Biotechnological slurry process
for the decontamination of
excavated polluted soils
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PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit Delft op gezag van de Rector Magnificus, prof. drs. P.A. Schenck, in het openbaar te verdedigen ten overstaan van een commissie aangewezen door het College van Dekanen op 18 juni 1991 te 14.00 uur.

door

René Henri Kleijntjens
Geboren te Leiden
Scheikundig Ingenieur
Dit proefschrift is goedgekeurd door

Prof.ir. K.Ch.A.M. Luyben Promotor
Prof.dr.ir. A.A.C.M. Beenackers Co-Promotor

Overige Commissieleden

Prof.dr. A.J.B. Zehnder
Prof.dr.ir. F.T.M. Nieuwstadt
Prof.dr.ir. J. de Graauw
Dr.ir. E.W.B. de Leer

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Motto

"The simpler the better, is a good rule for an engineer in search for theory. He is interested in truth of course; but it is the truth of the artist, which consists in emphasizing essentials and stimulating the imagination, not in accumulating detail without regard and relevance."

from "Mathematical models of Turbulence"
By B.E. Launder and D.B. Spalding,
Academic Press, New York (1972)

"The important thing is to get the pilot-plant built."

from "Pilot plants for medium-sized companies"
G.L.Constan in Chemical Engineering Progress, February 1984

Photograph at the cover shows the bottom part of the tapered 4 m³ DITS-reactor with the perspex dual (air-slurry) injector. To give an indication of the reactor size, a personal computer has been placed in the back ground and a screw-driver was stuck to the tapered front plate of the reactor.
VOORWOORD

Bij de start van een promotie-onderzoek weet je eigenlijk niet waar je aan begint. Je denkt "het zal wel gewoon werk zijn", en inderdaad, in het eerste onderzoeksjaar lijkt het op werken. Na verloop van tijd blijkt echter dat promoveren tijdelijk werk is. Dit aspect haalt een fikse streep door de plannen die met grenzeloos optimisme gemaakt waren. Het inzicht dat eveneens rijpt is, dat jij dan wel de promovendus mag zijn, maar dat zonder alle ondersteuning het onderzoek niet veel verder komt dan de mooie papieren plannen.

Een cruciaal punt wordt bereikt als de promovendus inziet dat technologie-onderzoek niet simpel is. Met name de ervaring dat grote opstellingen zo zwaar zijn dat ze door de fundamenten kunnen zakken, geeft een goed gevoel voor de complexiteit der dingen. In deze periode breken de slapeloze nachten aan. Immers, dan zijn er diverse afstudeerders in volle gang, werkplaatsen bouwen stalen ketels, computerprogramma’s draaien, terwijl voor experimenten duizenden kilo’s zand nodig zijn. De machine is in volle gang en bottlenecks worden één voor één omvergeworpen. Het belang van deze fase besef je eigenlijk pas als de volgende fase aanbreekt en de grote lijnen zichtbaar worden.

Hoe groot die lijnen zijn wordt meestal pijnlijk duidelijk in de eindfase van het onderzoek, de schrijverij. De promovendus realiseert zich het tijdelijke karakter van zijn werkzaamheden in deze periode volop. Elke tekst die moet worden geproduceerd neemt meer tijd in beslag dan hij denkt. Het opschrijven lijkt op het tekstuvel doden van een draak. Tijdens de eindfase wordt de moed er vooral ingehouden door te genieten van de mede promovendi die ook schrijven. Ook zij zien groene draken, hetgeen de saamhorigheid versterkt. Wanneer zelfs de schrijffase zijn eind nadert en de promovendus na 5 jaar de weg naar de drukker inzwaait, komen allerlei bespiegelende gedachten naar boven. Enkele van deze gedachten heeft u hierboven gelezen. Een afsluiting van het voorwoord kan alleen maar tot stand komen met het bedanken van iedereen.

Beginperiode
Het project "Biologische grondreiniging" BT3024, begon in de jaren '84-'85 als idee binnen de vakgroep Bioprocstechnologie. Mijn werkzaamheden (gestart in mei '86) bestonden in eerste instantie dan ook uit het vertalen van idee naar projektvoorstel. Dit ongebruikelijke begin van het onderzoek (meestal is er al een projektvoorstel), maakte dat ik mijn eigen baan op poten kon zetten. Ik wil mijn promotor Karel Luyben hierbij bedanken voor het vertrouwen dat hij mij gaf om het slurry-avontuur in gang te zetten. Al tijdens het schrijven van het voorstel werd er met zand-water-lucht reactor-en gewerkt in de kelder.

Uit deze tijd, er waren nog geen concrete fondsen binnen gehaald, stamt de benaming Low-Budget-Basement-project. In de LBB-project fase hielpen de afstudeerders Liesbeth Velthuisen en Michiel Bosse mee met het leggen van de basis voor het slurry proces. Praktische problemen kenmerkten de beginperiode. Het klankbord dat ik vond bij het toenmalige hoofd van de instrumentmakerij Gerard van der Toolen werd daarom één van de praktische fundamenten van het project. Het opzetten van de lastige analyses van grondmonsters werd in deze periode door Gert van de Steen uitgevoerd.

**Middenperiode**

In deze periode stond het zoeken naar theorie centraal. Hierbij speelden Karel Luyben en Rob van der Lans een grote rol. Samen met de afstudeerders Yvonne van Voorst, Mark de Groot, Astrid van de Graaf en Ad Smolders werd gezocht naar een "wetenschappelijke vorm" van het grond-slurry-gebeuren. Dat deze vorm soms wat ongebruikelijk was bleek uit het hoengelach dat aan de koffietafel opsteg als grondonderzoekers met vieze overalls binnen kwamen. Net zo als de vermaningen omtrent de "vieze rommel", werd dweilen een vast deel van het weekritme. De combinatie van aseptisch draaiende bioreactoren en het ontwikkelen van een slurry proces bleek niet altijd een even gelukkige.

In deze periode werd de 7 meter hoge pilot-plant gebouwd. Naast Gerard van de Toolen en de centrale TU werkplaats, werkte namens de instrumentmakerij van het Kluyverlab Peter Vetter met enorme inzet mee aan de constructie van de pilot-plant. Tevens was de ondersteuning van het bedrijfsburo, met name van George van Eybergen en Frans Bolman belangrijk. Namens de vakgroep
Bioprocesstechnologie speelden Sjaak Lisper, Bart Kerkdijk en Peter Kroon een rol bij het tot stand komen van de diverse opstellingen.

Het hebben van een goede analyse techniek bleek cruciaal voor het onderzoek. Een dankwoord voor de ondersteuning die ik vanuit de (toen nog zelfstandige) vakgroep Analytische Chemie kreeg, met name van Ed de Leer en Corry Erkelens, is daarom op zijn plaats. Andere universitaire ondersteuning werd gegeven door Lesley Robertson (vakgroep ME), professor Sef Heijnen (BPT), professor Nieuwstadt van het Laboratorium voor Aero- en Hydrodynamica en professor Beenackers van de Rijksuniversiteit Groningen. Uit de industrie was Piet de Boks van IWACO bij het project in deze fase betrokken.

Eindfase

Misschien werd de eindfase van mijn promotiewerk wel ingeluid door het patent dat door Luyben en Kleijntjens werd aangevraagd voor de DITS-reactor (Dual Injected Turbulent Separation-reactor). Deze spin-off van het onderzoek gaf het project de rust mee die noodzakelijk was om op de ingeslagen weg door te gaan. Dit laatste is iets dat binnen de turbulente milieuwereld niet onderschat moet worden. Het waren de afstudeerders Hilda van de Laar en René Langevoort die het project mee over de eindstreep trokken.

Van grote waarde was de inspanning die Peter de Groot, vaak met de klei tussen zijn oren, leverde bij het uitvoeren van de praktijkproeven met de Amsterdamse klei. Afroondende kostenberekeningen werden in deze periode door Ted Meeder gestart. Mijn opvolger Marcel Geerdink nam in deze fase hard meer en meer het onderzoek over. Hoe dynamisch promoveren kan zijn bleek uit de nieuwe bijdragen die Chris Hellinga in het project bracht. Met zijn computerprogramma MAJROBAL bleek het microbiologische onderzoeksdeel tot een goed einde te brengen.

Voor wat betreft de schrijffase, gaat mijn dank bijzonder uit naar Karel Luyben, Rob van der Lans, Lesley Robertson, Marcel Geerdink, Chris Hellinga en Henk Noorman. Zij hielden allen mee "de taal de baas te worden".

Omdat het, behalve promoveren, ook de bedoeling was om een praktisch haalbaar grondreinigingsproces te ontwikkelen, ben ik zeer verheugd dat C.
Broërkens, J. Verheul en J. Bos van het ingenieursbureau DHV genoemd kunnen worden in de laatste fase. Ook de samenwerking met de gemeente Amsterdam en met name Linda Maasdijk was voor het uitvoeren van de praktijkproeven kardinaal. In de toekomst bestaan er mogelijkheden om het TU-Delft slurry-proces met DHV en de gemeente Amsterdam tot een pilot installatie te brengen.

Extra vermeldenswaard is de samenwerking met Mieke Boon, Jacques Potters en Karel Luyben op het gebied van schaalvergrotingsonderwijs. Door te zoeken naar nieuwe concepten zijn we verder gekomen op het gebied van onderzoek/onderwijs van dit lastige vak.

Zonder verder toelichten wil ik verder de volgende mensen bedanken:

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Omdat onderzoek zonder inspiratie niet kan slagen, wil ik tot slot mijn Brabantse inspiratiebron alle lof en dank toe zwaaien.
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SUMMARY

Since the "Club of Rome" report (1972), environmental problems have gained increasing interest from industry, politics, science and other groups in society. Regarding the pollution of the three earthly compartments, air, water and land, the environmental threat originating from contaminated land has been recognized latest (since about 1980). Nowadays, most of the polluted sites in the industrialized countries have been mapped and the consequences of soil contamination (and contaminated groundwater) have been recognized as a serious problem. The growth in number of remediation technologies must be seen within this framework.

One of the first operational techniques for soil decontamination was thermal treatment. However, a major disadvantage of thermal treatment is the poor quality of the endproduct. To decontaminate the soil without destroying its fertility, biological techniques are considered attractive remediation alternatives.

In this thesis the development of a new biotechnological slurry process for the decontamination of excavated polluted soils is described. The slurry process is especially designed to decontaminate soils polluted with organic compounds (e.g. oil). Crucial in the slurry process is the use of three phase (soil-water-air) suspension bioreactors. In these reactors, an aerated suspension of polluted soil particles is sustained under conditions (pH ≈ 7, temperature = 30 °C) which favour the microbial conversion of the pollutant. In the bioreactors, natural microbial communities are used, originating from polluted harbour sludge.

In order to design a large scale biotechnological slurry process, the proper design parameters were identified. Kinetic experiments, carried out in a continuously operated mini-plant (40 l), showed that for a soil residence time of 6 days, microbial decontamination down to the Dutch b-level (1 g/kg soil) was reached. To achieve decontamination down to the a-level (50 mg oil/kg soil) a residence time of 28 days was estimated from an extrapolation of desorption data.

For the reactor development the research was directed at the hydrodynamical description of three phase (s-l-g) tapered slurry reactors, with a
polydisperse soil (particle size from 1 \( \mu \)m to 4 mm) as solid phase. For efficient soil processing, the coarse, relatively low polluted, soil fraction should be treated and separated at the beginning of the process. For this purpose, a new, gas-liquid agitated suspension reactor was developed, the Dual Injected Turbulent Separation (DITS) reactor. In this reactor, the coarse soil fraction (particle size larger than 700 \( \mu \)m) is fluidized at the bottom, while the polluted fines are kept in suspension in the bulk. Subsequently the fluidized fraction is withdrawn. Once the coarse fraction has been removed, the fines are further processed in a cascade of air agitated slurry reactors. Final step is the remixing of both fractions in a dewatering section.

As major design parameters, the solid hold-up and the power input were identified. Experiments carried out at four scales (0.08, 0.24, 0.4 and 4.0 m\(^3\)) showed that 40 wt% soil could be sustained in suspension efficiently, with moderate power input (100-300 W/m\(^3\)). To estimate the specific power input for the bottom and the bulk compartment of a large scale, 40 m\(^3\), DITS-reactor, experiments and a mathematical (turbulent) suspension model were used. Calculations resulted in a specific power input of \( \approx 50 \) W/m\(^3\) for the bulk and \( \approx 1000 \) W/m\(^3\) for the relatively small bottom compartment. For the cascade reactors a power input equal to the bulk of the DITS-reactor was considered appropriate. From the power inputs the required gas and slurry injection flows for all reactors could be estimated.

Based on the experimental results achieved, a step-wise scale-up strategy was considered appropriate for the future development of the slurry process. First, a 16 m\(^3\) pilot plant and a 160 m\(^3\) demonstration plant should be designed and tested. Based on the results achieved with these two plants, the construction of a 2680 m\(^3\) full scale plant can be considered. Cost estimations showed that the economics of the slurry process largely depend on the outlet concentration of the pollutant. Accepting the b-level, the cost estimation for the full scale plant resulted in about 50 Dfl/ton of soil treated. If the a-level has to be reached, these costs are expected to be about 200 Dfl/ton. Contaminated soils are considered as untreatable at this moment in the Netherlands, if the decontamination costs come above 250 Dfl/ton.
SAMENVATTING

Sinds het rapport van de "Club van Rome" (1972), wordt onderkend dat de vervuiling van lucht en water ernstige milieuproblemen met zich mee brengt. Vervuiling van de bodem daarentegen wordt pas sinds het begin van de jaren tachtig als probleem gekenmerkt (zo vond in Nederland rond 1980 de "Lekkerkerk-affaire" plaats). Tien jaar bodeminventarisatie bracht echter het merendeel van de verontreinigde locaties in kaart en maakte duidelijk dat het probleem omvangrijk is. Parallel hieraan ontstond de vraag naar saneringstechnieken voor vervuilde grond.

Een van de eerste, en veelvuldig gebruikte, bodemsaneringstechnieken was thermische reiniging. Aan deze techniek kleeft echter het bezwaar dat de behandelde grond een slechte eindkwaliteit heeft. Als mogelijk alternatief worden biotechnologische technieken ontwikkeld.

In dit proefschrift wordt de ontwikkeling van een nieuw biotechnologisch proces voor de reiniging van afgegraven, vervulde, grond beschreven. Het betreft een slurry proces, in principe ontwikkeld voor de reiniging van grond die met organische componenten (bv. olie) vervuild is. Centraal in het proces staan drie fasen (grond-water-lucht) slurry reactoren. In deze beluchte bioreactoren worden gunstige afbraakomstandigheden gecreëerd door de vervuilde gronddeeltjes te suspenderen. Een microbiële populatie (afkomstig uit vervuilde havenslib) is vervolgens in staat om, bij een optimale temperatuur en zuurgraad (30 °C, pH=7) de vervuilende componenten af te breken.

Cruciaal bij de procesontwikkeling, is het vaststellen van ontwerpparameters. Als eerste parameter werd de grondverblijftijd in het systeem bepaald. Op basis van kinetiek-experimenten, uitgevoerd in een 40 liter, continu bedreven mini-plant, werd bepaald dat een verblijftijd van 6 dagen voldoende is om grond (olievervuiling) te reinigen tot de b-norm (1 g/kg grond). Uit extrapolaties is afgeleid dat de a-norm (50 mg/kg grond) bij een verblijftijd van 28 dagen bereikt zou worden.

Reactorparameters werden bepaald op basis van hydrodynamisch onderzoek aan taps toelopende suspensie reactoren. Als vaste fasen werden zowel mono- dispers zand (260 μm) als polydisperse grond gebruikt (deeltjesgrootte variërend van 1 μm tot 4 mm) gebruikt. Bepaald werd dat een efficiënt slurry
proces alleen tot stand kan komen wanneer de grove, relatief weinig vervuilde, grondfraktie aan het begin van het proces verwijderd wordt. Om dit te berekenen is een nieuwe, gas-vloeistof geroerde tapse suspensie reactor ontwikkeld, de "Dual Injected Turbulent Separation" reactor oftewel DITS-reactor. De grove fraktie, met een deeltjesgrootte van boven de 700 µm wordt in het onderste gedeelte van deze reactor gefluidiseerd, terwijl de fijne fraktie (deze bevat het merendeel van de vervuiling) in de bulk wordt gesuspendeerd. Vanuit het gefluidiseerde deel wordt de grove fraktie afgescheiden. Nadat deze scheiding is uitgevoerd, worden de fijne gronddeeltjes verder behandeld in een cascade van lucht geroerde slurry reactoren. Laatste processtap is de opmenging van de behandelde fijne en grove frakties.

Als belangrijkste reactorparameters werden de slurrydichtheid en de vermogenstoever onderzocht. Aangetoond werd dat een grondslurry met een deeltjesdichtheid van 40 gew.%, op twee verschillende reactorschalen (0.40 en 4.0 m³), zonder problemen kon worden gesuspendeerd/gefluidiseerd in de DITS-reactor. Teneinde de vermogenstoever voor een 40 m³ DITS-reactor (grote schaal) te bepalen, werden de experimentele resultaten met behulp van een mathematisch (turbulentie) model geëxtrapoleerd. Berekend werd dat de specifieke vermogenstoever voor het onderste compartiment (grove fraktie) 1000 W/m³ en voor de gesuspendeerde fijne fraktie in het bulk compartiment 50 W/m³ bedraagt. Voor de luchtgeroerde cascade reactoren is eveneens 50 W/m³ nodig. Op basis van de specifieke vermogenstoever kunnen voor alle reactoren de benodigde lucht- en vloeistofinjectiestromen berekend worden.

In het proefschrift werd aangetoond dat een stapsgewijze schaalvergroting de meest geschikte manier is om tot een slurry proces op grote schaal te komen. Op basis van de ontwerpparameters worden achtereenvolgens een pilotinstallatie (16 m³), een demonstratie-installatie (160 m³) en een 2680 m³ full scale proces in beschouwing genomen. Een duidelijk verband bleek te bestaan tussen de kostprijs per ton behandelde grond en de vereiste reinigingsgraad. Wordt de b-norm geaccepteerd, dan kan de 2680 m³ installatie worden bedreven voor een kostprijs van 50 gulden per ton. Wanneer echter de a-norm bereikt moet worden, dan stijgen de kosten tot 200 gulden per ton. Op dit moment wordt in Nederland grond als niet reinigbaar beschouwd als de kostprijs boven de 250 gulden per ton ligt.
Introduction

Ever since the Club of Rome report "Limits to growth" (Meadows et al, 1972), environmental problems have gained increasing interest from industries, politicians, scientists and other groups in society. As one of the consequences of this increasing interest, a wide range of remedial action technologies has been developed in the last decades. The aim of remediation processes is the removal of polluting compounds. The material to be treated can be a gas (e.g. off gas treatment), a fluid (e.g. waste water treatment) or a solid (e.g. thermal treatment of domestic waste). In figure 1 a general diagram for a conversion process is given. The diagram can also be applied for remediation processes.

![Diagram of chemical process](image)

**Figure 1** Structure of a chemical process.

In the development of new remediation techniques bottlenecks can be expected in each of the three sections. To reduce these problems an integrated process design is of crucial importance. For instance, integration of the reactor section and the after-treatment section could facilitate downstream processing. The multi-disciplinary character of process design, especially comes to the fore in scaling-up. In scale-up, methods are used, or developed, to transform information from small scale experiments into (full scale) design parameters.

As depicted in figure 2, there are several ways to design large scale process operations. An elegant design method is found in the analogy approach. Crucial here is the assumption that the uncharacterized operation resembles in its major features a well defined operation. The advantage of this method is found in the literature and the theoretical models available for the analogue operation. Theoretically, a large scale operation can be designed. It should however be noted that the assumptions supporting the analogy, may have been incorrect.
A second design strategy can be called the engineering approach. Based on the information available and the aims of the research, either the empirical path or the more pragmatic path of first (engineering) principles is followed. According to the first path, structured experiments are to be carried out in combination with the creation of a mechanistic model. The model either has its roots in theoretical descriptions of rate limiting phenomena, or in empiric correlations. Design and scale-up is based on the extrapolation of trends observed in model parameters and experimental data (Berty, 1979; Rose, 1981). More pragmatic is the use of available engineering principles and empiric correlations, In this approach, a clear relationship with theory is not always present, rules of thumb and industrial experience play a key role (Jordan, 1968; Johnstone and Thring, 1957).

As a third method the trial and error approach is given. Without the burden of theoretical considerations, the full scale design is carried out straight from the concept. Since it is unlikely that the full scale design will be correct, failures become manifest during the practical operation at the full scale. Full scale trial and error experience thus is necessary to complete the design.

*Figure 2* Design philosophies for new process operations.
In the development of new remedial action technologies, the choice of an appropriate design and scale-up pathway is important due to the high demands which have to be fulfilled. A required pollutant conversion of 99% or higher is not unusual (Dennekamp and Robberse, 1990). Not only from the point of view of technology, remedial action suffers from severe restrictions. Also with regard to cost aspects, the position of cleaning techniques is not favourable. The best technical means often have to be achieved with the smallest financial means. These restrictions certainly are a hindrance in the development of new soil reclamation techniques. Since the necessity of efficient and cheap soil decontamination will increase in the future, the need of a structured, scientific, approach in the development of new processes becomes more important.

Soil decontamination

Soil pollution only recently has been recognized as a serious threat to the environment (in the Netherlands since the Lekkerkerk affair in 1980). Therefore remedial action technologies for the treatment of excavated polluted soils are emerging mainly this last decade. To be able to judge the merits of a soil decontamination process, five aspects have to be considered

* range of pollutants
* soil types
* level of decontamination
* end product quality
* costs

Two well known soil decontamination processes, thermal treatment and landfarming, will be briefly discussed with regard to these five aspects.

In thermal soil treatment excavated polluted soil is exposed to high temperatures (up to 1100 °C) in a (rotary) kiln in order to evaporate the pollution from the matrix (Reintjes, 1990). The vaporized pollutants subsequently are burned in an off-gas treatment section. In thermal installations, organic xenobiotics are treated with high decontamination levels. Different soil types have been treated in practice, for prices ranging from 150-250 DFL/ton. Major disadvantage of thermal processing is
the bad end product quality, and the difficulty to treat wet clay and silty soils.

The most widely applied biological treatment method is landfarming (Maag, 1990). In this "low-tech" treatment method, excavated polluted soils are evenly spread out in a so-called landfarm. By means of farming techniques such as fertilizing and ploughing the microbial activity in the soil is stimulated. In landfarming the organic pollutant is aerobically mineralized to carbon dioxide, water and biomass. Different soil types have been treated in landfarming although clay and silty soils are difficult to handle due to the large agglomerates present. The quality of the endproduct is a major advantage of this method. Also the costs, mostly around 100 DFl/ton are rather low. Major disadvantages of landfarming are the long time needed for treatment (up to 2 years), the low decontamination levels and the uncontrolled conditions under which the treatment takes place (outdoor conditions).

In order to benefit better from microbial soil processing (endproduct quality and mineralization of the pollutant), the disadvantages of a slow process have to be circumvented. For this purpose, it might be sensible to carry out the aerobic microbial conversion (of the organic pollutants) under controlled conditions in a bioreactor. It is for this reason that a soil slurry process has been developed.

**Biotechnological slurry process**

The core of the slurry process described in this thesis is formed by the bioreactors. To circumvent a limited availability of the pollutants due to the presence of particle agglomerates, and to improve mass transfer (oxygen), a suspension bioreactor was considered to be the most appropriate reactor type. In this three phase (solid-liquid-gas) system, soil particles are suspended in process water in the presence of air. In the reactor optimal process conditions for the microbial breakdown can be obtained (temperature, pH and mineral medium).

The aim of this research project was to develop a biotechnological slurry process for the decontamination of excavated polluted soils. With respect to the above mentioned process aspects, the following aims for the process design were set.
* treatment of excavated soils, polluted with oil products
* a large variety of soil types (including clay/silt) as feed
* optimization of the conversion by optimal bioreactor conditions
* healthy soil as end product
* minimal costs

Organization of the thesis

In line with the general discussion concerning remediation technologies, the bottleneck in the process design was the design of the slurry reactor. The main part of the research which is described in this thesis, therefore focuses on the design of the bioreactor. Due to the short history of soil processing, only recent literature is available on this subject. Scientific meetings on the subject take place since the mid-eighties (Sanning et al., 1990). The development of the process, and more specific of the bioreactor, thus falls into the profile of uncharacterized operations. The thesis contents will be discussed within the framework provided by figure 2. For this purpose, the relevant part of figure 2 is redrawn in figure 3.

Figure 3  Organization of the thesis.
The actual process design is described in the last chapter of the thesis (chapter 7), in this chapter also the economic feasibility is discussed. To be able to derive a realistic full scale design, research focused on both the microbial decontamination kinetics, and the development of a reactor having the ability to process soil slurries. As shown in figure 3, both a theoretical and an engineering approach were used.

Placed at the right side of figure 3 is the study focussing on the microbial kinetics of the decontamination process, described in chapter 1. This research was carried out in a continuously operated mini-plant with a total volume of 40 liter. Using two types of soil and a range of pollutant concentrations (2.5 - 15 g/kg), more than one year of continuous processing was carried out. Since data on kinetics are scarce in literature, experimentation dominated this part of the research (engineering approach). In order to evaluate results from the mini-plant experiments, statistical data analysis was performed (chapter 2). The theory for the second chapter originates from developments on monitoring and control. Aim of the analysis was to find on-line monitor parameters for the slurry process.

Due to the unfamiliarity of soil as solid phase in a multiphase slurry system, the reactor development was performed with a model system. For this purpose, the polydisperse solid phase (soil) was replaced by monodisperse quartz sand. In chapters 3 and 4, hydrodynamic aspects of the three phase (solid-liquid-gas) model reactor are discussed. Both chapters are located in between the blocks theory and model experiments, positioned at the centre of figure 3.

In chapters 5 and 6 the hydrodynamic results are integrated with the practical soil experiments. Aim of this integration is to develop an optimal reactor and to derive an "engineered model" of soil suspension phenomena. The chapters 5 and 6 are located to the right side of figure 3 since practical design arguments dominate this part of the research. Experimental scales for the design of the soil slurry reactor, varied from 0.080 m³ to 4.0 m³, with four different slurry densities (up to 40 wt%). In the experiments both gas agitated (model system) and gas-slurry agitated reactors were used.

This introduction was positioned as chapter 0 at the top of figure 3. This emphasizes that research methodology is crucial for the development of new remediation technologies.
LITERATURE


Meadows D.L, "Limits to growth", report of the Club of Rome, (1972)


MICROBIAL DECONTAMINATION
Microbial decontamination of polluted soils in a slurry process

Part 1 Mini-plant operation

Kluyver Laboratory for Biotechnology, Delft University of Technology

ABSTRACT

To examine the feasibility of a biotechnological slurry process for the decontamination of excavated polluted soils, continuous soil processing was carried out in a mini-plant. The mini-plant consists of a cascade of two (20 l) aerated tapered suspension bioreactors. Independent of the pollutant's concentration (ranging from 2.5-16.8 g oil/kg soil) an overall reduction of about 70% was reached for a soil residence time of 200 hours. Based on this experience, a 40 year old oil polluted clay soil was decontaminated below the dutch B-level (1 g/kg). In the second bioreactor, low conversion rates were observed despite the presence of significant pollutant concentrations. This result suggests that the rate-limiting step in the conversion of the pollutant is the desorption of the pollutant from the soil particles.

KEYWORDS Soil decontamination, bioreactors, mini-plant, slurry process, oil pollution.
INTRODUCTION

Historical and modern industrial and military activities have resulted in large amounts of polluted soil in many different countries. The environmental and public health dangers of these polluted soils are not only limited to the sites themselves, which have to be closed down or lose their multi-functional use, but often extend to the contamination of groundwater over much larger areas. Therefore, the development of environmentally and economically feasible methods for the treatment of contaminated soils is becoming ever more important in most industrial countries. This is certainly the case in the Netherlands. In a recent survey, it was estimated that there are over ten thousand polluted sites in the Netherlands, a fair percentage of which are contaminated with oil products (Brinkman et al., 1989). An extensive governmental programme, aimed at the development of new soil reclamation techniques, has therefore been started (Eijsackers, 1989; Brinkman et al., 1989).

In soil treatment, a wide range of cleaning techniques is used. Distinction must be made between in-situ and ex-situ treatments. In urban areas, it is often not possible to excavate the polluted site, in situ treatment is then most appropriate. Biorestitution, which aims at the stimulation of microbial activity at the site by the injection of organisms, nutrients and oxygen, is an example of an in-situ method (Staps, 1990). For ex-situ techniques, the polluted soil is excavated and decontaminated in an installation either at the site, or transported and treated elsewhere. Excavated soils are often treated by means of physico-chemical or thermal reclamation technologies (Tuin, 1989; Assink, 1988). A disadvantage associated with these methods is the poor condition of the soil after the treatment. It is generally sterile and much of its organic content has been destroyed. In addition, the pollutant is not always mineralized. The latter is the case in soil extraction with organic solvents, a form of treatment that generates a waste stream to be cleaned.

To overcome these problems, biological decontamination techniques may be emerging as alternative treatments for excavated polluted soils. One of these biological methods is landfarming (Soczo and Staps, 1988; Maag, 1990).
In landfarming, excavated polluted soil is spread out in a special depot as a layer, 0.5-1.0 m deep. To this layer, specific bacteria are added in combination with nutrients, oxygen transport is enhanced by means of ploughing or forced aeration. In this method, organic pollutants are mineralized under relatively mild conditions (pH=6-8, T=10-50 °C) to harmless products such as biomass, carbon dioxide and water with hardly any toxic compounds being formed.

There are some disadvantages associated with landfarming. Most obvious is the requirement for large (costly) areas of land. Secondly, the conversion rates are low to very low, resulting in treatment periods which may exceed 2 years (Bonnier, 1980). High residual concentrations have been found after the treatment. In addition, biological systems in the open can be rather sensitive to changes in environmental conditions (summer-winter). To take more advantage of the benefits of biological soil decontamination, it is therefore desirable to process the soil in a better controlled environment. For this purpose, a biotechnological soil slurry process has been developed.

CHARACTERISTICS OF BIOTECHNOLOGICAL SOIL PROCESSING

In biotechnological soil processing, one can distinguish four different factors that play key roles

- soil type
- pollutant
- microbial community
- environmental conditions

Each of these four factors will be discussed briefly as an introduction to the slurry process.

Soil type

Soil is a complex multi-phase system in which several physical, chemical and microbiological processes occur simultaneously (Brady, 1984). It is composed of inorganic materials (sand, silt, clay), organic material (e.g. partially
degraded plant remains and colloidal humus), water and air. Soil can be classified in terms of its organic content and/or particle size distribution. Several standards for soil classification, based on particle size distribution, are shown in figure 1. Sand particles are mainly composed of \( \text{SiO}_2 \) (quartz), have a specific surface area in between 0.1 and 1 \( \text{m}^2/\text{g} \) and are relatively inert with respect to sorption processes. This contrasts with clay, which has a specific surface area of 10-1000 \( \text{m}^2/\text{g} \) and adsorbs water, nutrients and organic pollutants. The colloidal organic soil (humus) fraction also has the capacity to adsorb or chemically bind pollutants. Depending on the origin, the organic content in soil can vary from almost zero (sand) up to almost 100% (peat) (Brady, 1984).

<table>
<thead>
<tr>
<th>British Standards</th>
<th>Clay</th>
<th>Fine</th>
<th>Medium</th>
<th>Coarse</th>
<th>Fine</th>
<th>Medium</th>
<th>Coarse</th>
<th>Gravel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Silt</td>
<td></td>
<td></td>
<td></td>
<td>Sand</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>International Society of Soil Science</td>
<td>Clay</td>
<td>Silt</td>
<td></td>
<td></td>
<td>Sand</td>
<td></td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
<td>Fine</td>
<td></td>
<td></td>
<td>Coarse</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>0.002</td>
<td>0.02</td>
<td>0.06</td>
<td>0.2</td>
<td>0.6</td>
<td>2.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.002</td>
<td>0.02</td>
<td>0.2</td>
<td>2.0</td>
<td>1.0</td>
<td>2.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>United States Department of Agriculture</td>
<td>Clay</td>
<td>Silt</td>
<td>Very fine</td>
<td>Fine</td>
<td>Med. Coarse</td>
<td>Very Coarse</td>
<td>Gravel</td>
<td></td>
</tr>
<tr>
<td></td>
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<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td></td>
<td>0.005</td>
<td>0.5</td>
<td>0.25</td>
<td>0.25</td>
<td>0.5</td>
<td>1.0</td>
<td>2.0</td>
<td></td>
</tr>
</tbody>
</table>

Particle diameter (mm, log scale)

**Figure 1** Classification of soil particles based on particle size (Brady, 1984).

At each location soil characteristics are different, therefore each polluted site has its own identity in terms of particle size distribution, texture, organic content, permeability etc. Due to these variations, the most appropriate remedial techniques will differ from site to site. It is generally accepted that sandy soils can be decontaminated without difficulty by existing, non-biological methods. This biotechnological slurry process
chapter 1

therefore aims at the treatment of soils containing a broad range of particles, and especially at soils containing clay.

Pollutant

Biotechnological soil reclamation is mainly used for the decontamination of soils polluted with mineral oils. These oil pollutants can contain a mixture of several hundreds of different hydrocarbons (e.g., alkanes, aromates, asphaltenes) (Colwall and Walker, 1977; Teschner, 1985; Altgelt and Gouw, 1979; Speight, 1982). The susceptibility of oil components to microbial attack has been ranked as: n-alkanes > branched alkanes > low molecular weight aromatics > cyclic alkanes (Perry, 1984).

When soil is contaminated, the pollutant interacts with the solid matrix, thereby seriously decreasing its availability for microbial breakdown. Previous experiments on soil-pollutant interactions have shown the irreversibility of the adsorption of polycyclic aromatics (Knezovich et al., 1987) and pesticides (Di Toro and Horzempa, 1982). It should be noted that because of the different characteristics of the particles, adsorption to the clay and humic fractions is more pronounced than to the coarse fractions (Assink, 1988).

At higher concentrations of the more water soluble toxic oil components, such as low-molecular alkanes, the microbial community may be disrupted (Ratledge, 1978). It should, however, be noted that due to adsorption, the chance of lethal toxic effects in soil systems is lower than in systems without a solid phase.

In an earlier study on the slurry process, diesel was selected as model pollutant, since its composition is considered to be representative of an extensive range of oil products. Diesel contains over 300 detectable components, varying from linear and branched alkanes to mono- and polyaromatic hydrocarbons. The presence of a series of homologous linear alkanes varying from \( C_{10} \) to \( C_{40} \) is characteristic of diesel. To test the practical feasibility of the process, a 40 year old oil pollution containing both light and heavy oil fractions has been treated as well as a model system.
Microbial community

A microbial community containing a diversity of organisms will be necessary for the rapid and complete degradation of oil. In the literature, many (mostly aerobic) microorganisms with the appropriate metabolic potentials for the breakdown of oil components have been described (Beerstecher, 1954; Davis, 1967; Ratledge, 1978; Trudgill, 1978; Hopper, 1978; Atlas, 1981). Bacteria, yeasts and fungi are all capable of degrading hydrocarbons. The most important genera of hydrocarbon utilizers so far described are Pseudomonas, Arthrobacter, Micrococcus, Nocardia, Candida and Rhodotorula. Pure cultures are able to degrade an extensive range of single and mixed hydrocarbons, including very persistent polyaromatics (Weissenfels et al., 1990; Shialabhai et al., 1985). Nevertheless, as in other waste treatment systems, mixed cultures are much more stable and effective (Ratledge, 1978; Trudgill, 1978; Hopper, 1978).

Hydrocarbon degradation is not, however, the only reaction taking place during microbial soil reclamation (Alexander, 1977). In an aerated slurry, biological processes such as nitrification, humification and sulphur oxidation may run simultaneously with the hydrocarbon conversion. In addition, protozoa feed on the bacteria, thereby reducing their number. This range of activities makes a complete description of the decontamination process difficult.

Environmental conditions

The presence of microorganisms with the appropriate metabolic potential is no guarantee for a successful biological reclamation process. The bacteria have specific environmental requirements which, to a large extent, have to be fulfilled in order to establish microbial breakdown. A major requirement is, besides oxygen and nutrient supply, the accessibility of the pollutant to microbial attack (Rijnaarts et al., 1990). In soil, bioavailability is impeded by

1) the presence of agglomerates enclosing the pollutant
2) the adsorption of the pollutant to individual particles
Ad 1 To improve the conditions under which the microbial decontamination takes place, the use of bioreactors has become popular during the last few years (Sozcz and Staps, 1988). Within this framework, aerated three phase (solid-liquid-gas) slurry bioreactors are considered (Kleijnjens et al., 1987; Kleijnjens et al., 1989). Because in slurries individual particles are suspended and agglomerates desintegrate (Rijnaarts et al., 1990), availability problems due to the presence of agglomerates mostly are avoided.

Ad 2 Concerning the desorption from individual particles, surfactants can be used to stimulate the availability. Oberbremer and Müller-Hurtig (1989) described the step wise aerobic degradation of a model oil mixture and the formation of biosurfactants by a natural soil community in a stirred batch fermentor. The most soluble hydrocarbons were first degraded. The subsequent breakdown of the other compounds only began when the interfacial tension was reduced by the production of surfactants at the beginning of the second degradation phase.

Also, the addition of microbial surfactants to the soil slurry resulted in a faster and more complete reduction of the hydrocarbon level (Oberbremer et al., 1990). In order to improve the availability of the contaminants, the addition of artificial surfactants might be considered in biological soil processing. In addition, the temperature, pH and nutrient concentrations can be set at the most appropriate levels and adequately controlled in a bioreactor.

**SLURRY PROCESS**

A continuous slurry process was designed for the biological treatment of excavated polluted soils (Kleijnjens et al., 1990). First stage in the process is a pretreatment section, in which soil is crushed and sieved. From this section, polydisperse polluted soil ($1 < d_p < 4000 \mu m$) is transported to the first reactor where it is mixed with process water and nutrients. The tapered reactors shown in figure 2 were selected as the most appropriate for soil slurry processing (Kleijnjens et al., 1991a).
In the first reactor, the coarse soil fraction \( (d_{p} > 700 \, \mu m) \) is fluidized in the bottom zone by means of an upward injected slurry flow (originating from the recirculation unit), while fines are kept in turbulent suspension in the gas agitated bulk compartment.

From the bottom of the first reactor, the coarse fraction is withdrawn and passed directly to the dewatering section. As a result of this mode of operation, the first reactor simultaneously acts as reactor and separator, therefore denoted as Dual Injected Turbulent Separation (DITS) reactor (Luyben and Kleijntjens, 1988). The fine material, containing most of the adsorbed pollutants (Assink, 1988), is given a further treatment in the cascade of air agitated bioreactors. In the dewatering section, the effluent flow of the last bioreactor is spread on top of the coarse fraction which is used as filter aid. In order to reduce the amount of waste water, percolation water is recycled to the DITS-reactor.
MATERIALS AND METHODS

Mini-plant

In order to investigate the feasibility of the slurry process and to determine design parameters, experiments were carried out in a mini-plant. Mini-plant experiments were considered beneficial since, "performance data derived from correctly designed and operated small scale plants are more accurate than data scaled-up directly from the laboratory" (Johnstone and Thring, 1957). In a mini-plant process conditions can be chosen close to the expected full scale conditions.

In line with the flowsheet for the full scale, the mini-plant was designed as a set of tanks in series. In the mini-plant, four stages are to be distinguished (figure 3)

1) Soil supply elevator. The elevator contained dried, polluted soil which has been crushed and sieved to give a particle size distribution ranging from clay ($d_p < 1 \mu$m) up to coarse sand ($d_p = 1.5 \text{ mm}$).

2) First slurry bioreactor (DITS-reactor) with air and slurry injection. In this two compartment system, fluidized coarse material is withdrawn from the bottom, while the suspended fine material is removed from the top. Two different effluent flow rates are thus obtained, allowing a short residence time for the relatively clean coarse fraction and a longer residence time for the fines in the bulk compartment.

3) Second bioreactor (air agitated). The effluent from the top of the first reactor is used as feed for this second reactor. After a residence time about equal to that in the bulk compartment of the first stage, the effluent from the second reactor is transported to the dewatering section.

4) In the dewatering section, the slurry from the second bioreactor is spread over a filter bed which contains the coarse material from the
first reactor. In line with the flowsheet, percolation water is recirculated to the DITS-reactor.

![Diagram](image)

**Figure 3** Schematic representation of the mini-plant.

Considering a flow of several kilograms of soil per day appropriate, the total volume of the mini-plant reactors was equal to 40 liter (2 * 20 l). The maximum size of the soil particles which could be treated in the mini-plant was determined experimentally to be 1.5 mm. Experiments in the mini-plant focussed on kinetic aspect of the process design. Most crucial research parameter within this framework was the residence time of the solid phase. In addition to the kinetics, the slurry handling was investigated by means of monitoring the solid hold-up and particle size distributions at several points in the system.

**Operating conditions in the mini-plant**

The operating conditions used during processing in the mini-plant are summarized in table 1. The volume of the fluidized bed in reactor 1 is about 10 % of the working volume (20 l) of the first reactor. Two different residence times for the suspended soil fraction were investigated; 50 and 100 hours per reactor (100 and 200 hours total). In both cases, the residence time of the coarse fraction was about 24 hours. The residence time
ratios (bulk : bed) thus were 4:1 and 8:1. Based on these times, the solids
hold-up in the bulk of both reactors was $\epsilon_s = 0.05$, and $\epsilon_s = 0.23$ for the
bottom compartment in the DITS-reactor.

Air flows to the mini-plant were 10 l/min for the first reactor and 7.5
l/min for the second reactor (0.5 and 0.375 VVM (volume/(volume*minute)),
respectively). These air flows were chosen such that

* the fine soil fraction was kept in turbulent suspension
* the air flow was low enough to monitor (on-line) a significant
difference between the incoming and outgoing oxygen concentration.

The slurry injection flow (to fluidize the coarse fraction) in the first
reactor was set at 2 l/min (0.1 VVM). At higher flow rates, obstructions
caused practical problems in the recirculation loop, and at lower rates the
coarse fraction could not be fluidized.

The nutrients (medium) were added to the first reactor in a concentrated
flow (see figure 3), containing per liter tap water: 20 grams of NPP
fertilizer, corresponding to a concentration of 2.4 g/l nitrogen (75 % NH$_4^+$
and 25 % NO$_3^-$), 2.0 g/l phosphorus (P$_{2}$O$_5$) and 3.6 g/l potassium (K$_2$O). Also
0.5 gram MgSO$_4$ and 0.25 gram FeCl$_3$ were added. This concentrated nutrient
flow is diluted with the water flow in a ratio of about 1 : 7. The true
influent liquid concentrations of N thus are about 0.01 mol/l for NH$_4^+$ and
0.001 mol/l for NO$_3^-$. In the slurry reactor, this composition corresponds to
a molar C:N ratio of about 10:1.
Table 1 Operating conditions in the mini-plant.

<table>
<thead>
<tr>
<th>Volume: [l]</th>
<th>reactor 1</th>
<th>reactor 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>total effective</td>
<td>20.0</td>
<td>20.0</td>
</tr>
<tr>
<td>bulk</td>
<td>18</td>
<td>20.0</td>
</tr>
<tr>
<td>fluidized bed</td>
<td>2</td>
<td>-</td>
</tr>
</tbody>
</table>

Agitation flows: [l/min]
- air injection                  | 10        | 7.5       |
- liq injection                   | 2         | -         |

Soil residence time: [h]
- bulk                            | 50/100    | 50/100    |
- fluidized bed                   | 24        | -         |

Soil feed (from elev) [g/h]
-                                       | 76/38     | -         |

Flow rates: [ml/h] in
- medium supply                     | 48/24     | 24/12     |
- tap water + recycle                | 396/198   | -         |
- slurry                            | -         | 360/180   |
- fluidized bed                     | 360/180   | 384/192   |

The contribution of soil organics is not taken into account. Trace elements were assumed to be sufficiently available in the soil and the tap water. The simple NPP medium was chosen in order to keep the conditions as close as possible to real situation. On the basis of an estimated complete conversion of hydrocarbons, the nutrients supply rate was 0.048 l/hr for a residence time of 50 hours per reactor, and 0.024 l/hr for a residence time of 100 hours per reactor (Table 1). In the second reactor, the medium supply flow rate was 50% of that in the first reactor.

The temperature in both reactors was kept at 30 °C. Because protons are released during the degradation of the pollutant as well as in humification and nitrification, the pH was controlled at 7.0 using a 2 N NaOH solution.

Preparation of the (model) soil pollution
For the model experiments (runs 1 to 5), a clean soil with a polydisperse particle size distribution range of 1-1500 μm was used. It had been dried at ambient temperatures for about 2 months, giving a dry weight percentage of 95.97%. The organic content was about 2-4 wt%. Diesel was added to the dried soil two weeks before the soil was put into the elevator, in order to ensure good adhesion to the soil matrix. The polluted soil was frequently mixed during the two adsorption weeks. For the run with real polluted soil (number 6), polluted clay soil was excavated, dried and sieved over the 1.5 mm mesh. More than 90 wt% of this soil was smaller than 100 μm, confirming the very fine character. The organic content in this clay was about 5 wt%.

Microorganisms

A mixed microbial community capable of degrading oil pollutants was used in the process. The community was obtained from a polluted harbour sludge originating from an oil refinery. Species from the genera *Pseudomonas*, *Acinetobacter*, *Xanthomonas* and *Alcaligenes* were shown to be present. At the start of the first run, the first reactor was inoculated with an overnight culture of the community. This culture was made in the mini-plant medium using a shake-flask at 30 °C, without solid phase, diesel was added as carbon and energy source. The experiments were started in fed batch mode until the desired solid hold-up of 20 wt% was reached, and then switched to continuous operation. The second reactor was switched to the continuous mode as soon as a total volume of 20 liter had been pumped over from the first reactor.

Monitoring the decontamination process

The solids hold-up and the particle size distribution in the different reactor compartments, were monitored during the mini-plant operation. The solids hold-up was determined by frequently sampling the reactor compartments and measuring the soil fraction. To determine the particle size distribution, different soil fractions were flushed with water over a series of vibrating sieves with decreasing mesh.
The pollutant concentrations at the different locations in the mini-plant were monitored by frequently sampling the slurry and determining the total amount of pollutant present in the soil. The slurry samples were first dewatered by means of filtration using paper Soxhlet holders. The filtered samples were subsequently treated in a continuous Soxhlet extraction for five hours, using a methanol/toluene mixture (4:1) (Puttman, 1988). Hexadecene, not dominantly present in oil products as diesel, was used as an internal standard. The extract was analyzed for its total pollution content by capillary gas chromatography (G.C.).

Finally, the amount of pollutant present in the extract was combined with the dry weight of the slurry sample, in order to determine the pollutant concentration present in the solid phase. Using this method, an average extraction yield of 92% was achieved. A standard deviation for the Soxhlet-gas chromatography procedure of about 10% was determined. This is acceptable when compared to other quantitative analysis methods for mineral oils (Ouboter, 1988).

To determine the dissolved pollutant concentration, slurry samples were centrifuged. After separation of the pellet and the supernatant, the latter was extracted with a solvent (ether) in a shake-flask. Subsequently the solvent was analysed for its pollutant content (using hexadecene as internal standard) using G.C.

Since it is possible that dissolved pollutant is stripped by the air flow, the gas flows leaving the reactors were monitored for pollutants. For this purpose, off gas was lead through tubes containing solid adsorption material suitable for the gas phase adsorption of hydrocarbons (Tenax). After several days of being exposed to the off gas the adsorption material was extracted with a solvent which subsequently was analyzed by G.C.

Oxygen- and carbon dioxide concentrations in the in- and outlet gas were monitored on-line using a (paramagnetic) O₂-analyzer (Taylor Analytics) and a CO₂-infrared analyzer (Beckman model 864); dissolved oxygen concentrations were measured with an oxygen electrode.

**Experimental strategy**
Degradation kinetics and mini-plant performance were studied under continuous process conditions for a period of more than 1 year. In six different runs the kinetics were determined, one duplicate run was carried out (run 2'). Table 2 presents an overview of the experiments. The first 3 runs (no 1, 2 ans 2') were test runs, mainly used to test practical features of the system.

Table 2 Experimental conditions, steady state concentrations, overall reduction and conversion rates for the 7 mini-plant runs.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>1</th>
<th>2</th>
<th>2'</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Recirculation flow</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>+</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Residence time (hr)</td>
<td>2*100</td>
<td>2*100</td>
<td>2*100</td>
<td>2*100</td>
<td>2*50</td>
<td>2*50</td>
<td>2*100</td>
</tr>
<tr>
<td>Oil conc [g/kg soil]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Entering soil Reactor 1 -fines</td>
<td>16.8</td>
<td>11.5</td>
<td>10.8</td>
<td>4.8</td>
<td>8.6</td>
<td>5.2</td>
<td>2.4</td>
</tr>
<tr>
<td>Reactor 1 -coarse</td>
<td>20.4</td>
<td>13.6</td>
<td>11.0</td>
<td>12.1</td>
<td>16.7</td>
<td>8.7</td>
<td>2.2</td>
</tr>
<tr>
<td>Reactor 2 -fines</td>
<td>3.0</td>
<td>1.7</td>
<td>1.6</td>
<td>1.8</td>
<td>0.9</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td>Remixed soil after dewat.</td>
<td>18.0</td>
<td>10.5</td>
<td>8.5</td>
<td>5.7</td>
<td>12.8</td>
<td>6.5</td>
<td>2.4</td>
</tr>
<tr>
<td>Overall reduction (%)</td>
<td>67</td>
<td>65</td>
<td>72</td>
<td>76</td>
<td>74</td>
<td>79</td>
<td>67</td>
</tr>
<tr>
<td>Overall conversion rate [g/kg/h]</td>
<td>1.4</td>
<td>1.0</td>
<td>1.0</td>
<td>0.4</td>
<td>1.7</td>
<td>1.0</td>
<td>0.2</td>
</tr>
</tbody>
</table>

The first four experiments (numbers 1, 2, 2' and 3) were performed with a soil residence time of 200 hours (100 hours per reactor), and decreasing influent concentrations of 16.6, 11.5, 10.8 and 4.8 g pollutant/kg soil. Experiments 4 and 5 were carried out using a shorter residence time of 100 hours (50 hours per reactor) and influent concentrations of 8.6 and 5.2 g/kg. The runs 1 to 5 were carried out with the deliberately polluted soil (distribution from 1 μm to 1500 μm).

The last experiment was carried out with a residence time of again 200 hours for a 40 year old oil spill, having an average concentration of 2.4 g/kg.
The polluted soil originated from an oil refinery and contained for more than 90% silt and clay (smaller than 100 μm). Since recirculation of the process water appeared to be problematic (see results and discussion) during the test runs, no recycling was applied during the last three runs.

The separation performance of the DIITS-reactor was of crucial importance in the mini-plant. For this purpose and to test other practical matters, three test runs were carried out (runs numbered 1, 2 and 2'). After a satisfactory operation had been established, the mini-plant was monitored during the last four runs (3, 4, 5 and 6) by means of the following parameters:

* solids hold-up
* concentration of pollutant
* oxygen and carbon dioxide conversion rates
* ammonium and nitrate concentrations
* proton production rate

RESULTS AND DISCUSSION

Mini-plant slurry characteristics

During the test runs, the slurry characteristics in the mini-plant were investigated. The particle size distributions shown in figure 4 demonstrate that the soil entering the reactor separates in the DIITS-reactor into a suspended fine fraction and a coarse fraction, which is fluidized at the bottom of the reactor. Around 70% of the suspended particles had diameters smaller than 100 μm, while about 90% percent of the fluidized material was larger than 100 μm, and can therefore be classified as sand (see figure 1). Although the suspended solids present in the bulk of the first and second reactor showed a similar pattern, a small difference can be seen. In the second reactor, the 100 and 210 μm fractions were smaller than in the first reactor, the opposite holds for the 53 μm fraction. The desintegration of the soil aggregates during slurry processing were considered responsible for this effect.
Figure 4 Particle size distribution of the solid phase in the mini-plant, monitored at several points.

In figure 5 the particle size distribution in the bulk and bottom compartments of the DITS-reactors are shown. From figures 4 and 5 it was calculated that about 50% (on weight basis) of the solids entering the first reactor leave the system by the bottom effluent flow. The other half was transported with the bulk effluent flow to the second bioreactor.
Solids hold-up

During runs 3, 4, 5 and 6 the process parameters mentioned above were monitored. With respect to the steady state solids hold-up figure 6 shows a considerable variation. The average hold-up of suspended solids was around 0.1 v/v (~20 wt%) in the bulk of both reactors, and about 0.5 v/v (60 wt%) in the bottom compartment of the DITS-reactor. The measured hold-ups thus were larger than the set points given in table 1. An increase in solids hold-up in the bottom compartment (day 31) corresponded with a decrease in particle loading in the bulk compartment. The development of the solids hold-up in the second reactor was comparable with that in the bulk zone in the first reactor. Since the power input in the reactors was constant during the runs, the drift in the solids hold-up was considered to be due to changes in the entering soil composition.
Figure 6 Solids hold-up in the two mini-plant reactors.

Steady state pollutant concentrations

Figure 7 shows the courses of the influent pollutant concentrations for the two bioreactors. The aim of the experiments was to investigate the influence of the pollutant concentration and the soil residence time on the decontamination process. For the first reactor, the concentrations are given for the dry soil added from the elevator. For the second reactor, the influent concentration corresponds to the bulk effluent concentration of reactor 1. Figure 8 shows the difference between the concentrations (in g/kg soil) measured in the bottom zone (coarse fraction) and in the bulk compartment. It was concluded that about ten times more pollutant adsorbed to the fines than to the coarse fraction. This inhomogeneous distribution is in line with our expectations (Assink, 1988). It is clear that the steady state pollutant concentration in the second reactor was only slightly
smaller than that in the first reactor. The final concentration in the effluent soil was determined after recombination (mixing and dewatering) of the slurry from the second reactor and the coarse fraction from the first reactor. The calculated overall conversions are given in table 2.

![Graph](Image)

**Figure 7** Pollutant concentration (g/kg soil) in the influent soil for both mini-plant reactors.

To see whether the pollutant was partly stripped by the aeration flow, or partly removed by way of the liquid phase, the pollutant concentration in the effluent gas and liquid flows were monitored. The concentrations in the water phase were found to be below detection level (≈ 100 μg/l). Also no pollutant could be detected in the gas phase leaving the reactors.
Figure 8 Steady state pollutant concentrations (g/kg soil) in the DITS-reactor (bottom and bulk) and the second reactor.

Oxygen consumption and carbon dioxide production rates

In order to determine whether oxygen transfer from the gas phase to the slurry was rate-limiting, the oxygen mass transfer coefficient (k_{la}) and the dissolved oxygen concentration were determined for the mini-plant. For a bulk solids hold-up of 0.1 v/v and an air flow rate of 10 l/min (superficial gas velocity = 0.0023 m/s), the k_{la} in the first reactor was found to be 0.003 s^{-1}. In the second reactor, for an air flow of 7.5 l/min (superficial gas velocity = 0.0018 m/s), the k_{la} was 0.0024 s^{-1}. During processing, an average dissolved oxygen concentration of 2-3 mg/l was measured. Although
mass transfer in aerated dense slurry systems can be a problem (De Lasa, 1986), this was obviously not the case in the soil slurry reactors.

By measuring the difference between the oxygen and carbon dioxide concentrations in the influent and effluent air flows, the courses in the oxygen uptake and carbon dioxide production rates can be calculated by means of mass balances over the gas phase (see part 2). Figure 9 shows the oxygen consumption rates for the first and second reactors.

![Graph of Oxygen Consumption Rates](image)

**Figure 9** Oxygen consumption rates (mol/l/hr) in the mini-plant.

Figure 10 shows the carbon dioxide production rates for both reactors. Comparing the two reactors, it becomes clear that in the second reactor the oxygen consumption rates and the carbon dioxide production rates were only about 40 to 50% of those measured in the first reactor.
Figure 10 Carbon dioxide production rates (mol/l/hr) in the mini-plant.

Steady state proton production and ammonium and nitrate concentration

Figure 11 shows the steady state ammonium and nitrate concentrations for the four runs considered. It can be clearly seen that recirculation of the process water (run 3) resulted in the build up of nitrate in the two reactors. This was not considered beneficial since high levels of nitrate in bioreactors frequently result in high levels of (toxic) nitrite. At the end of run 3 (around day 24), the recirculation was stopped. As is shown, the nitrate concentration dropped within a period of about 1 residence time (≈ 8 days) to much lower concentrations. The increase during run 6 is due to the different origin of the soil. From the results depicted it was concluded that, recirculation of the process water can only succeed in combination
with an adjustment of the medium supply flow. This was not done in the experiments described.

In addition to the change in nitrate concentration caused by recirculation effects, microbial nitrification (conversion of $\text{NH}_4^+$ into $\text{NO}_3^-$) took place in the system. It is shown that, for each of the runs, the steady state concentrations of $\text{NH}_4^+$ in reactor 1 were a factor 10 lower than the influent $\text{NH}_4^+$ concentration of about 0.01 mol/l. The conversion of ammonium is confirmed in reactor 2 where the ammonium concentration practically dropped to zero.

![Diagram showing ammonium and nitrate concentrations](image)

**Figure 11** Ammonium and nitrate concentrations (mol/l) in the mini-plant.

Figure 12 gives an illustration of the NaOH control flow which is used to maintain the pH at 7. It is assumed that this control equals the proton production rate. Shown is that in the first reactor the production rate of
protons rather fluctuated. These fluctuations were probably due to changes in the buffering capacity of the soil entering the system. In the second reactor the fluctuations were much more damped.

**Figure 12** NaOH addition rates (≈ proton production rates) in the mini-plant.

**Overall degradation rates in the mini-plant**

Table 2 shows the steady state pollutant concentrations, and the overall reduction and conversion rates. The data given are averaged values measured over a period of several weeks. From these data, it can be seen that in the mini-plant about 70% overall reduction was reached. This overall reduction was independent of the residence time of the solid phase and the influent concentration of the pollution.
Based on an analytical error in the pollutant concentration (Soxhlet and GC together) of about 10%, in combination with the error made in slurry sampling (estimated at 5%), the total relative measurement error in the overall reduction was about 30%. On the basis of this error, the overall reductions given in table 2 do not differ significantly.

CONCLUSIONS

The mini-plant as a scale model

From the first year of mini-plant operation, the following features can be given for the slurry process

* Microbial decontamination of oil polluted soil can be performed in a slurry reactor. Overall degradation rates varying from 1.7 to 0.2 g oil/kg soil/day were measured (table 2). From the fact that the amount of pollutants analyzed in the off gas and in the liquid phase were negligible it follows that the measured removal (table 2) of the pollution must be due to microbial conversion.

* The conversion rates achieved in the slurry process were about 100-200 times higher than those obtained in landfarming (Bonnier, 1980).

* In accordance with the full scale design, the separation of incoming soil into fine and coarse fractions could be carried out without difficulty in the DITS-reactor.

* The conversion rates in the second reactor were smaller than those in the first reactor (figure 8). Consequently, the oxygen consumption rate in the second reactor was 40-50% of that in the first reactor, 0.6 mmol/l/hr and 1.3 mmol/l/hr respectively (figure 9).

* Recirculation of the process water can only succeed in combination with an adjustment (decrease in nitrogen concentration) of the nutrient supply flow.
* Significant nitrification took place in the system. As shown in figure 11, ammonium was converted to steady state levels of around zero.

* The overall amount of reduction reached with the deliberately polluted soil was comparable to the reduction achieved with soil (clay) polluted with oil about 40 years ago (run 6). This validates the use of deliberately polluted soil in a model system.

Degradability of soil pollutants

To investigate the difference in degradation between the 40 years old landspill and the deliberately pollution (diesel), some chromatograms taken from mini-plant samples are compared. The composition of the diesel pollution, before and after treatment, is shown in figure 13.

![Chromatogram of diesel contaminated soil](image-url)
If these chromatograms are compared to those made for the old oil spill, shown in figure 14, a similar trend can be seen. The large peaks represent the linear aliphatics, while the branched and aromatic hydrocarbons are gathered in the "bump" (Gough and Rowland, 1990). The total peak and "bump" area were significantly reduced after processing. Low molecular compounds were more readily degraded than high molecular compounds, as can be seen from the shape of the chromatograms. The same kind of hydrocarbon patterns were also found in other biodegraded oils (Teschner and Wehner, 1985; Fedorak and Westlake, 1981).

![Chromatograms of oil contaminated soil](image)

**Figure 14** Effect of processing on oil chromatographs.

**Bio-availability and soil decontamination**
The small difference in the steady state pollutant concentrations between reactors 1 and 2 (figure 8), indicates that a relatively small amount of pollution was converted in the second reactor. This relatively low conversion rate was confirmed by the differences in the oxygen consumption and carbon dioxide conversion rates in the two reactors (figures 9 and 10). These lower conversion rates were apparent despite the fact that biomass, generated in the first reactor, is continuously transported to the second reactor, the biomass concentration therefore will be larger than that in the first reactor.

The combination of high steady state pollutant concentrations and low oxygen consumption rates in the second reactor can be explained by a restricted availability of the pollutant.

If the rate limiting step in the second reactor is assumed to be the desorption process by which the pollutant is released from the solid phase, the desorption kinetics can be estimated. From the constant overall reduction measured for runs 1 to 3 (equal \( r_{\text{soil}} \), different \( C_{\text{in}} \)), it follows that the desorption kinetics are not zeroth order. Assuming first order desorption kinetics, with \( k_{\text{des}} \) being the desorption constant, and \( C_{\text{p}} \) the pollutant concentration in the soil, the following equation holds

\[
C_{\text{out}} = C_{\text{in}}^p \exp[-k_{\text{des}}*r_{\text{soil}}]
\]

For a constant value of \( k_{\text{des}} \), equation 1 results in a constant ratio between the inlet and outlet concentrations of reactor 2. This is in reasonable agreement with the results given in table 2.

Assuming this first order desorption process to be rate-limiting in the decontamination, it can be calculated that in order to reach the Dutch A-level (50 mg/kg) for the oil spill, a soil residence time of about 4 weeks is required. If decontamination down to the Dutch B-level (1000 mg/kg) is sufficient, a residence time of 6 days would be enough.
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LITERATURE


Microbial decontamination of polluted soils in a slurry process

Part 2 Monitoring conversion rates

R.H. Kleijntjens, C. Hellinga, M.J. Geerdink
Kluver Laboratory for Biotechnology, Delft University of Technology,

ABSTRACT

Adequate monitoring of the microbial decontamination activity during soil processing is desired in order to take immediate action in case toxic compounds enter the slurry system. Time-consuming and expensive, off-line analysis of the pollutant is not appropriate for this purpose. Experiments have been performed to find out if on-line gas phase reactants (O_2 and CO_2) can be used for monitoring purposes. Correlations have been established between the conversion rates of the gas phase reactants and the pollutant conversion rate. To eliminate the influence of measurement errors in these correlations, the (mini-plant) data were evaluated by means of a consistency test. From consistent data sets, it could be concluded that the correlations varied significantly (about a factor 3). Despite these variations, there are good reasons to prefer the carbon dioxide production rate above the oxygen consumption rate as on-line parameter.

KEYWORDS Biological soil reclamation, mineral oils, suspension bioreactor, degradation kinetics, consistency test.
INTRODUCTION

A first question with respect to the applicability of a biotechnological slurry process for the reclamation of excavated polluted soils, is the degradability of the pollutant under treatment (see part 1). In order to establish this, biological tests should be carried out in advance. However, due to the large amounts of soil to be treated in an installation, and its inhomogeneous character, it is unlikely that a representative number of samples can be investigated. This leaves open the possibility for toxic compounds such as pesticides or heavy metals to enter the continuous treatment system.

A possible indicator of contamination by toxic compounds is a decrease in the microbial activity. Should this occur in the continuous process, immediate action should be taken. The most straightforward would be to stop the intake of the soil causing the problems. This action would result in a decrease of the solids hold-up, leading to a reduction of the toxic concentration. Other options are the addition of chemical oxidants to destroy the toxic compounds, the use of chelators or a pH increase to remove harmful metals.

Adequate on-line monitoring of the degradation process is therefore required. For this purpose, the time consuming, off-line analysis of the pollutant is not an appropriate monitoring method. Most appropriate are gas phase reactants, since they show a fast response and can be sampled on-line.

This paper examines the possibility to determine the pollutant conversion by means of an on-line monitoring of oxygen or carbon dioxide conversion rates. For this purpose, correlations between the pollutant and the gas phase reactants must be established. A prerequisite is the absence of large measurement errors in the experimental data. To investigate the reliability of the data from the mini-plant soil processing (see part 1), a data consistency test will be applied. Using reliable data sets, the existence of unambiguous correlations was investigated. As a first step in the derivation of the test, a black box conversion rate model for the slurry system will be constructed.
BLACK BOX CONVERSION RATE MODEL

A black box model is constructed for the microbial decontamination of oil polluted soil in a slurry bioreactor (see part 1, Kleijnjtjens et al., 1991). In the model only the major chemical compounds involved will be regarded. Between these major compounds, the following stoichiometric equation is proposed for oil conversion (Roels, 1980)

\[ \text{CH}_{1.8}^+ + a \cdot \text{NH}_4^+ + b \cdot \text{O}_2 \rightarrow c \cdot \text{CH}_{1.8}^0 \cdot 0.5 \cdot \text{N}_0.2^+ + d \cdot \text{CO}_2 + e \cdot \text{H}_2\text{O} + a \cdot \text{H}^+ \]  

[1]

In this equation, one C-mole of pollutant (\text{CH}_{1.8}) is aerobically converted into biomass (\text{CH}_{1.8}^0 \cdot 0.5 \cdot \text{N}_0.2), carbon dioxide and water (Kleijnjtjens et al, 1989). It is assumed that no toxic compounds are formed, and that the mineralization is complete (Schlegel, 1962; Bossert and Bartha, 1984, Atlas, 1981). For the elemental composition of the pollutant, the composition of diesel, which is assumed to have an average composition for oil products (see figures 13-14 in part 1), was taken. The elemental biomass composition was taken from literature (Roels, 1980). In equation 1, the stoichiometric coefficients a, b, c, d and e are not further specified within this paper. Despite the fact that this reaction may give the essence of the microbial conversion, it is unlikely that this is the only conversion taking place in the slurry system. Since the contents of soil organics may add up to 30-40 grams per kilogram dry soil (see part 1), a significant turnover of these organic compounds (humification) can be expected in addition to the oil breakdown. In addition, it was shown in part 1 that nitrification takes place, thus nitrate is also a relevant compound (Schlegel, 1985). Since it is impossible to distinguish between all of these parallel reactions, it was considered appropriate to lump all bio-organics (excluding the pollutant) into one "compound", the soil organics. For the elemental composition of this lumped "compound" (which thus includes micro-organisms), the average composition of humus is taken from literature (\text{CH}_{1.9}^0 \cdot 0.5 \cdot \text{N}_0.02) (Povoledo and Golterman, 1975). Thus eight relevant compounds are defined in the slurry system
The black box model is depicted in figure 1. Arrows towards the box indicate that the compound is consumed, arrows from the box represent production in the slurry reactor.

**Figure 1** Schematic representation of the black box model.

Using the elemental composition of the eight compounds considered, the matrix, denoted as $E$, is defined according to figure 2. In combination with this matrix, the conversion rates of the compounds can be represented in a vector $r_a$. In this vector, rates have a positive value when the compound is formed and a negative value when it is consumed. The laws of conservation
for elements and charge, dictating that no elements or charge can be formed or consumed, is expressed as (Roels, 1980)

\[ E \times r_a = 0 \]  

According to equation 2, the black box consists of a set of five linear equations (see figure 2), one for each element and one for the charge. This set of five equations contains eight unknown conversion rates, one for each of the compounds. In order to solve these equations, at least three rates have to be determined experimentally. For the continuous slurry process at steady state, the conversion rates can be determined (by means of the macroscopic mass balances) from the net exchange flows with the environment.

\[
\begin{array}{cccccc}
\text{pollutant} & \text{C} & \text{H} & \text{O} & \text{N} & \text{charge} \\
\text{carbon diox.} & 1 & 1.8 & 0 & 0 & 0 \\
\text{oxygen} & 1 & 0 & 2 & 0 & 0 \\
\text{ammonium} & 0 & 0 & 2 & 0 & 0 \\
\text{nitrate} & 0 & 4 & 0 & 1 & 1 \\
\text{protons} & 0 & 0 & 3 & 1 & -1 \\
\text{water} & 0 & 0 & 0 & 1 & 1 \\
\text{soil organics} & 1 & 2 & 1 & 0 & 0 \\
\end{array}
\]

\[
\begin{array}{cccccc}
\text{E} & \text{r}_a \\
\text{C} & \text{H} & \text{O} & \text{N} & \text{charge} \\
\end{array}
\]

\[
\begin{array}{cccccc}
r_p & r_{CO_2} & r_{O_2} & r_{NH_4} & r_{NO_3} & r_H \\
r_{H_2O} & r_{org} \\
\end{array}
\]

**Figure 2** Elemental matrix and conversion rates for the black box model.

**MACROSCOPIC MASS BALANCES**

Of the relevant conversion rates in the degradation process, the water and biomass conversion rates could not be determined in the slurry reactor.
Starting with the soil mass balance, the mass balances for the six measured compounds will now be derived for the first bioreactor.

Soil

In a continuous mode, assuming steady state, the mass flow of soil entering the slurry reactor, $\Phi_{m,in}$, is equal to the sum of the mass of soil in the bulk effluent flow, $\Phi_{v,s}$ (with a solid hold-ups of the suspended fraction equal to $\epsilon_s$, see part 1), and in the bottom effluent flow, $\Phi_{v,fb}$ (with a hold-up equal to $\epsilon_{fb}$)

$$\Phi_{m,in} = \Phi_{v,s} \cdot \rho_{s,fb} \cdot \epsilon_s + \Phi_{v,fb} \cdot \rho_{s,fb} \cdot \epsilon_{fb} \quad [3]$$

Pollutant

During degradation, the major part of the pollutant is converted to biomass, carbon dioxide and water, while the remainder leaves the system in the two effluent flows. The macroscopic steady state mass balance is derived from the general mass balance expression

$$\text{input of pollutant} = \text{output of pollutant} + \text{conversion} \quad [4]$$

Considering the slurry bioreactor (figure 3), it can be seen that the input originates from the pollutant present in the incoming soil $\Phi_{m,in}$. The output is divided over three flows which each may contain a certain amount of pollutant

* the upper effluent flow $\Phi_{v,s}$
* the bottom effluent flow $\Phi_{v,fb}$
* the gas phase, due to stripping, $\Phi_{v,gas}$
Figure 3 Flows to and from the DITS-reactor in the mini-plant.

The last term in equation 4, the amount of pollutant which is consumed in the reactor is not shown in figure 3 (this term equals $r_p \cdot V_r$). In equation 5 pollutant is considered to be present in both the solid phase ($C_{p,s}$ and $C_{p,fb}$) and the liquid phase ($C_{p,l}$). In equation 5 $r_p$ is negative when consumtion takes place

$$
\Phi_{m,in} \cdot C_{p,in} = \Phi_{v,s} \cdot \left( \rho_{s,s} \cdot \epsilon_{s} \cdot C_{p,s} + (1-\epsilon_{s}) \cdot C_{p,l} \right) + \\
\Phi_{v,fb} \cdot \left( \rho_{s,fb} \cdot \epsilon_{fb} \cdot C_{p,fb} + (1-\epsilon_{fb}) \cdot C_{p,l} \right) + \\
\Phi_{g} \cdot C_{p,g} - V_{r} \cdot r_p
$$

[5]

From the experimental results discussed in part 1, it was concluded that the concentrations of the pollutant in the water and gas phase were negligible. Considering the relevant terms in equation 5 the pollutant conversion rate can be expressed as
\[ \frac{r}{p} = -\frac{\Phi_{m, in} \cdot C_{p, in}}{V_r} + \left( \rho_{s, s, \epsilon_s} \cdot \phi_{p, s} \cdot \rho_{s, \phi} \cdot \epsilon_{fb} \cdot \phi_{p, fb} \cdot V_{r} \right) + \left( \rho_{s, fb} \cdot \epsilon_{fb} \cdot C_{p, fb} \cdot \phi_{v, fb} \right) \]  

[6]

From experiments determining the steady state values of the parameters on the right of equation 6, the pollutant conversion rate can be calculated.

Oxygen

In a steady state, the oxygen uptake rate in the first slurry reactor is related directly to the difference in the oxygen concentration measured in the injection and effluent air flows. Under the experimental conditions \((r_{gas, phase} \ll r_{slurry})\), the amount of dissolved oxygen leaving the system with the slurry effluent flow can be neglected compared with the consumption rate. The oxygen consumption rate reads as

\[ r_{O_2} = \left( \frac{\Phi}{V_r} \right) \cdot (C_{O_2, g, out} - C_{O_2, g, in}) \]  

[7]

Carbon dioxide

For the carbon dioxide production rate under steady state conditions, a similar expression to that for oxygen holds. The rate of removal of dissolved carbon dioxide or (bi)carbonate transported by the slurry effluent flows is again much smaller than the consumption rate. The carbon dioxide production rate can then be calculated from on-line gas analysis as

\[ r_{CO_2} = \left( \frac{\Phi}{V_r} \right) \cdot (C_{CO_2, g, out} - C_{CO_2, g, in}) \]  

[8]

Nitrogen

Because the mass balances for ammonium and nitrate are similar, only that for ammonium consumption is given here. The complete balance contains incoming
terms due to ammonium present on the soil and in the medium supply, two effluent terms (slurry flows) and the ammonium conversion rate

\[
\Phi_{m,\text{in}} C_{\text{NH}_4,\text{in}} + \Phi_{v,\text{med}} C_{\text{NH}_4,\text{med}} = \\
\Phi_{v,s} (1-\epsilon_s) C_{\text{NH}_4,\text{l}} + \Phi_{v,fb} (1-\epsilon_{fb}) C_{\text{NH}_4,\text{l}} - V_r r_{\text{NH}_4}. \tag{9}
\]

To simplify the expression for the ammonium conversion rate, the underflow \(\Phi_{v,fb}\), containing only a small portion of the liquid phase, is neglected. In addition, the amount of ammonium present in the soil entering the system, \(C_{\text{NH}_4,\text{in}}\), is considered to be negligible. The expression for ammonium consumption now reads as

\[
r_{\text{NH}_4} = \frac{-\Phi_{v,\text{med}} C_{\text{NH}_4,\text{med}} + C_{\text{NH}_4,\text{l}} (1-\epsilon_s) \Phi_{v,s}}{V_r} \quad \tag{10}
\]

The expression for the nitrate production rate can be derived in a similar way.

**Protons**

For the proton balance, both the NaOH flow (to keep the pH ≈ 7) and the neutralizing capacity of soil are considered. This buffering capacity, \(\delta\text{ANC}\), is defined as the difference in acid neutralizing capacity between the soil entering and leaving the reactor. The two neutralizing flows are counteracted by two "production" terms the medium supply flow rate and the proton production rate due to pollutant conversion and nitrification. The proton balance reads as

\[
\Phi_{v,\text{med}} C_{\text{H,med}} + V_r r_{\text{H}} = \Phi_{v,\text{NaOH}} C_{\text{OH}} + \Phi_{m,\text{in}} \delta\text{ANC} \quad \tag{11}
\]

This results in the following expression for the proton conversion rate:

\[
r_{\text{H}} = \frac{\Phi_{v,\text{NaOH}} C_{\text{OH}} + \Phi_{m,\text{in}} \delta\text{ANC} - \Phi_{v,\text{med}} C_{\text{H,med}}}{V_r} \quad \tag{12}
\]
The conversion rates in the slurry system can now be calculated from experimental data on the parameters in the above equations. Since the conversion rates determined this way will contain measurement errors, the consistency of the data is tested.

CONVERSION RATES UNDER THE RESTRICTION OF THE LAWS OF CONSERVATION

As was stated above, the set of linear equations describing the conservation of the chemical elements and the charge can be solved in this case, when three out of the eight conversion rates are measured. However, it is unlikely that the (three) measured rates are without measurement errors (standard deviations are presented). The solved set of equations will therefore contain errors which depend on these standard deviations. In addition, the system description may not be correct. Relevant compounds may not have been taken into account or the elemental or charge composition of the compounds incorrectly defined. To be able to detect these errors, it is necessary that the set of equations is overdetermined. More than the minimum of three rates must be measured for this.

When the system is overdetermined, the validity of the system description and the measurements can be checked. The logic behind this check is that, from measurements, balanced conversion rates that exactly fulfill the constraints given by equation 2 can be generated. Madron et al. (1977) presented a procedure that generates the new balanced set of rates. The adjustments applied to the measured rates are a function of the standard deviations of the measured rates and the structure of the set of equations (the procedure which generates the adjustments is called balancing). Accurate measurements will be adjusted only slightly, while inaccurate measurements can be subject to larger corrections. By comparing the adjustments to the measured standard deviations, one can determine whether they are reasonable (i.e. whether they are within the range of the measured deviations).

A quantitative statistical test for this comparison was given by Wang and Stephanopolous (1983). If the measured data set cannot be balanced according to the test, it must be concluded that either the measurements are
inconsistent or that the system description is incorrect. A distinction between these two conclusions cannot be made.

To find out whether a specific measured rate disturbs the balancing procedure, the system must be overdetermined by more than one rate. It is then possible to discard one of the measured rates and perform the balancing procedure and the consistency check again. If the inconsistency vanishes, it can generally be assumed that the omitted rate is faulty, or is closely related to the errors in the system description. If the data set is considered consistent, it can be concluded that the data set on which the balancing is based is not in contradiction with itself or with the black box description. For the calculations described in this paragraph, the computer program MACROBAL was used (Hellinga, 1991).

MATERIALS AND METHODS

To determine the consistency of the experimental data from the mini-plant (see table 2 of part I) runs number 3, 4, 5 and 6 were considered. For each of these runs, the following parameters were monitored during the steady state

* solids hold-up in reactor and effluent flows
* pollutant concentrations in the different soil fractions and in the water and gas phases
* oxygen and carbon dioxide conversion rates
* ammonium and nitrate concentrations
* proton production rate

From the values of these measured quantities and the other process parameters, the conversion rates were calculated using the mass balances.

To estimate the measurement error in the pollutant conversion rate, not only variations in the effluent flows and other process parameters should be taken into account. Also errors in the analysis of the compounds are relevant. For the sake of simplicity, small errors are omitted (see table 1) and variance cross terms (e.g. \( \text{var } C_d \times \text{var } \Phi_{m,in} \)) are considered negligible.
For the conversion rate of the pollutant, the variance, \( \text{var}(r_p) \), can be calculated according to

\[
\text{var}(r_p) = \frac{1}{v_r^2} \left [ \Phi_{m, in}^2 \text{var}(C_{p, in}) + C_{p, in}^2 \text{var}(\Phi_{m, in}) + \rho_s^2 \right ]
\]

\[
\epsilon_{s, s}^2 \left ( C_{p, s}^2 \text{var}(\Phi_{v, s}) + \Phi_{v, s}^2 \text{var}(C_{p, s}) + C_{p, s}^2 \Phi_{v, s}^2 \text{var}(\epsilon_{s, s}) + C_{p, fb}^2 \Phi_{v, fb}^2 \text{var}(\epsilon_{s, fb}) \right ]
\]  \[13\]

**Table 1** Mean value and variance of the slurry reactor parameters.

<table>
<thead>
<tr>
<th>parameter</th>
<th>mean value</th>
<th>unit</th>
<th>st. dev.</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Phi_{m, in} )</td>
<td>0.07</td>
<td>kg/h</td>
<td>0.0035</td>
</tr>
<tr>
<td>( C_{p, in} )</td>
<td>10</td>
<td>g/kg</td>
<td>1</td>
</tr>
<tr>
<td>( C_{p, s} )</td>
<td>15</td>
<td>g/kg</td>
<td>1.5</td>
</tr>
<tr>
<td>( C_{p, fb} )</td>
<td>2</td>
<td>g/kg</td>
<td>0.2</td>
</tr>
<tr>
<td>( V_r )</td>
<td>20</td>
<td>l</td>
<td>negl.</td>
</tr>
<tr>
<td>( \rho_{s, s} )</td>
<td>2.05</td>
<td>kg/l</td>
<td>negl.</td>
</tr>
<tr>
<td>( \rho_{fb, s} )</td>
<td>2.3</td>
<td>kg/l</td>
<td>negl.</td>
</tr>
<tr>
<td>( \epsilon_{s, s} )</td>
<td>0.07</td>
<td>1/l</td>
<td>0.007</td>
</tr>
<tr>
<td>( \epsilon_{fb, s} )</td>
<td>0.48</td>
<td>1/l</td>
<td>0.048</td>
</tr>
<tr>
<td>( \Phi_{v, s} )</td>
<td>0.10</td>
<td>1/hr</td>
<td>0.01</td>
</tr>
<tr>
<td>( \Phi_{v, fb} )</td>
<td>0.01</td>
<td>1/h</td>
<td>0.001</td>
</tr>
</tbody>
</table>

For the conversion rates of oxygen, carbon dioxide, protons, ammonium and nitrate the errors can be calculated in the same way. The values of the various variances are given in tables 2, 3, 4 and 5.

**RESULTS/DISCUSSION**
For run numbers 3, 4, 5 and 6 the experimental data from the first reactor in the mini-plant (see part 1) were used to calculate the averaged conversion rates according to equations 2 to 12. In the first three columns of tables 2, 3, 4 and 5, the measured conversions rates are given with their variances. It can be seen that the measurement errors in all of the six conversion rates are considerable (varying between 2 and 100% relative error).

**Table 2** Measured and "balanced" set of conversion rates in the first reactor from run 3

<table>
<thead>
<tr>
<th>compound</th>
<th>measured rates</th>
<th>absolute measur. error</th>
<th>&quot;balanced&quot; rates</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(mmol/1/hr)</td>
<td>(mmol/1/hr)</td>
<td>(mmol/1/hr)</td>
</tr>
<tr>
<td>pollutant</td>
<td>-0.32 B</td>
<td>0.19</td>
<td>-0.28</td>
</tr>
<tr>
<td>carbon dioxide</td>
<td>0.38 B</td>
<td>0.02</td>
<td>0.37</td>
</tr>
<tr>
<td>oxygen</td>
<td>-1.12 B</td>
<td>0.195</td>
<td>-1.19</td>
</tr>
<tr>
<td>ammonium</td>
<td>-0.35</td>
<td>0.043</td>
<td>-0.35</td>
</tr>
<tr>
<td>nitrate</td>
<td>-0.72</td>
<td>0.26</td>
<td>0.36</td>
</tr>
<tr>
<td>protons</td>
<td>0.84 B</td>
<td>0.25</td>
<td>0.71</td>
</tr>
<tr>
<td>water</td>
<td>-</td>
<td>-</td>
<td>0.6</td>
</tr>
<tr>
<td>soil org.</td>
<td>-</td>
<td>-</td>
<td>-0.1</td>
</tr>
</tbody>
</table>

**Table 3** Measured and "balanced" set of conversion rates in the first reactor from run 4

<table>
<thead>
<tr>
<th>compound</th>
<th>measured rates</th>
<th>absolute measur. error</th>
<th>&quot;balanced&quot; rates</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(mmol/1/hr)</td>
<td>(mmol/1/hr)</td>
<td>(mmol/1/hr)</td>
</tr>
<tr>
<td>pollutant</td>
<td>-0.75 B</td>
<td>0.5</td>
<td>-0.46</td>
</tr>
<tr>
<td>carbon dioxide</td>
<td>0.95 B</td>
<td>0.12</td>
<td>0.93</td>
</tr>
<tr>
<td>oxygen</td>
<td>-1.18 B</td>
<td>0.33</td>
<td>-1.37</td>
</tr>
<tr>
<td>ammonium</td>
<td>-0.53</td>
<td>0.07</td>
<td>-0.14</td>
</tr>
<tr>
<td>nitrate</td>
<td>-0.09</td>
<td>0.037</td>
<td>0.18</td>
</tr>
<tr>
<td>protons</td>
<td>0.38 B</td>
<td>0.17</td>
<td>0.33</td>
</tr>
<tr>
<td>water</td>
<td>-</td>
<td>-</td>
<td>0.58</td>
</tr>
<tr>
<td>soil org.</td>
<td>-</td>
<td>-</td>
<td>-0.47</td>
</tr>
</tbody>
</table>
Table 4 Measured and "balanced" set of conversion rates in the first reactor from run 5

<table>
<thead>
<tr>
<th>compound</th>
<th>measured rates (mmol/l/hr)</th>
<th>absolute measur. error (mmol/l/hr)</th>
<th>&quot;balanced&quot; rates (mmol/l/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pollutant</td>
<td>-0.32 B</td>
<td>0.07</td>
<td>-0.34</td>
</tr>
<tr>
<td>carbon dioxide</td>
<td>1.47 B</td>
<td>0.18</td>
<td>1.35</td>
</tr>
<tr>
<td>oxygen</td>
<td>-1.83 B</td>
<td>0.29</td>
<td>-1.97</td>
</tr>
<tr>
<td>ammonium</td>
<td>-0.55</td>
<td>0.038</td>
<td>-0.29</td>
</tr>
<tr>
<td>nitrate</td>
<td>-0.048</td>
<td>0.033</td>
<td>0.38</td>
</tr>
<tr>
<td>protons</td>
<td>0.93 B</td>
<td>0.36</td>
<td>0.68</td>
</tr>
<tr>
<td>water</td>
<td>-</td>
<td>-</td>
<td>0.64</td>
</tr>
<tr>
<td>soil org.</td>
<td>-</td>
<td>-</td>
<td>-1.0</td>
</tr>
</tbody>
</table>

Table 5 Measured and "balanced" set of conversion rates in the first reactor from run 6

<table>
<thead>
<tr>
<th>compound</th>
<th>measured rates (mmol/l/hr)</th>
<th>absolute measur. error (mmol/l/hr)</th>
<th>&quot;balanced&quot; rates (mmol/l/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pollutant</td>
<td>-0.13 B</td>
<td>0.17</td>
<td>-0.12</td>
</tr>
<tr>
<td>carbon dioxide</td>
<td>0.24 B</td>
<td>0.13</td>
<td>0.21</td>
</tr>
<tr>
<td>oxygen</td>
<td>-1.02 B</td>
<td>0.49</td>
<td>-1.32</td>
</tr>
<tr>
<td>ammonium</td>
<td>-0.26</td>
<td>0.062</td>
<td>-0.50</td>
</tr>
<tr>
<td>nitrate</td>
<td>0.36</td>
<td>0.14</td>
<td>0.50</td>
</tr>
<tr>
<td>protons</td>
<td>1.04 B</td>
<td>0.02</td>
<td>1.04</td>
</tr>
<tr>
<td>water</td>
<td>-</td>
<td>-</td>
<td>0.71</td>
</tr>
<tr>
<td>soil org.</td>
<td>-</td>
<td>-</td>
<td>-0.089</td>
</tr>
</tbody>
</table>

Consistency of the data

From the experimentally derived conversion rates, the balanced rates were calculated according to the method of Wang and Stephanopoulos (1983). For
the periods considered, the balanced rates are shown in the last columns of tables 2, 3, 4 and 5. For all the four steady states, the data sets were not consistent on the basis of the six measured rates. However, being three times overdetermined, it was possible to leave out two measured rates. Two conversion rates were selected for which, when omitted, all four runs would give consistency of the four remaining rates. By trial and error, it was found that the ammonium and nitrate conversion rates should be omitted to suit this purpose. The consistent rates are denoted with a B in the first column of the tables.

In agreement with the measurements, the consistent data sets give a negative sign (consumption) for oxygen, pollutant and soil organics, while carbon dioxide and protons are produced. The fact that the data consistency was only obtained when the ammonium and nitrate were excluded, illustrates that either the nitrification is not well described by the black box model or that large measurement errors are present in these rates. The conversion of soil organics is in agreement with previous reports, stating that humification is a relevant factor in microbial soil processing (Lotter et al., 1990).

**Correlation between gas phase reactants and the pollutant**

The established consistent data sets incorporate the conversion rates of the oxygen, carbon dioxide and the pollutant. The correlations between these rates can now be examined. Figure 4 shows the correlations for the 4 runs under consideration. It is clear that both the ratios \( \frac{r_p}{r_{O_2}} \) and \( \frac{r_p}{r_{CO_2}} \) varied significantly (about a factor 3). These variations indicate that, although sometimes found in the literature (Barenschee et al., 1990), oxygen and carbon dioxide do not provide an unambiguous measure for the degradation of pollutants.

However, oxygen and carbon dioxide do have a purpose for the monitoring of the slurry process. As shown in the black box model (figure 1), oxygen and carbon dioxide are clearly related to the conversion of the pollutant. The element carbon only is present in the pollutant, the soil organics and carbon dioxide. According to the model, \( CO_2 \) not originating from the pollutant breakdown, is due to the turnover of soil organics. Oxygen
however, is not only present in the pollutant and the soil organics but also in the nitrate. Due to the fact that the nitrification process in the reactor is not well understood, the role of oxygen is less unambiguous than that of carbon dioxide. Using the carbon dioxide conversion rate as a parameter for on-line monitoring, will avoid the problems related to the nitrification.

![Graph showing correlations between \( \frac{r_p}{r_{O_2}} \) and \( \frac{r_p}{r_{CO_2}} \).]

**Figure 4** Correlations between \( \frac{r_p}{r_{O_2}} \) and \( \frac{r_p}{r_{CO_2}} \).

It is clear that, for monitoring purposes, the correlation between \( r_{CO_2} \) and \( r_p \) is to be preferred above the correlation between \( r_{O_2} \) and \( r_p \). From the mini-plant results, the average ratio for \( \frac{r_p}{r_{CO_2}} \) was about 0.5, with the other 50% of the carbon dioxide production coming from the humification processes.

**Complexity of the soil slurry**

The fact that the role of nitrification in the process remains unclear, and that, even for consistent data sets, the \( \frac{r_p}{r_{O_2}} \) and \( \frac{r_p}{r_{CO_2}} \) correlations are scattered, emphasizes the complexity of the phenomena in the slurry reactor.

* The presence of many different compounds in the oil pollutants (over 500 different compounds may be found), each with its own
affinity for the soil and its specific microbial degradation rate, results in a diffuse breakdown pattern (Perry, 1984).

* A wide variety of microorganisms are responsible for the breakdown of the pollutant.

* The chemical and physical properties of the soil particles determine the extent of the adsorption of the pollutant. Much of the pollutant is not directly available for microbial breakdown (Knezovich et al., 1987), desorption phenomena play a crucial role in the breakdown.

* The presence of organic soil material in the reactor leads to an aerobic turnover of carbon (Povoledo and Golterman, 1975). In addition to this humification process, oxygen consumption due to processes including nitrification, sulphur oxidation and protozoa activity also proceed in parallel with the conversion of the pollutant.

Considering these complicating factors, it becomes obvious that from the mini-plant experiments, the degradation kinetics cannot be revealed in detail. In addition, it must be noted that because of the complexity described above, the pollutant itself should be monitored in order to detect whether the soil is decontaminated to the desired level. On-line gas phase measurements cannot replace the time consuming off-line detection of the pollutant, they can only be used as an indicator to monitor the overall activity in the slurry.

CONCLUSIONS

* Based on the law of element and charge conservation, a black box model for the conversion of the major compounds in the slurry reactor has been derived. In this model, all organic matter except the pollutant is lumped into one compound.
* Balancing techniques provide a means of checking the validity of the black box description.

* Using the balancing technique, the consistency of the experimental mini-plant data (see part 1) was checked. It was established that the ammonium and nitrate conversion rates disturbed the consistency of the data. Omitting these two rates, the remaining oxygen, carbon dioxide, protons and pollutant conversion rates were shown to be consistent.

* From the problems that the ammonium and nitrate conversion rates caused in the consistency tests, it was concluded that nitrification is not well understood/described in the slurry system.

* In line with literature, the aerobic conversion of soil organics in the slurry reactor (humification) was incorporated in the black box model.

* Based on the four reliable conversion rates (pollutant, oxygen, carbon dioxide and protons), it was concluded that the correlations between the oxygen and carbon dioxide conversion rates and the pollutant conversion rate are not unambiguous.

* On-line gas phase measurements cannot replace the time consuming off-line monitoring of the pollutant in order to determine the conversion in the slurry process. However, the carbon dioxide production rate can be used as a control parameter to monitor the overall activity in the slurry.

ACKNOWLEDGEMENT

The authors are grateful to A. v.d. Graaf, L. Velthuisen, L.A. Robertson, M. van Loosdrecht and H. Noorman for their theoretical support. Also they are grateful for the cooperation of C. van der Veen, C. Erkelens and the indispensable technical support of S. Lispet, P. Kroon, P. Vetter and G. van der Toolen.
### SYMBOLS

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANC</td>
<td>acid neutralizing capacity</td>
<td>g/kg</td>
</tr>
<tr>
<td>C</td>
<td>concentration</td>
<td>g/kg</td>
</tr>
<tr>
<td>C_{p,s}</td>
<td>based on solid phase</td>
<td>g/kg</td>
</tr>
<tr>
<td>C_{p,l}</td>
<td>based on aqueous/slurry phase</td>
<td>g/l</td>
</tr>
<tr>
<td>kla</td>
<td>mass transfer coefficient</td>
<td>h^{-1}</td>
</tr>
<tr>
<td>r</td>
<td>conversion rate</td>
<td>mol/l/h</td>
</tr>
<tr>
<td>V_r</td>
<td>reactor volume</td>
<td>l</td>
</tr>
<tr>
<td>x_s</td>
<td>volume fraction of soil</td>
<td>[-]</td>
</tr>
<tr>
<td>\Phi_m</td>
<td>mass flow of soil</td>
<td>kg/h</td>
</tr>
<tr>
<td>\Phi_v</td>
<td>volumetric liquid flow</td>
<td>l/h</td>
</tr>
<tr>
<td>\Phi_g</td>
<td>volumetric gas flow</td>
<td>l/h</td>
</tr>
<tr>
<td>\rho_s</td>
<td>soil density</td>
<td>kg/l</td>
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<tr>
<td>\tau</td>
<td>residence time</td>
<td>h</td>
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</table>

### INDICES

<table>
<thead>
<tr>
<th>Index</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>accumulated compound</td>
</tr>
<tr>
<td>evap</td>
<td>evaporation</td>
</tr>
<tr>
<td>fb</td>
<td>fluidized bed</td>
</tr>
<tr>
<td>g</td>
<td>gas</td>
</tr>
<tr>
<td>H</td>
<td>protons</td>
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<tr>
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<td>medium</td>
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<tr>
<td>s</td>
<td>soil, suspension</td>
</tr>
<tr>
<td>tot</td>
<td>total</td>
</tr>
<tr>
<td>x</td>
<td>biomass</td>
</tr>
</tbody>
</table>

### LITERATURE


Madron F., Veverka V., Vanecek V., AIChE-journal, 23, 482 (1977)


THREE PHASE SOIL SUSPENSION REACTORS
Particle suspension hydrodynamics of an air agitated tapered reactor

R.H. Kleijntjens, R.G.J.M. van der Lans, K.Ch.A.M. Luyben
Kluver Laboratory for Biotechnology, Delft University of Technology
Julianalaan 67, 2628 BC Delft, The Netherlands

ABSTRACT

In order to describe the particle suspension hydrodynamics in a tapered gas agitated three-phase slurry reactor, a mathematical model was developed. This model is based on turbulent energy conversion. To determine the model parameters, suspension experiments with sand particles (260 μm diameter) have been carried out at four reactor volumes (up to 4 m³) with four different slurry densities (up to 40 wt%). It is shown that the hydrodynamic state in a particle suspension can be described by two turbulent parameters, the liquid fluctuation velocity and the eddy length. The latter parameter is the most sensitive with respect to scale and slurry density.

KEYWORDS

slurry reactor, particle distribution, suspension hydrodynamics, turbulence, eddy length
INTRODUCTION

Suspension reactors have found many applications both in the chemical and mining industries for different multi-phase processes. As well in the case of two (s-l) as in the case of three phase (s-l-g) suspension reactors it is crucial to keep the solid phase in suspension and mix the phases intensively (Nienow, 1968; De Lasa, 1986). Mixing in two phase (s-l) systems is generally done by mechanical stirring. In three phase systems, agitation is often performed by the gas phase (Shah, 1979). Especially when aerobic processes are dealt with, gas agitation is beneficial since particle suspension, mixing and oxygen transfer is achieved efficiently. In the design of a slurry process for the biological decontamination of excavated polluted soils, suspension and aeration of the solid phase were crucial. In addition, it was shown that only for a low power input the full scale process is economic feasible (Kleijntjens et al., 1990 and 1991). Therefore an efficient air agitated system was looked for. Among the various types described in literature, the concept of the air agitated Pachuca tank was considered most appropriate. This system, originating from mineral industries, operates in practice at low a power input (50-100 W/m³) (Merriman, 1958; Hallett et al., 1981).

In the soil slurry process, a tapered gas agitated (s-l-g) tank with a height/width ratio equal to one is used as a bioreactor. In this reactor suspended polluted soil particles are aerobically decontaminated by means of micro organisms under controlled conditions (Kleijntjens et al., 1989 and 1990). For an efficient decontamination process, it is necessary to obtain complete particle suspension at minimum power input with sufficient oxygen transfer. The suspension of the solid phase however, was shown to be a major bottleneck in the process. In order to achieve a proper reactor design and scale-up, it was considered neccessary to study first a model system. In the model system, the complex, not well defined, polydisperse soil is replaced by monodisperse quartz sand (d_p = 260 μm).

This paper describes the investigation of the particle suspension behavior as a function of power input, solid hold-up and scale for the tapered reactor. For this purpose, experiments were carried out at several reactor scales, and a mathematical model describing the suspension hydrodynamics of the monodisperse solid phase has been developed.
The suspension hydrodynamics of a monodisperse solid phase in slurry bubble columns (S-L-G) is often described by the sedimentation-dispersion model (Jean et al., 1989; Cova, 1966). According to the model, the axial solid hold-up distribution in a batch operated slurry column is

$$
\epsilon_s(z) = \epsilon_s(0) \times \exp \left( - \frac{u_p^\infty \times H}{E_s} \times z \right)
$$

[1]

In the derivation of equation 1 (in which $z$ is the dimensionless axial height), it is assumed that no radial gradients are present in the column, and that both the settling velocity and the dispersion coefficient are height independent. The dimensionless group in the exponent, determining the course of the axial distribution, is defined as the particle Peclet number (Kato et al., 1972)

$$
Pe_p = \frac{u_p^\infty \times H}{E_s}
$$

[2]

A small Peclet number indicates that the dispersion process proceeds faster than the settlement giving a small coefficient in equation 1 and therefore results in a homogeneous state of suspension. An inhomogeneous state of suspension corresponds to large Peclet numbers, resulting in an curved axial hold-up distribution with the highest hold-up at the reactor bottom. In slurry columns the measured bottom hold-up generally doesn't exceed values of $\epsilon_s \sim 0.15$-0.2 (Pandit and Joshi, 1983). For most of the experimentally established axial solids hold-up distributions in columns, the $Pe_p$ number can be determined without difficulty by means of curve fitting.

For two phase (s-l) stirred suspension reactors, the sedimentation-dispersion model is not frequently used. In these systems, the particle behavior is often described by means of the minimum power input required to just sustain a state of suspension. In this state of minimal suspension, the particles do not remain on the reactor bottom for longer than 1-2 seconds (the Zwietering criterion). For stirred tank reactors the stirrer speed at minimal suspension is directly related to the properties of the system

$$
N_{ms} = S_{sys} \times \nu^{0.1} \times d_p^{0.2} \times \left( g \Delta \rho / \rho_1 \right)^{0.45} \times \epsilon_{(tot)}^{0.13} \times D_i^{-0.85}
$$

[3]
Equation 3 is based on a dimensional analysis, combined with a set of experimental data from which the numerical values of the exponential coefficients were derived (Zwietering, 1958; Harnby et al., 1986). The required minimal stirrer speed depends on properties such as the particle size, the viscosity and the solids hold-up (which normally doesn't exceed values of 0.2 - 0.3 (Chapman et al., 1983)). The reactor scale determines also the minimal stirrer speed since, for a standard vessel, the impeller diameter $D_1$ is proportional to the reactor diameter $T$, which is related to the volume. Based on this relationship, the power input equation for stirred vessels $P \sim N^3 \cdot D_1^5$ (Beek and Mutzall, 1975), and the Zwietering equation, the scale dependency of the power input can be derived as

$$P/V_{ms} = \frac{N^3 \cdot D_1^5}{V_r} \sim \frac{D_1^{-2.55} \cdot D_1^5}{T^3} \sim \frac{D_1^{2.45}}{D_1^{-0.55} \cdot T^{-0.55} \cdot V_r^{-0.18}}$$

[3a]

The sedimentation-dispersion model and the Zwietering equation are not based on mechanistic concepts. This disadvantage becomes manifest in the fact that neither approach provides a relationship between the power input, the liquid motion and the solid hold-up distribution. To achieve this relationship, more fundamental modelling of suspension phenomena, using turbulent theory, has been emphasized by several authors. (Baldi et al., 1977; Joshi, 1983; Shamlou and Koutsakos, 1989).

In this paper a mathematical model is described, which determines the solid phase dispersion coefficient ($E_s$) from turbulent parameters. The mechanistically derived $E_s$ is used in the sedimentation-dispersion model to predict the solids hold-up distribution in the tapered reactor.

**HYDRODYNAMIC SUSPENSION MODEL**

In the following section, a hydrodynamic suspension model is introduced in three steps. Firstly, the turbulent particle suspension is described, followed by the structure of the mathematical model for the tapered reactor. A parameter estimation procedure, in which the model parameters are derived from suspension experiments, is then given.
Turbulent particle suspension

In particle suspension theories, turbulence has been used to describe the interaction between the motion of a discrete particle and the surrounding flow (Hetseroni, 1982; Hinze, 1972, Narayanan et al., 1969). Within this framework, a particle response number was introduced characterizing the hydrodynamic state of a particle suspension (Kleijntjens et al., 1991). This number represents the ability of the particle to follow turbulent motion. It is defined as the ratio of the particle response time, $\tau_p$, giving a measure for the particle inertia, and a characteristic time, $\tau_t$, for the change of motion in a turbulent flow

$$\frac{\tau_p}{\tau_t} = \frac{(0.04 \times (\rho_p + \beta \times \rho_1) \times d_p^2) / \eta_f}{l_e / u'}$$  \hspace{1cm} [4]

For response numbers smaller than 0.1, the particle inertia time is smaller than the characteristic time of the liquid motion, indicating that the particle is able to follow the turbulent flow (suspension regime). With values above 0.1, complete particle suspension is impossible since the particle is too inert to respond adequately to changes in the supporting flow. To calculate the value of the response number, the liquid and solid phase properties, the fluctuation velocity, $u'$, and the length of the energy containing eddy, $l_e$, have to be known.

Not only for the particle response number the eddy length and the fluctuation velocity are important, also these parameters are crucial in the description of the liquid dispersion coefficient which is, in an isotropic turbulent flow, characterized by (Hinze, 1959)

$$E_1 = u' \times l_e$$  \hspace{1cm} [5]

Also, the specific power dissipation in isotropic turbulent flows can be described using $u'$ and $l_e$ (Batchelor, 1953)

$$\epsilon_t = \frac{u'^3}{l_e}$$  \hspace{1cm} [6]

Equation 6 shows that the energy transfer rate in a turbulent flow is determined by the hydrodynamics of the energy containing eddies. Based on
the crucial role of these eddies in turbulence, the mathematical suspension model introduced in this paper relates both the eddy length and the fluctuation velocity to the parameters used in the sedimentation-dispersion model: $E_s$ and $Pe_p$.

**Mathematical model**

The first step in the model development is the analysis of the steady state flow pattern in the tapered reactor. As is shown in figure 1, the flow pattern is assumed to consist of three zones, with equal cross sectional areas.

![Diagram of flow pattern in tapered reactor](image)

**Figure 1** Overall flow pattern in the tapered reactor.

The plume zone contains most of the rising bubbles, resulting in an upwardly directed bulk motion. For continuity reasons, the total capacity of the downward directed bulk flow in the two wall zones must equal the upflow capacity. The average upward liquid velocity must therefore be twice the average downward liquid velocity. The flow pattern is based on the pattern proposed by Lamont for conical Pachuca reactors (Lamont, 1958). A major difference between the flow pattern in the tapered system and in a bubble column (Cananda et al. 1978, Chen et al. 1989) is the presence of two relative stable shear layers over the total reactor height at both sides of
the plume zone. In these shear layers turbulence is generated in relation to
the velocity gradient as described by (Tennekes and Lumley, 1973)

\[
\frac{u'}{l_e} = \frac{\Delta U_Z}{\Delta y}
\]  

[7]

According to this equation, the ratio of the velocity difference between the
up and downward bulk motion \(\Delta U_Z\), and the width of the layer \(\Delta y\) determines
the ratio of \(u'\) and \(l_e\).

For simplicity, it is assumed that the characteristic eddy length, \(l_e\), and
the turbulent fluctuation velocity, \(u'\), are constant over the reactor at a
specific power input. The turbulence is considered to be isotropic. With
these assumptions, the flow of energy corresponding to the depicted flow
pattern can be seen in sequential steps

1) The first step is the introduction of power to the system. The
   amount of power present in the gas phase at the point of entry is
given by the work equation for rising bubbles (Lamont, 1958)

\[
p_{\text{in}}^{\text{gas}} = p^0 \ast Q^0 \ast \ln \left[ \frac{p^0 + \rho_{sl} \ast g \ast H}{p^0} \right]
\]

[8]

2) The incoming power is used for the generation of bulk motion. It
   is assumed that the complete buoyant power is transferred to the
   bulk of the system (Nicklin, 1962).

3) Turbulence is generated in the shear layers and subsequently
   spread through the reactor (Hinze, 1958). During this generation
   process power is withdrawn from the bulk motion.

4) Part of the turbulent power generated is used for maintenance of
   the slurry motion (kinetic energy), and the remainder maintains
   the potential energy of the particles in suspension.

5) Assuming that in each of the preceding steps the amount of power
   transferred is equal to \(P_{\text{in}}\), the heat finally produced by viscous
dissipation equals the incoming power (Davies, 1972).
The third and fourth steps in this sequence are crucial since the generation of turbulence is rate determining for both the energy transfer rate, and the suspension of particles. The power balance in the fourth stage may be derived in terms of the (total) change in potential energy of the particles per unit of time and the change in kinetic energy of the slurry motion per unit of time. Since \( \frac{dE}{dt} \) is equivalent to power, the specific kinetic power dissipation \( (\epsilon_{kin}) \) is used to incorporate the slurry motion in the power balance. The power balance is written as

\[
p_{gas}^{\text{part}} \frac{d}{dt} \left[ -P_{\text{pot}}^{\text{tot}} \right] + \epsilon_{kin} \ast v \ast \rho_{sl}
\]

Being the crucial, but unmeasurable, model parameter, the rate of kinetic energy transfer \( (\epsilon_{kin}) \) will be expressed in two experimental quantities: the power input and the solids hold-up distribution. For this purpose the total rate of change in potential energy of a particle suspension is considered, first the rate of change for one discrete particle is given

\[
\frac{d}{dt} \left[ -P_{\text{pot}}^{\text{part}} \right] = \frac{p}{p} \ast (\rho_{s} - \rho_{1}) \ast g \ast u_{p}^{\infty}
\]

In equation 10 the rate of loss in potential energy is defined as the resulting gravity force acting on the particle \( (V_{part} \ast \Delta p \ast g) \) multiplied by its settling velocity. In the general case, for a particle suspension with a three dimensional solids hold-up distribution \( \epsilon_{s}(x,y,z) \), the total rate of change in potential energy is given by integration over all the particles in the reactor. Since in particle swarms the settling velocity is related to the solids hold-up, the local velocity \( u_{p}(x,y,z) \) must be considered. The generalized form of equation 10 reads as

\[
\frac{d}{dt} \left[ -P_{\text{pot}}^{\text{part}} \right] \text{tot} = g \ast (\rho_{s} - \rho_{1}) \ast \int \int \int_{0-1,1} u_{p}(x,y,z) \ast \epsilon_{s}(x,y,z) dx dy dz
\]

To reduce the complexity of this integration, two simplifications are made. First, the solids hold-up distribution is not regarded in its three dimensional form. As indicated in figure 1, gas injected at the bottom of the reactor results in upward and downward directed bulk flow in the \( y,z \)-plane. Gradients in the \( x \)-direction are assumed to be negligible in the
tapered reactor. Second, the radial profile in the y-direction is incorporated into the axial term. For this purpose, the radial-averaged solids hold-up at the dimensionless height \( z^* / H \), is defined as

\[
\varepsilon_s(z) = \frac{\int_0^z \varepsilon_s(y, z) \, dy}{Y_z} \quad [12]
\]

In this equation \( Y_z \) is the maximal radial distance defined from the center of the tapered reactor at height \( z \). The radial averaged hold-up is also used to determine the settling velocity as a function of the axial distribution only. To relate the settling velocity in the suspension to the solids hold-up, the correlation from Barnea and Mirzahi (1973) was considered most appropriate.

Using the radial averaged axial solids hold-up distribution, and approximating the total reactor volume as being a completely tapered volume \( V_R = \tan 30^\circ \cdot D \cdot H^2 \), equation 11 can be rewritten as

\[
\frac{d \varepsilon_{part}^p}{dt} \bigg|_{tot} = 2 \cdot V_R \cdot g \cdot \Delta \rho \cdot \int_0^1 (u_p(z) \cdot \varepsilon_s(z) \cdot z) \, dz \quad [13]
\]

By means of equation 13 it is possible to determine the total rate of change of potential energy from an experimentally determined particle hold-up distribution. Having established the power input and the rate of change in potential energy, equation 9 can be rewritten in order to determine the rate of kinetic energy transfer in the slurry

\[
\varepsilon_{kin} = \frac{\frac{d \varepsilon_{part}^p}{dt}}{V_R \cdot \rho_{sl}^p} \bigg|_{tot} \cdot \frac{P_{in}}{\varepsilon_s(z)} \quad [14]
\]

In equation 14 the average slurry density, \( \rho_{sl}^p \), can be calculated from the total solids hold-up in the system. Neglecting the gas hold-up, and gradients in solids hold-up the average slurry density is described as

\[
\rho_{sl} = (1 - \varepsilon_s(z)) \cdot \rho_1^p + \varepsilon_s(z) \cdot \rho_s^p \quad [15]
\]
A crucial assumption for the model is the statement that the kinetic energy transfer according to equation 14, is equal to the rate of energy transfer maintaining the turbulent motion ($\varepsilon_t$). This assumption is in agreement with the proposed energy conversion sequence. Using the equality of $\varepsilon_t$ and $\varepsilon_{\text{kin}}'$ it is possible, by means of equation 6, to relate the turbulent fluctuation velocity to $\varepsilon_{\text{kin}}$ and the eddy length

$$u' = \left( \varepsilon_{\text{kin}}' \right)^{1/3} l_e$$  \[16\]  

Considering the turbulence generation in the tapered reactor as determined by free shear layers, the particle dispersion process is described as (Hinze, 1959)

$$E_s = 2 \times u' \times l_e$$  \[17\]

From the turbulent fluctuation velocity (equation 16) and a known eddy length, the solids dispersion coefficient thus can be derived. It should however be noted that for this purpose the liquid and solids dispersion coefficients are assumed to be equal. As discussed in the introduction, this equality may only be assumed for mobile, small, particles having $r_p/r_t < 0.1$. Although depending on reactor scale and power input, the mobility of (sand) particles with $d_p < 300-400 \mu m$ are in agreement with this criterion (Kleijntjens et al, 1991).

Next step in the model is the calculation of the Pe number from the dispersion coefficient from the equations 17 and 2. To predict the solids hold-up distribution in the tapered reactor, finally the sedimentation-dispersion model is rewritten using the radial averaged hold-up

$$\frac{\varepsilon_s(z)}{\varepsilon_s(0)} = \exp[-Pe \times \frac{z}{p}]$$  \[18\]

However, to describe the solids hold-up distribution according to equation 18, its validity for the tapered reactor has to be proven. For this purpose a derivation of the sedimentation-dispersion model for the tapered system was carried out. It was shown that the final result is equal to the one achieved for bubble columns. Taking into account only the settling velocity and an axial dispersion coefficient, the sedimentation-dispersion model obviously does not incorporate effects in directions other than axial. Equation 18 may therefore be used for the tapered system.
The aim of the model described above, is to determine the hydrodynamic state of a particle suspension by means of the parameters \( l_e \), \( u' \), \( E_s \) and \( P_e \). As shown in figure 2, of these four state parameters, only the eddy length is used as an input parameter. To solve the model for a particular experiment, the numerical value of \( l_e \) is therefore necessary. Since \( l_e \) can not directly be determined experimentally, an estimation procedure is used. The parameter estimation for \( l_e \) comes from a comparison between the predicted radial averaged distribution according to the model (figure 2), and the experimentally derived hold-up data.

![Figure 2 Calculation scheme of the mathematical model.](image)

**EXPERIMENTAL EQUIPMENT AND METHODS**

Experiments were carried out in a three phase (solid-liquid-gas) gas agitated tapered reactor with an angle of the tapered part equal to 60°. Particle suspension was studied at 4 four different scales with a volume range of 0.080-0.240-0.4-4.0 m³ and for four solids hold-up 10-20-30-40 wt% (volume fraction of 0.0414, 0.0885, 0.143 and 0.205 respectively). Figure 3
shows a schematic representation of the reactor configuration. According to the principle of geometrical similarity, all reactors were designed with a height to width ratio of 1; the heights were 0.56, 0.97, 1.27 and 2.70 m, respectively. The only difference in the geometrical similarity were variations in the depth, D (x-direction); on the three smaller scales the depth of the system was 0.45 m, while at the pilot scale of 4.0 m³, the reactor depth was 1 m.

Gas was introduced at the bottom by means of an injection manifold with equidistant injection points made of vertical tubes with an inner diameter of 2 mm and a mutual distance of 2 cm. The gas flow was measured at the manifold with a rotameter. The pressure registered in the rotameter was used to express the gas flow for atmospheric conditions (Coleman, 1956). The standardized gas flow was used in the work equation for rising bubbles (equation 8), in order to determine the power input (Lamont, 1958). In order to circumvent maldistribution in the manifold, the cross sectional area of the manifold was chosen 3 times larger than the total injection surface area, as given by the sum of the injection openings (Dow and Shreveport, 1950).

**Figure 3** Schematic representation of the tapered reactor.
To verify the proposed flow pattern shown in figure 1, a liquid velocity probe has been developed for the measurement of local velocities. The probe consisted of an injection point, from which tracer material was pulsewise released, and a detection satellite 2 cm further out in the fluid (schematically this is depicted in figure 4). Velocities were measured by means of the tracer residence time between two detection rings in the satellite. The scale at which turbulence could be detected was determined by the distance between the rings in the satellite (2.5 cm). Due to the width of the satellite, tracer detection took place within a range of directions, as indicated by the windows of observation (figure 4). The satellite could be rotated around the central tracer injection point.

Two types of experiments were carried out with the probe. Firstly, the statistical chance on a specific flow direction was determined. For this purpose, the satellite is located at a specific position and the percentage of a number of pulses which reached the satellite was determined. The two directions for which the score percentage was determined are shown in figure 5: the vertical upward direction (a) and the downward direction parallel to the tapered wall (b). Secondly, the actual velocity of the pulses which reached the satellite was determined.

![Figure 4](image)

**Figure 4** Schematic representation of the velocity probe in the y,z plane.
Suspension experiments were carried out with monodisperse quartz sand with a diameter of 260 (+/-30) μm and a density of 2500 kg/m³. Experiments were conducted with minimal power input, since this gives the only really comparable hydrodynamic state on different scales and different solids hold-ups. To establish the minimum power input, excess gas (air) was injected to create a suspension, the critical input was then determined by a stepwise decrease in gas flow. Minimal suspension was detected by visual observation, and defined as that state in which a dense collar of slow slipping sand appeared just above the injection manifold (Hallett, 1981). With this method the minimal gas input could be reproduced within 5 %.

The axial particle hold-up distribution at the axis of the tapered system was determined with minimal suspension for all 16 experiments. The solids hold-up distribution was established using a sampling probe with a content of 9.42 ml. To measure the hold-up, the open probe is brought at the desired location, after several seconds, the probe is closed and withdrawn from the slurry. Subsequently, the probe content is analyzed for its dry weight. From the dry weight, the sampling volume of the probe and the densities of the solid and liquid phases, the volume fraction solids can be determined. At each location, three samples were taken within a period of several minutes. The standard deviation in the calculated solids hold-up varied between 5 and 30 %, probably because of fluctuations due to turbulence. Radial distributions were determined at scales of 0.4 and 4.0 m³ by radial sampling at several heights in the reactor.

**EXPERIMENTAL RESULTS**

Firstly, the proposed flow pattern, as depicted in figure 1, has been verified. Figure 5 presents the results for the statistical experiments carried out under two phase (g-1) conditions at a volume of 0.4 m³, a reactor height of 1.27 meter and a P/V equal to 120 W/m³. Each point in the figure represents the score percentage calculated from 300 injections. As is shown, in the center of the system, with a dimensionless radial distance of 0 (the systems center, note figure 3) at half reactor height, the chance of detecting upward liquid motion was 60 %. Close to the wall at a dimensionless distance of 0.8, the chance of detecting upward motion was reduced to 5 %. The opposite holds, for the velocity parallel to the wall
(regarded as downward motion). Close to the wall there was more than 90% detection chance, while in the middle of the plume this chance was reduced to less than 5%. Halfway between the plume center and the wall, the chances of detecting upward or downward motion were almost equal, which agrees with the proposed flow pattern.

![Diagram showing percentages of tracer pulses reaching the detection satellite.](image)

**Figure 5** Percentages of tracer pulses reaching the detection satellite.

For the average and maximum liquid upward velocities in the plume, values of 0.52 m/s and 0.9 m/s, respectively, were detected for a gas input of 0.0036 m³/s. A turbulence fluctuation velocity of 0.33 m/s, equal to 62% of the average velocity, was measured upwards in the plume zone. It became clear from measurements in the shear layers that the turbulence generated was not fully isotropic. In the z+ direction, the turbulence fluctuation velocity was more than twice the fluctuation velocity in other directions. If the measured velocities are averaged over the six main directions (x⁺, x⁻, y⁺, y⁻, z⁺, z⁻), fluctuation velocities in the order of 0.2 - 0.25 m/s were determined.

Experiments at a scale of 4.0 m³ showed that u' was rather insensitive for power input and scale, fluctuation velocities were found to be in between 0.2 and 0.3 m/s.
In figure 6, the specific minimal power input \((P/V)\) for the four scales and the four different solids hold-up are given in a logarithmic plot. It can be seen that at larger scales, less specific power was required to sustain a suspension. This trend held for all four slurry densities tested. Furthermore, it can be concluded that denser slurries require a larger specific power input to achieve a state of minimal suspension. To determine the coefficient which dictates the scale dependency (compare equation 3a), the regression lines are depicted in figure 6. As denoted in the figure the coefficient \(\alpha\) was rather constant, with an average value of -0.14.

![Figure 6 Double logarithmic representation of the measured specific power input \((P/V)\) at the condition of minimal suspension.](image)

The axial particle hold-up distribution at the central axis of the reactor \((x=0 \text{ and } y=0)\) was determined under minimal suspension condition. Figure 7 shows the results for each of the four slurry densities investigated.
Figure 7 Measured axial solids hold-up distribution at the axis of the system.

The data for the lowest hold-up of 0.041 (v/v) demonstrate that in the three smaller volumes (0.080, 0.240 and 0.4 m³) the axial profiles were comparable and had a shape agreeing with the standard axial profile described in equation 1. The profile at the larger volume (4.0 m³) and the profiles
depicted in the other three figures are somewhat S-shaped with a rather homogeneous bottom zone containing a maximal local solids hold-up of about 0.3-0.4 (v/v). These figures show that the volume of the homogeneous zone increased at higher slurry densities while the hold-up in this zone did not increase significantly. The observed S-shape in the axial distribution, dividing the suspension into a dense homogeneous bottom zone and a dilute inhomogeneous upper zone, differs considerably from the smooth curves reported for slurry bubble columns. The establishment of a single smooth axial curve is obviously impeded by density effects at the bottom of the reactor.

In contrast to slurry bubble columns, the axial gradient alone does not give all the information concerning the particle suspension behavior in the tapered reactor. In figure 8, radial particle hold-up profiles at different heights, for a volume of 0.4 m³ and an overall slurry density of 0.210 (v/v) are given. On the vertical axis, the ratio of the local hold-up at position (y,z) and the hold-up at the center with position (0,z) is given at four different heights. On the horizontal axis, the dimensionless radial distance from the center is given. The tapered reactor shape can be recognized if the points at the right side of each profile are connected.

The dimensionless representation \( \varepsilon_s(y,z)/\varepsilon_s(0,z) \) clearly demonstrates the shape of the radial profiles. In the homogeneous bottom zone (z=0.25) radial gradients were not present, but with increasing height, the radial profiles were more pronounced. To be able to define a radial averaged solids hold-up, and to capture these trends into a shape factor, the following relation is introduced

\[
q(z) = \frac{\varepsilon_s(z)}{\varepsilon_s(0,z)}
\]  

[19]

In the definition of \( q(z) \), equation 12 is used in order to calculate the radial averaged hold-up at a height \( z \). If the radial profile is rather flat, \( q(z) \) comes close to unity, for a pronounced radial profile \( q(z) \) reaches values below 0.5. Figure 8 shows that the shape of the radial profiles depends on height, making \( q \) a function of \( z \). However, for reasons of simplicity, a single average height independent shape factor is aimed at. From the measured data, other experiments carried out at the 0.4 and 4.0 m³ scale gave similar results to those depicted in figure 8, an estimation of
the average value of $q$ was made. Incorporating the flat profiles at the bottom and the sharp profiles at the top of the system, the average value for $q$ could be approximated with 0.6.

![Graph showing radial solids hold-up distribution](image)

**Figure 8 Measured radial solids hold-up distribution.**

**MODEL RESULTS**

In the parameter estimation, the axial hold-up distributions at the system axis, as shown in figure 7, are first transformed into radial averaged distributions using equation 19 with an average $q$ equal to 0.6. Secondly, since the sedimentation-dispersion model describes a free particle dispersion, only data from the upper zone are considered. Therefore equation 18 is rewritten, using the average solids hold-up at $z=1$, denoted as $\epsilon_s(1)$, as fixed point

$$\epsilon(z) = \epsilon_s(1) \times \exp\left[\frac{Pe}{p} \times (1-z)\right]$$

From the model and the experimentally established data, the eddy lengths can now be estimated. The results are shown as symbols in figure 9, the solid
lines are fitted curves. Also shown in figure 9 are the ratios of \( l_e/d_p \). The modelled eddy lengths increase as the scale increases but are also a function of slurry density.

Figure 9 Modelled eddy length, \( l_e \), as a function of scale and solids hold-up. Also shown is the ratio \( l_e/d_p \).

Figure 10 Modelled fluctuation velocity, \( u' \) as a function of scale and solids hold-up.
In figure 10, the model results for the fluctuation velocities are depicted. This parameter is rather insensitive for scale and slurry density. The dispersion coefficient and the Peₚ number, derived from \( l_e \) and \( u' \), are given in figures 11 and 12 (solid lines are fitted curves).

**Figure 11** Modelled dispersion coefficient, \( E_s \) as a function of scale and solids hold-up.

**Figure 12** Modelled Peₚ as a function of scale and solids hold-up.
Despite some irregularities, the overall trends in both parameters can clearly be seen. The increase of $E_s$ with scale is in agreement with the literature on slurry bubble columns (Joshi, 1980; Walter and Blanche, 1983).

**DISCUSSION**

The aim of the model was to provide a mechanistic background for the dispersion coefficient, $E_s$, in order to relate the particle suspension hydrodynamics to the power input. As shown in figures 9 and 10, the model parameters $l_e$ and $u'$ depend on the reactor scale and the solids hold-up. In order to quantify the modelled trends, $l_e$ and $u'$ are given as fitted functions of scale and solids hold-up in equations 21 and 22

$$l_e = 0.0063 \times \frac{V}{r}^{0.15} \times \epsilon_s^{(\text{tot})^{-0.78}}$$  \[21\]

$$u' = 0.11 \times \epsilon_s^{(\text{tot})^{-0.15}}$$  \[22\]

Equation 21 gives the volume dependence of $l_e$ as determined by means of regression analysis (the four solid lines drawn for the four slurry densities), from the data given in figure 9. As average value for the four slopes 0.15 was found, with a standard deviation of 0.013. For the density dependence an average value of -0.78, with a standard deviation of -0.07 was determined. Since both standard deviations were smaller than 10%, the trend in $l_e$ is reasonably covered by the correlation given in equation 21.

In contrast to the behavior of $l_e$, the model results for the turbulent fluctuation velocities (figure 10), were considered to be scale independent. The average value for the exponent representing the density dependency was found to be -0.15. This value was the calculated mean of four regression lines determined for each of the slurry densities respectively. The regression lines are not shown in figure 10. The standard deviation was relatively large, -0.055, giving a relative error of more then 30%, this indicates that the results for $u'$ are somewhat scattered.

If the correlations for $l_e$ and $u'$, are substituted in equation 6, a relation for the specific turbulent dissipation rate $\epsilon_t$ as a function of the reactor volume and the solids hold-up results
\[ \epsilon_t = 0.21 \times V_r^{-0.15} \times \epsilon_s^{(tot)}^{0.33} \quad [23] \]

By means of equation 23, the experimentally determined course in minimal power input (figure 6) can be explained. Considering the fact that more than 70 - 80% of the power present in a turbulent flow is used to maintain the turbulent dissipation rate (Turner, 1973), equation 23 may be rewritten as

\[ \frac{P}{V} = V_r^{-0.15} \times \epsilon_s^{(tot)}^{0.33} \quad [24] \]

Since in equation 21 the reactor volume and the solids hold-up have a more pronounced influence than in equation 22, the volume and solids hold-up dependencies according to equation 24 are largely determined by the behavior of the eddy length. In the interpretation of the scale effect in \( \frac{P}{V} \), the eddy length therefore is the most crucial parameter.

Also in the interpretation of the suspension behavior at higher slurry densities, the eddy length is the important parameter. This is illustrated by the sensitivity of \( l_e \) and \( l_e/d \) for the reactor scale and solids hold-up (figure 9). Concerning the relationship between reactor scale and eddy length, the ratio \( l_e/T \) is regarded. This ratio has been derived for slurry bubble columns as 0.08 (Pandit and Joshi, 1984). However, when the modelled eddy lengths (figure 9) are interpreted in terms of \( l_e/T \) ratios, no constant ratio is found. Taking as characteristic length scale for the tapered reactor the width at half height, the ratio \( l_e/T \) varies from 0.15 (\( V_r = 0.080 \ m^3 \)) and 0.06 (\( V_r = 4.0 \ m^3 \)) at low slurry densities (10 wt%) down to 0.05 (\( V_r = 0.08 \ m^3 \)) and 0.017 (\( V_r = 4.0 \ m^3 \)) at 40 wt% slurry density. Obviously the \( l_e/T \) ratio in the tapered reactor is a function of scale and slurry density.

The influence of the solids hold-up on the eddy length at higher densities is such that \( l_e/d \) becomes insensitive for power input and the scale. This means that free turbulent dispersion is hampered by particle interactions. If it is assumed that a critical limit of this ratio \( l_e/d \) exists, this limit was obviously reached in the bottom zone of the tapered reactor for a value of \( \epsilon_s(z) = 0.3 - 0.4 \), since in none of the experiments this hold-up was exceeded (figure 7). The measured axial profiles show that the only way of increasing the capacity of the reactor can be found in the enlargement of the homogeneous bottom zone and not by increasing the hold-up in the bottom part.
CONCLUSIONS

From the experiments in combination with the mathematical model the following conclusions can be drawn

* The mathematical model presented gives a relationship between the power input \( P_{in} \) and the hydrodynamic state in a particle suspension, \( P_{e,p} \). This relationship is useful for the design and scale-up of suspension reactors.

* The overall flow pattern, as depicted in figure 1, is in agreement with the trends in the measured score percentages (figure 5). In between two downward directed bulk flows at the side of the system, an upward directed bulk flow (the plume zone) was identified. At the interface of upward and downward liquid motion a highly turbulent region exists, in which the chance on upward and downward motion are about equal. This is the region where the shear layers, as depicted in figure 1, are located.

* The experimentally determined fluctuation velocities (0.20-0.25 m/s), are close to the model predictions (0.14-0.18 m/s). Regarding the restricted accuracy of the model, it may be concluded that the proposed model describes the energy conversion process reasonably well. Velocity measurements carried out on two different scales (0.4 and 4.0 m³) showed a limited sensitivity of the fluctuation velocity for scale and power input. These measurements also are in line with the model predictions.

* The experimentally determined scale effect in the \( P/V \) which gave \( \alpha = 0.14 \) (figure 6), is in accordance with the modelled scale effect which resulted in \( \alpha = 0.15 \) (equation 24). In addition, both exponents from equation 24 are in good agreement with the exponents of -0.18 and 0.39, as reported for two phase (s-l) stirred vessels (Zwietering 1958). The modelled effects in \( u' \) and \( l_e \), obviously cover the mechanisms implicitly present in the Zwietering equation.

* In contrast with the constant \( l_e/T \) ratio reported for slurry bubble columns (\( l_e/T = 0.08*T \)), for the tapered reactor \( l_e/T \) was shown to be variable. Both the increase of reactor scale and slurry density...
resulted in the decrease of the $l_e/T$ ratio, indicating that
turbulence is largely influenced by these two factors.

* In turbulent particle suspensions a critical ratio $l_e/d_p$ exists.
According to the data in figure 9, this critical ratio will be below 5.
Since in turbulent suspensions, the energy containing eddies have to
support the motion of the particle, the eddy size should be larger than
the particle diameter. It therefore is assumed that the critical ratio
$l_e/d_p$ is in between 1 and 5.

* The experimentally determined upper limit in solids hold-up reported for
bubble columns ($\varepsilon_s = 0.2/0.3$) and stirred vessels ($\varepsilon_s = 0.3$), as discussed
in the introduction, also might be due to the phenomenon of the critical
$l_e/d_p$ ratio.

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project.

LIST OF SYMBOLS

\begin{align*}
D & = \text{depth of tapered reactor (in direction x), m} \\
D_i & = \text{impeller diameter, m} \\
d & = \text{particle diameter, m} \\
E_{l} & = \text{dispersion coefficient for liquid phase, m}^2/\text{s} \\
E & = \text{dispersion coefficient for solid phase, m}^2/\text{s} \\
E_{\text{part}} & = \text{potential energy of the particles in suspension, J (kg.m}^2/\text{s}^2) \\
E_{\text{slur}} & = \text{kinetic energy of the slurry, J (kg.m}^2/\text{s}^2) \\
g & = \text{gravitational constant, 9.8 m/s}^2 \\
G & = \text{gas phase} \\
H & = \text{reactor height, m}
\end{align*}
\[ L = \text{liquid phase} \]
\[ l = \text{length of energy containing eddy, m} \]
\[ N_{ms} = \text{stirrer speed at condition of minimal suspension, s}^{-1} \]
\[ p = \text{pressure, kg/m.s}^2 \]
\[ \text{Pe} = \text{dimensionless Peclet number for the solid phase} \]
\[ p_{gas}^{in} = \text{incoming power with the gasphase, W} \]
\[ P/V_{ms} = \text{specific power input at minimal suspension, W/m}^3 \]
\[ q(z) = \text{shape factor for the radial profile} \]
\[ Q = \text{volumetric flow, m}^3/\text{s} \]
\[ S = \text{solid phase} \]
\[ S_{sys} = \text{system constant} \]
\[ t = \text{time, s} \]
\[ T = \text{reactor width (in direction y), m} \]
\[ U_Z = \text{velocity difference between up- and downward bulk motion, m/s} \]
\[ u(x,y,z) = \text{local particle settling velocity, m/s} \]
\[ u' = \text{turbulent fluctuation velocity (defined as the root mean square of the instantaneous fluctuation velocities), m/s} \]
\[ V_r = \text{reactor volume, m}^3 \]
\[ Y_{z\text{max}} = \text{maximum radial distance at height z, m} \]
\[ y_z = \text{radial distance at height z, m} \]
\[ z^* = \text{axial height in the reactor, m} \]
\[ z = \text{dimensionless reactor height (z^*/H)} \]

Greek symbols

\[ \alpha = \text{exponential coefficient representing the volume dependency of the specific power input P/V} \]
\[ \beta = \text{virtual coefficient for liquid displacement} \]
\[ \varepsilon_{(\text{tot})} = \text{total solids hold-up present in the reactor, m}^3 \text{solid/m}^3 \text{reactor} \]
\[ \varepsilon_{x,y,z} = \text{solids hold-up at position x,y,z, m}^3 \text{solid/m}^3 \text{reactor} \]
\[ \varepsilon_{y,z} = \text{solids hold-up at position y,z, m}^3 \text{solid/m}^3 \text{reactor} \]
\[ \varepsilon_{(0)} = \text{axial solids hold-up at height 0, m}^3 \text{solid/m}^3 \text{reactor} \]
\[ \varepsilon_{z} = \text{axial solids hold-up at height z, m}^3 \text{solid/m}^3 \text{reactor} \]
\[ \varepsilon_{z} = \text{radial averaged solids hold-up at height z, m}^3 \text{solid/m}^3 \text{reactor} \]
\( \varepsilon_{\text{kin}} \) - specific kinetic energy conversion rate (equals the specific kinetic power dissipation), W/kg

\( \varepsilon_t \) - specific turbulent energy conversion rate (equals the specific turbulent power dissipation), W/kg

\( \eta \) - dynamic viscosity, kg/(m.s)

\( \rho \) - density , kg/m\(^3\)

\( \tau_p \) - particle response time, s

\( \tau_t \) - characteristic fluctuation time, s

\( \nu \) - kinematic viscosity, m\(^2\)/s

**Super- and subscripts**

- \( f \) = fluid phase
- \( \text{gas} \) = gas phase
- \( l \) = liquid phase
- \( o \) = atmospheric conditions
- \( p \) = particle
- \( s \) = solid phase
- \( sl \) = slurry
- \( \text{tot} \) = total present in the system
- \( x, y, z \) = specific directions
- \( \infty \) = in infinite medium
LITERATURE

Canada G.S., Mclaughlin M.H., Staub F.W., "Flow regimes and void fraction distribution in gas fluidization of large particles in bed without tube banks", AIChE Symposium Series, 74, 14, 1978
Scale-up of an air agitated tapered suspension reactor

R.H. Kleijntjens, R.J.G.M. van der Lans, K.Ch.A.M. Luyben
Kluyver Laboratory for Biotechnology, Delft University of Technology
Julianalaan 67, 2628 BC Delft, The Netherlands

ABSTRACT

Suspending sand particles ($\rho_s = 2500$ kg/m$^3$) in water by means of air agitation requires considerable amounts of energy. Scale-up criteria, predicting the minimal power input per volume ($P/V$), are therefore indispensable for the sound scale-up of (s-l-g) slurry reactors. For the scale-up of a tapered reactor, the suspension efficiency ($\eta_{\text{sus}}$), being the ratio of the power required to sustain the solid phase (d$E_{\text{part}}$/dt) and the power input ($P_{\text{in}}$), is shown to be a valid criterion. Experiments determining the numeric value of $\eta_{\text{sus}}$ were carried out at two reactor scales (0.4 and 4.0 m$^3$) using slurry densities of 20 and 40 wt%. A monodisperse solid phase was used with a diameter of 260 µm. On the basis of a constant axial profile and the established value of $\eta_{\text{sus}}$ ($\approx 0.2$), the power input at a large scale (40 m$^3$) is predicted to be $P/V = 122$ W/m$^3$. The $P/V$-scale effect is quantified as $P/V \cdot V^{-0.13}$.

Keywords

Scale-up, reactor, particle suspension, specific power input, suspension efficiency
INTRODUCTION

In the development of multiphase reactors, often the stage of scale-up is difficult because of the lack of design and scale-up criteria. This lack partly is due to the complexity of multiphase systems and partly to the enormous diversity of solid, liquid and gas phases which can be combined (Shah, 1979; Schugerl, 1989). It is therefore important to recognize scale-up bottlenecks in an early stage, since it is then possible to focus on these aspects both from a theoretical point of view as well as from the practical side.

If scale-up theory is developed, it will generally not be derived straight for the complex practical system, but focus first on a simplified model system. In such a model system, one or more of the phases are replaced by well defined model phases. In the theoretical model, the rate limiting phenomena in kinetics, mass or heat transport are described. Subsequently, model parameters are determined from experiments carried out at laboratory and pilot scale (Rose, 1981). Finally, scale-up is achieved by means of the extrapolation of model parameters.

In the development of a biotechnological slurry process for the reclamation of excavated polluted soils, a new type of tapered suspension reactor was designed with soil as the solid phase (Kleijntjens et al., 1990). To scale-up the three phase (soil-water-air) slurry reactor properly, it was considered necessary to study first the power input as a function of scale and slurry density for a model system. In this model system, the complex, not well defined, polydisperse solid phase (soil) is replaced by monodisperse sand. In this paper, a combination of suspension experiments carried out at two different scales (0.4 and 4.0 m³), and a mechanistic model is described. A mechanistic scale-up relation and the scale effect in P/V will be presented as result.

To provide a background for the mechanistic suspension model, first the scale-up of the two phase (s-l) mechanically stirred tank reactor is discussed. Secondly, the scale-up of slurry bubble columns is considered.

Scale up of mechanically stirred tank reactors

In the industrial practice of slurry processing, two phase (solid-liquid) mechanically stirred tank reactors are frequently used. Although the
technology of these mixing operations has advanced tremendously in the last decades, an uniform scale-up procedure still does not exist. As a consequence, a choice between several contradictory rules of thumb has to be made. (Jordan, 1968). To consider the consequences of the different criteria, four criteria will be discussed in terms of scale effect in the specific power input (Harnby et al., 1986)

* constant specific power input (P/V)
* constant Reynolds number
* constant Froude number
* experimental scaling rules (Zwietering, 1958)

In the comparison made, unaerated monodisperse suspensions are considered. It is assumed that the scale-up is carried out under the condition of geometrical similarity.

The first criterion, prescribing a constant P/V is considered as a reference, since it incorporates no net scale-effects. This means that identical results are achieved, at every scale, for the same amount of specific power input. To express the criteria prescribing a constant Reynolds and Froude number in terms of P/V, the derivation given in the equations 1 to 8 is considered. First, the definitions of both numbers are given

\[ \text{Re} = \frac{\rho L * v * D_i}{\eta_1} \quad \text{Fr} = \frac{v^2}{D_i * g} \]  \[ [1] \]

In the given definitions of the Reynolds and Froude number the impeller diameter is taken as the characteristic length in the stirred vessel. To eliminate v, the product of stirring speed and impeller size is customary regarded \((v = N * D_i)\)

\[ \text{Re} = \frac{\rho L * N * D_i^2}{\eta_1} \quad \text{Fr} = \frac{N^2 * D_i}{g} \]  \[ [2] \]

Assuming constant liquid properties and using the expressions given above, the criteria for constant Re and Fr read as
\[ N \sim D_i^{-2} \quad \quad N \sim D_i^{-0.5} \quad [3] \]

To translate these criteria into the specific power input, the impeller diameter \( D_i \) is taken proportional to the reactor diameter \( T \), and the reactor volume is proportional to \( T^3 \) (standard stirred vessel). Following these relationships, and the general power input equation for mechanically stirred vessels (Beek and Muttall, 1972) the following derivation may be given

\[ \frac{P}{V} = \frac{N^3 \cdot D_i^5}{V_r} - \frac{N^3 \cdot D_i^5}{T^3} - \frac{N^3 \cdot D_i^5}{D_i^3} - \frac{N^3 \cdot D_i^2}{D_i} \quad [4] \]

With the relations given in 3 and 4, the criteria of constant Reynolds and Froude number respectively read as

\[ \frac{P}{V} \sim D_i^{-4} \quad \quad \frac{P}{V} \sim D_i^{-0.5} \quad [5] \]

Using the proportionality between \( D_i \) and \( T \), and rewriting the reactor diameter \( T \) as \( V^{1/3} \) the two criteria finally read as

\[ \frac{P}{V} \sim V_r^{-1.33} \quad \quad \frac{P}{V} \sim V_r^{0.166} \quad [6] \]

In stirred suspension reactors, the criterion of a constant \( \text{Re} \) obviously can be achieved at decreasing \( P/V \). The criterion of a constant Froude number can only be achieved at the expense of an increasing specific power input at larger scale. In figure 1 the two curves for constant Reynolds and Froude number are depicted.

The fourth scale-up criterion was derived empirically. From suspension experiments, the power input necessary to achieve a state of minimal suspension, was determined at several scales (Zwietering, 1958). Combining these experimental results with a dimensional analysis, the following relation was derived for the stirrer speed at minimal suspension

\[ N_{ms} = S_{sys} \cdot \nu^{0.1} \cdot d_p^{0.2} \cdot (g \cdot \Delta p / p_l)^{0.45} \cdot \epsilon_{(tot)}^{0.13} \cdot D_i^{-0.85} \quad [7] \]
Using the correlation between specific power input, stirrer speed and impeller diameter as given in equation 4, the trend in $P/V$ according to equation 7 reads as

$$ P/V_{ms} = D_i^{-0.55} - T^{-0.55} - \nu^{-0.18} \quad [8] $$

This scale-up correlation differs from the other scale-up criteria as is clearly shown in figure 1. If, for instance, a scale factor 100 is considered, the predicted $P/V$ according to equation 8 is about half of the $P/V$ predicted by the reference criterion assuming a constant $P/V$. For scale-up in practice, the consequence of these effects in terms of equipment, power consumption, heat production and thus costs can be large. For the mechanically stirred suspension reactor, although being a known system, obviously no clear rules of thumb are available.

![Figure 1: Scale effects in the minimal power input for two phase (s-l) mechanically stirred tank reactors.](image)

Scale-up of slurry bubble columns

Having replaced soil by monodisperse sand, the tapered soil slurry reactor can also be seen in the perspective of three phase (s-l-g) slurry bubble columns. However, the vast literature on bubble columns makes it impossible
to cover all aspects from these systems. From reviews it is clear that the behavior of the solid phase in bubble columns is mostly studied by means of the sedimentation-dispersion model ((Murray and Fan, 1989; Jean et al., 1989; Deckwer, 1985). This model however, gives no relationship between power input, liquid motion and solid hold-up. Scale-up criteria can not be generated by the sedimentation-dispersion model. Its use is mainly restricted to the derivation of the particle dispersion coefficient from experimental distribution data (Kato et al, 1973).

From the empirical scale-up of two phase (1-g) bubble columns, dimensional correlations were derived predicting the dispersion coefficient from the column diameter and the liquid circulation velocity (Walter and Blanche, 1983; Joshi, 1980; De Lasas, 1986). Nevertheless, similar design correlations for the particle dispersion in three phase bubble columns are rare. Studies regarding the critical superficial gas velocity, necessary to sustain a particle suspension, have resulted in various design relations for which however, "a significant disagreement exists for the presented correlations" (Fan, 1989).

MECHANISTIC SUSPENSION MODEL

Turbulent particle suspension

To achieve a mechanistic description of the particle suspension hydrodynamics, the use of turbulence has been emphasized by several authors (Baldi et al., 1978; Joshi, 1983)). In understanding turbulent suspension, the fluid-particle interaction is regarded as crucial, since it is the fluid motion which supports the solid phase (Narayanan et al., 1969). Two conditions can be formulated which have to be fulfilled in order to establish a turbulent particle suspension

* each particle must be able to follow the turbulent liquid motion (Hinze, 1972)
* the turbulence must be non-decaying and sufficient in intensity to replenish the continuous energy dissipation of suspended particles (Narayanan et al., 1969; Soo, 1956)
The first condition stresses that a particle can be kept in turbulent suspension unless it is not able to follow the turbulent liquid motion. To capture this condition in a single parameter, a particle response number was defined in previous work. Experiments, confirming the theory, have shown that at pilot scale (4.0 m³), sand particles with a diameter larger than 700 μm are unable to follow the turbulent flow. Above this diameter, the particles settled (Kleijntjens et al., 1991-A).

The major difference between the first and the second condition, is the fact that the first statement is a necessary condition but not a sufficient one. The second, energetic, condition is both necessary and sufficient. Being the most complete, the second condition will be used in the mechanistic suspension model.

According to the second condition, liquid motion has to replenish the energy which is lost by the particles due to settling. If particle suspension is seen as a dynamic process in which settling particles are lifted by the turbulent liquid motion, the origin of the viscous dissipation is found in the loss of potential energy of the particles. To quantify the amount of work which is done by the liquid motion at the solid phase, the loss of potential energy for one particle is defined

\[
\frac{dE_{\text{part}}^{\text{pot}}}{dt} = g \Delta \rho \frac{V}{p} u_p^\infty
\]  

[9]

In this definition the rate at which a particle loses its energy is determined by its relative mass and its terminal settling velocity. In a suspension of particles however, the actual settling velocity at a specific location is determined by hindrances of the surrounding particles. Therefore a correlation from literature is used which relates the settling velocity in a cloud of particles to the solid hold-up. For the system discussed the correlation proposed by Barnea and Mirzahi (1973), covering a wide range of experimental results, was considered appropriate

\[
u_p = u_p^\infty [\frac{1 - \epsilon_s}{1 + \epsilon_s^{1/3}}] \exp[-\frac{5 \epsilon_s}{3(1 - \epsilon_s)}]
\]

[10]
Regarding a particle suspension with a three dimensional spatial hold-up distribution \( \epsilon_s(x,y^*,z^*) \), the summation of all the individual energy losses provides the total amount of viscous dissipation per time

\[
\frac{d\epsilon_{\text{Part}}}{dt}_{\text{tot}} = g \Delta \rho \int_0^V \epsilon_s(x,y^*,z^*) \, dV_r
\]  

[11]

In agitated systems however, not only a particle suspension has to be sustained, also turbulent motion itself must be maintained. Incoming power thus is divided between two processes (Kleijntjens et al., 1991-B)

\[
P_{\text{in}} = \frac{d\epsilon_{\text{Part}}}{dt}_{\text{tot}} + \epsilon_t \, V_r \, \rho_{\text{sl}}
\]  

[12]

In equation 12 the generated turbulence is assumed to be isotropic, therefore it is allowed to use the specific turbulent energy conversion rate \( \epsilon_t \) (Hinze, 1959).

In order to develop a scale-up criterion, the interaction between turbulence and the solid phase is considered in more detail. In a particle suspension work is done by the "turbulent fluctuating velocity components directed opposite to gravity" (Hetseroni, 1982). Under isotropic conditions however, it is impossible that the turbulence generated in the system results in upward directed velocity fluctuations only. In each direction particles are transported. Although being executed in every direction, only work in the vertical upward direction will be effective. For this reason, the "suspension efficiency" can be defined as the ratio of the power effectively used to sustain the particles, and the total amount of power introduced into the system

\[
\eta_{\text{sus}} = \frac{\left[ \frac{d\epsilon_{\text{Part}}}{dt} \right]_{\text{tot}}}{P_{\text{in}}}
\]  

[13]

Taking \( P_{\text{in}} \) equal to the amount of turbulent power produced, and the loss in potential energy equal to the buoyancy force, \( \eta_{\text{sus}} \) is similar to the flux Richardson number. This number is defined as the ratio between the rate of removal of energy by buoyancy forces and the rate of turbulent energy production due to shear (Turner, 1973). From its definition the flux
Richardson number cannot exceed unity. In experiments, carried out in a meteorological context, a value for the flux Richardson number of about 0.2 has been established. This implies that 20% of the power is efficiently used, meaning that most of the energy is dissipated by turbulent viscosity and only a fraction by working against gravity (Turner, 1973; Tennekes and Lumley, 1972).

If \( \eta_{\text{sus}} \), in line with the \( \text{Ri}_f \) number, is a constant for turbulent suspension reactors, it can be used as a scale-up criterion linking the power input directly to the solid hold-up distribution. To investigate the hypothesis of a scale independent \( \eta_{\text{sus}} \), experiments were carried out in the tapered reactor at two scales of 0.4 and 4.0 m\(^3\) and for two different different slurry densities (0.088 and 0.210 v/v).

**SPATIAL HOLD-UP DISTRIBUTION IN THE TAPERED SUSPENSION REACTOR**

In order to determine \( \eta_{\text{sus}} \), both the power input and the rate of loss of solid phase potential energy have to be derived. To establish the latter, the integral given in equation 11 has to be solved. For this purpose, first the flow pattern and the spatial solid hold-up distribution in the tapered reactor are studied.

As indicated in figure 2, gas injected at the bottom of the tapered reactor results in the upward and downward directed bulk flow in the y,z plane. Since the reactor developed has a relative small depth (figure 3), liquid motion in the x-direction is restricted compared to motion in the y,z plane. It therefore is assumed that solid hold-up gradients in the x-direction are negligible. As a result a two dimensional hold-up distribution, \( \epsilon_s(y^*,z^*) \), is considered. Using the total depth \( D \) as parameter in the x-direction, equation 11 therefore can be rewritten,

\[
\frac{dE_{\text{part}}}{dt}_{\text{tot}} = g^* \Delta \rho \Delta z \int_0^H \int_{-Y(z)}^{Y(z)} \epsilon_s(y^*,z^*) dy^* dz^* \]

\( Y(z) \) is the maximum radial distance defined from the centre of the tapered reactor at height \( z^* \).
From equation 14 and the experimentally established two dimensional solid hold-up distribution, the loss of the potential energy of the solid phase can be determined. However, because hindered settling must be incorporated, a complex integral results after the substitution of $u_p(y^*,z^*)$. To reduce this complexity, the inner integral of equation 14 is solved by means of defining the radial averaged axial solid hold-up

$$\varepsilon_s(z) = \frac{1}{2Y(z)} \int_{-Y(z)}^{Y(z)} \varepsilon_s(y^*,z^*) \, dy^*$$

[15]

In equation 15, the radial hold-up distribution at a height $z^*$ is integrated over the total width of the reactor (from $-Y(z)$ to $Y(z)$) and subsequently divided by that width. In this way, a set of radial data points is converted into one parameter, $\varepsilon_s(z)$, representing the average and located at the central axis of the system. From this point on, the dimensionless height $z$ is used as axial parameter, defined as $z = z^*/H$, as dimensionless radial
distance \( y = y^*/Y(z) \) is used. After substitution of \( Y(z) \varepsilon_s(y^*, z^*)dy^* \) equation 14 reads as

\[
\frac{dE_{\text{part}}}{dt}_{\text{tot}} = g \Delta \rho D H \int_0^1 (u_p(z) - \varepsilon_s(z) * 2 Y(z)) dz \tag{16}
\]

Since in equation 16 the two dimensional distribution is replaced by the axial distribution, also the hindered settling velocity (equation 10) depends on the axial parameter only. If the reactor shape is approximated as being completely tapered, with an angle of 60°, \( Y(z) \) may be written as \( \tan 30^\circ \times H \times z \); the reactor volume as \( V_r = \tan 30^\circ \times D \times H^2 \). Substitution of \( Y(z) \) in equation 16 results in

\[
\frac{dE_{\text{part}}}{dt}_{\text{tot}} / V_r = 2g \Delta \rho \int_0^1 (u_p(z) - \varepsilon_s(z) * z) dz \tag{17}
\]

Combining the definition of the suspension efficiency according to equation 13 and equation 17, the specific power input reads as

\[
P/V = \frac{1}{\eta_{\text{sus}}} * 2g \Delta \rho \int_0^1 (u_p(z) - \varepsilon_s(z) * z) dz \tag{18}
\]

With equation 18 a direct relation between the behavior of the solid phase, expressed as dimensionless profile, and the specific power input is established. From the measured radial averaged axial profiles and the specific power input, the suspension efficiency can be empirically derived.

**EXPERIMENTAL EQUIPMENT AND METHODS**

Experiments were carried out in a three phase tapered s-l-g gas agitated reactor with an angle of the tapered part equal to 60° and a height over width ratio equal to unity (Kleijntjens et al., 1991-A).
In figure 3 a representation of the experimental set-up is shown, in the H/T ratio the maximum height at the centre, and the maximum width are regarded.

![Diagram of experimental set-up](image)

**Figure 3** Experimental set-up of the tapered reactor.

Particle suspension was studied at laboratory scale (0.4 m³) and at pilot scale (4.0 m³). Both reactors were designed according to the principle of geometrical similarity, with the 4 m³ reactor having the height, width and depth a factor $10^{1/3}$ larger than at the 0.4 m³ scale. The slurry height at the smaller scale is 1.27 m and 2.70 m at the pilot scale. The reactor depth D was 0.45 m at the 0.4 m³ scale and 1 meter at the 4.0 m³ scale.

The suspension behavior was studied at both scales for a slurry density of 20 and 40 wt% (respectively 0.088 and 0.210 v/v). In order to obtain a suspension, gas is introduced at the lower end of the reactor by means of an injection manifold with equidistant injection points. The injection points were designed as vertical tubes with an inner diameter of 2 mm and a mutual distance of 2 cm. The gas flow is measured at the manifold with a rotameter. The pressure registered was used to express the gas flow for atmospheric conditions (Coleman, 1956). The ratio of the total injection area, given by the sum of the tube openings, and the cross sectional area of the manifold
pipe, was chosen equal to 3 in order to circumvent maldistribution (Dow and Shreveport, 1950).

The standardized gas flow is used in the work equation for rising bubbles in order to determine the power input in the system (Lamont, 1958)

\[ P_{\text{Gas}}^{\text{in}} = p^0 \cdot Q_{\text{Gas}}^0 \cdot \ln \left( \frac{\rho_{\text{sl}} \cdot g \cdot H}{p^0} \right) \]  \hspace{1cm} [19]

In equation 19 the average slurry density is used, defined as

\[ \rho_{\text{sl}} = (1 - \epsilon_s^{(\text{tot})}) \cdot \rho_1 + \epsilon_s^{(\text{tot})} \cdot \rho_s \]  \hspace{1cm} [20]

Suspension experiments were carried out with monodisperse quartz sand, having a diameter equal to 260 (+/- 30) \( \mu \)m and a density of 2500 kg/m\(^3\). Experiments were conducted under the condition of minimal power input, since this critical input gives the only really comparable hydrodynamic state at different scales and different solid hold-ups. To establish the minimum power input an excess of gas (air) is injected to create a suspension. The critical minimal input is then determined by a stepwise decrease in gas input. The state of minimal suspension is detected by visual observation and, in line with the Zwietering criterion (Zwietering, 1958), is defined as that state in which a dense collar of slow slipping sand appeared just above the manifold. With this method the critical gas input could be reproduced within 5%.

The axial and radial particle hold-up distributions were determined at the state of minimal suspension with a sampling probe (figure 4). To measure the hold-up firstly the open probe is brought to the desired location, then, after several seconds, the probe is closed and withdrawn from the slurry. Subsequently, the probe contents is analysed for its dry mass. From the dry mass, the sampling volume of the probe (9.42 ml), using the densities of the solid and the liquid phase, the volume fraction of solids can be calculated. At each location three samples were taken within a period of several minutes. The locations were chosen at regular intervals giving a complete image of the distribution in the \( y,z \) plane of the tapered reactor. The standard deviations in the calculated solid contents varied between 5 and 30\% of the mean value. This variation was a result of turbulent fluctuations.
RESULTS AND DISCUSSION

Power input and solid hold-up distributions

In table 1 the measured minimal power input and corresponding specific power input \((P/V)\) is given. The table shows that at larger scale less specific power input is required to sustain a state of minimal suspension. Furthermore, it can be seen that a larger specific minimal power input is necessary at higher slurry densities.
Table 1 Measured minimal power input and minimal specific power input.

<table>
<thead>
<tr>
<th>( (tot) )</th>
<th>( V = 0.4 , m^3 )</th>
<th>( V = 4.0 , m^3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_s )</td>
<td>P</td>
<td>P</td>
</tr>
<tr>
<td>in</td>
<td>W</td>
<td>W</td>
</tr>
<tr>
<td>( v/v )</td>
<td>( v/v )</td>
<td>( W/m^3 )</td>
</tr>
<tr>
<td>0.088</td>
<td>50.8</td>
<td>374.4</td>
</tr>
<tr>
<td>0.210</td>
<td>87.3</td>
<td>610.0</td>
</tr>
<tr>
<td>( v/v )</td>
<td>( v/v )</td>
<td>( W/m^3 )</td>
</tr>
<tr>
<td>0.088</td>
<td>127</td>
<td>94</td>
</tr>
<tr>
<td>0.210</td>
<td>218</td>
<td>152</td>
</tr>
</tbody>
</table>

The axial solid hold-up distributions at the central axis of the reactor \((x=0, y=0)\) measured during minimal suspension, are depicted in figure 5.

**Figure 5** Measured axial solids hold-up distribution at the axis.
The data for a slurry density of 0.088 v/v show that the axial profiles at both scales are rather similar in their S-shape. The two profiles show for the lower half of the reactor, up to a dimensionless height of 0.4-0.5, a rather homogeneous distribution. In the upper reactor zone, the distribution shows an exponential decrease with height. At a slurry density of 0.210 v/v the axial profiles also are similar at both scales although they have lost their S-shape. The homogeneous zone at this density extends to a height in the reactor of z equal to 0.7-0.8, in the top part of the reactor almost a linear decrease in solid hold-up was measured.

The radial profiles are depicted in the figure 6. On the vertical axis the ratio of the local solid hold-up at position \( \epsilon_s(y,z) \) and the hold-up at the axis of the system \( \epsilon_s(0,z) \) is given for several heights in the reactor. On the horizontal axis the radial distance from the centre is given, as the ratio of the actual distance to the centre (y) and the maximal radial distance at that specific height, Y(z). The tapered reactor shape can be recognized if the points at the right side of each profile are connected.

The dimensionless representation of \( \epsilon_s(y,z)/\epsilon_s(0,z) \) demonstrates clearly that the shape of the radial profiles is height dependent. In the homogeneous bottom zone (z=0.25), radial gradients are far less pronounced than at larger heights. As is shown, the trend in the radial profiles is rather independent of scale. However, since in the upper reactor part the radial distribution at a slurry density of 0.210 v/v is more pronounced than at a slurry density of 0.088 v/v, the trend in the radial profiles is dependent on the solid hold-up.
Figure 6  Measured radial solids hold-up distributions.
The experimental suspension efficiency

To determine $\eta_{\text{sus}}$ from the experiments, the measured specific power input was combined with the solid hold-up distribution curves. Using equation 15 to average the hold-up data, and substituting $u_p(z)$ according to equation 10, equations 17 and 18 were solved for each of the four experiments. In table 2 the results of the numerical integrations are given. Shown are the rate of loss of the potential energy, the rate of specific energy loss (put between brackets) and the corresponding suspension efficiencies. At one specific solid hold-up, a constant rate of specific loss of potential energy was found. This was due to the assumed similarity of the profiles measured at 0.4 and 4.0 m³ scale (figures 5 and 6).

The experimental $\eta_{\text{sus}}$ (table 2) is somewhat lower than the efficiency derived under ideal conditions. This means that in a reactor turbulence counteracts gravity effects less efficient compared to turbulence under meteorological conditions. It should however be noted that at pilot scale (4.0 m³) for a slurry density of 0.088 v/v, $\eta_{\text{sus}}$ equals 0.19, a value which comes close to the expected value.

Table 2 Calculated potential energy loss rates of the solid phase, specific rates and suspension efficiencies.

<table>
<thead>
<tr>
<th>$\epsilon_{\text{tot}}$</th>
<th>$V = 0.4$ m³</th>
<th>$V = 4.0$ m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>$v/v$</td>
<td>$W$</td>
<td>$W$</td>
</tr>
<tr>
<td></td>
<td>$W/m^3$</td>
<td>$W/m^3$</td>
</tr>
<tr>
<td>0.088</td>
<td>6.9 (17.3)</td>
<td>72.3 (18.1)</td>
</tr>
<tr>
<td>0.210</td>
<td>9.5 (23.7)</td>
<td>96.9 (24.4)</td>
</tr>
<tr>
<td>$v/v$</td>
<td>$\eta_{\text{sus}}$</td>
<td>$\eta_{\text{sus}}$</td>
</tr>
<tr>
<td>0.088</td>
<td>0.14</td>
<td>0.19</td>
</tr>
<tr>
<td>0.210</td>
<td>0.12</td>
<td>0.16</td>
</tr>
</tbody>
</table>
Considering the trends in table 2, it seems likely that the counteraction of gravity at large scale, with minor wall effects, can proceed under circumstances similar to those under meteorological conditions. It is concluded that at large scale $\eta_{\text{sus}}$ equals 0.2.

The decreasing suspension efficiency at higher slurry density is in line with expectations. From previous experiments in the same reactor, it was derived that free particle dispersion is impeded at higher solid hold-up (Kleijntjens et al. 1991-B). At higher slurry density particle interactions obviously decrease the efficiency of suspension.

CONCLUSIONS

From equation 18 the mechanistic scale-up relation can be derived as

$$\frac{P/V}{1.\text{s.}} = \frac{1}{\eta_{\text{sus}}} \times 2g*\Delta \rho \times \int_{0}^{1}(u_{p}(z) * \varepsilon_{s}(z) * z)_{r.s.} \, dz \quad [21]$$

The P/V at large scale is predicted from the product of $1/\eta_{\text{sus}}$ and the dimensionless radial averaged axial hold-up profile at the reference scale (r.s). As reference scale, the largest scale at which suspension experiments were carried out can be taken. The use of equation 21 as scale-up relation implies two assumptions

* the dimensionless hold-up distribution at large scale is equal to that at the reference scale (constant specific power loss)

* the suspension efficiency ($\eta_{\text{sus}}$) at large scale is equal to 0.2

Using these conditions and the results from table 2, the P/V at large scale is determined as 88.5 and 122 W/m$^3$ for the slurry densities of 0.088 v/v and 0.210 v/v respectively. These predictions are irrespective of the size of the large scale, the reactor volume is not used as individual parameter in equation 21. If these predicted values are compared to the measured values (table 1), a definite scale effect in the P/V was established. This scale effect in P/V was mainly due to the scale dependency of $\eta_{\text{sus}}$ (table 2). The specific power loss was scale independent due to the similarity of the dimensionless profiles at 0.4 and 4.0 m$^3$. 
To express the scale-up according to equation 21 in terms of P/V, the exponential scale factor is determined (note equation 8). For this purpose, first the large scale volume is specified. Considering a scale factor 10 appropriate for this type of slurry operation (Peters and Timmerhaus, 1980), 40 m³ is taken for the large scale. The three scales (0.4, 4.0 and 40 m³) now are considered, together with their P/V values. For \( \varepsilon_s \text{(tot)} = 0.088 \text{ v/v} \) these values are 127 W/m³, 94 W/m³ and 88 W/m³. For \( \varepsilon_s \text{(tot)} = 0.210 \) the P/V values are 218 W/m³, 152 W/m³ and 122 W/m³ for the three volumina respectively. Using a double logarithmic plot (note figure 1), regression analysis provides the scale exponent as -0.08 and -0.13 for the 0.088 v/v and 0.210 v/v slurry density respectively. The exponent of -0.13 comes close to the scale effect predicted by the Zwietering correlation for stirred vessels (equation 8).

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LIST OF SYMBOLS

\[ D = \text{depth of tapered reactor (in direction x), m} \]
\[ D_1 = \text{impeller diameter, m} \]
\[ d = \text{particle diameter, m} \]
\[ E_1 = \text{dispersion coefficient for liquid phase, m}^2/\text{s} \]
\[ E_s = \text{dispersion coefficient for solid phase, m}^2/\text{s} \]
\[ \frac{dE_{\text{part}}}{dt} = \text{rate of loss of potential energy of the particles in suspension, kg.m}^2/\text{s}^3 \text{ or W} \]
\[ g = \text{gravitational constant, 9.8 m/s}^2 \]
\[ G = \text{gas phase} \]
\[ H = \text{reactor height, m} \]
\[ L = \text{liquid phase} \]
\[ l_e = \text{length of energy containing eddy, m} \]
\[ N = \text{impeller speed, s}^{-1} \]
**N**<sub>ms</sub> = impeller speed at condition of minimal suspension, \( \text{s}^{-1} \)

\( p \) = pressure, kg/m.s²

\( \text{Pe}_p \) = dimensionless Peclet number for the solid phase

\( \text{Pin} \) = incoming power, W

\( \text{Pin}_{\text{gas}} \) = power input due to gas agitation, W

\( \frac{\text{P}}{\text{V}} \) = specific power input, W/m³

\( \frac{\text{P}}{\text{V}}_{\text{ms}} \) = specific power input at minimal suspension, W/m³

\( Q^0_{\text{gas}} \) = standardized volumetric gas flow, m³/s

\( S \) = solid phase

\( S_{\text{sys}} \) = system constant

\( t \) = time, s

\( T \) = reactor width in the tapered reactor (in direction y), m

\( u_p \) = settling velocity, m/s

\( u_{p}(x,y,z) \) = local settling velocity at position \( x,y,z \), m/s

\( u_{p}(y,z) \) = local settling velocity in the plane \( y,z \), m/s

\( u_{p}(z) \) = local settling velocity at height \( z \), m/s

\( u' \) = turbulent fluctuation velocity (defined as the root mean square of the instantaneous fluctuation velocities), m/s

\( v \) = characteristic bulk liquid velocity, m/s

\( V \) = volume, m³

\( Y(z) \) = maximum radial distance from at height \( z \), defined from the centre of the reactor, m

\( y^* \) = radial distance in the reactor, m

\( y \) = dimensionless radial distance in the reactor, \( y^*/Y(z) \)

\( z^* \) = axial distance in the reactor, m

\( z \) = dimensionless reactor height, \( z^*/H \)

---

**Greek symbols**

\( \epsilon_s \) = solid hold-up, m³ solids/m³ reactor

\( \epsilon_{(\text{tot})} \) = total solid hold-up present in the reactor, m³ solid/m³ reactor

\( \epsilon_{s}(x,y,z) \) = local solid hold-up at position \( x,y,z \), m³ solid/m³ reactor

\( \epsilon_{s}(y,z) \) = local solid hold-up in the plane \( y,z \), m³ solid/m³ reactor

\( \epsilon_{s}(z) \) = radial averaged solid hold-up at height \( z \), m³ solid/m³ reactor

\( \epsilon_t \) = specific turbulent energy conversion rate, W/kg

\( \eta \) = dynamic viscosity, kg/(m.s)

\( \eta_{\text{sus}} \) = efficiency of suspension, -
\rho \quad - \text{density, } \text{kg/m}^3
\Delta \rho \quad - \text{density difference between solid and liquid phase, } \text{kg/m}^3
\nu \quad - \text{kinematic viscosity, } \text{m}^2/\text{s}

Super- and subscripts

f - fluid phase
l - liquid phase
l.s - large scale
o - atmospheric conditions
p - particle
r - reactor
s - solid phase
p.s - pilot scale
sl - slurry
x,y,z - specific directions
\infty - in infinite medium
Literature

Three phase slurry reactor for soil processing

Part 1 Design

R.H. Kleijntjens, R.G.J.M. van der Lans, K.Ch.A.M. Luyben
Kluivver Laboratory for Biotechnology, Delft University of Technology
Julianalaan 67, 2628 BC Delft

ABSTRACT

For the efficient treatment of excavated polluted soils (particle sizes in between $1 \mu m$ and $4000 \mu m$) in a slurry process, the coarse non-polluted soil fraction has to be removed at the beginning of the process. For this purpose a tapered integrated slurry reactor/seperator was developed: the Dual Injected Turbulent Separation (DITS-) reactor. In reasonable agreement with the theoretically derived particle response numbers for the turbulent suspension behavior of soil, a separation between the fines and the coarse fraction could be achieved in the DITS-reactor. At pilot scale (4.0 m$^3$), the fines ($<700 \mu m$) are suspended in a gas agitated bulk compartment, while the (settled) coarse particles are fluidized in the bottom compartment. By way of independent slurry withdrawal from bottom and bulk, particle separation is achieved.

Keywords

Reactor design, slurry, particle suspension, response number, soil
INTRODUCTION

Over the last decade, soil pollution has been recognized as a serious environmental problem. New remedial action technologies are therefore developed and tested in practice (Wolf et al., 1988). Among the various techniques under development are biological reclamation techniques (Soczo and Staps, 1988). Major advantages of these processes, are the mineralization of the (organic) pollutant into harmless compounds like biomass and carbon dioxide, and the quality of the soil after treatment. To obtain maximum profit of the opportunities microbial soil decontamination offers, a biotechnological slurry process for the aerobic treatment of excavated polluted soils was developed (Kleijntjens et al., 1990).

In this continuous slurry process, polluted soils are treated in three phase (soil-water-air) suspension reactors. Experiments carried out in a miniplant have shown that a soil residence time in the order of days is needed to obtain the required decontamination level (Kleijntjens et al., 1989). Regarding this residence time and the fact that the solid hold-up in suspension reactors normally does not exceed 0.2-0.3 m³/m³ (Nienow, 1968; Hallett, 1981), a reasonable process capacity can only be achieved by the use of large reactors. Considering a slurry process with a capacity appropriate in soil processing (10 tons per hour), a total reactor volume of several thousands cubic meters is necessary.

Preliminary cost estimates have shown that an economic large scale slurry process largely depends on the specific power input (P/V). This should preferable be minimized to values as low as 50-100 W/m³. At this low power input, not only particle suspension must be achieved, but also oxygen must be provided for the aerobic microbial decontamination. In search for multiphase reactors, suitable for this purpose, air agitated slurry reactors were considered most appropriate. Among this class of reactors, the air agitated suspension reactor with a tapered bottom offers the best possibilities to achieve particle suspension, mixing and oxygen transfer at a low power input (Bos et al, 1988).

Such a reactor, called Pachuca reactor, was orginally developed for mineral ore leaching. It is a cylindric tank (H/T = 2-3) with a conical bottom
(Merriman, 1958). The agitation of the slurry is caused by the air introduced at the apex of the cone. Due to the air agitation, the Pachuca tank differs profoundly from a mechanically stirred vessel. In the latter, power is transferred from the impeller to the tank contents, while in the Pachuca reactor the power is transferred as the air expands in rising through the vessel (Lamont, 1958). It was reported that Pachuca tanks at large scale (200-300 m³) can be operated with a power input per unit of volume (P/V) as low as 45 W/m³ (Hallet, 1981). Due to this moderate power input, a moderate oxygen transfer rate in Pachuca systems is achieved (a factor 10-100 lower than an aerated stirred vessel) (Joosten, 1977). Since the microbial oxygen demand for soil processing was shown to be moderate, this aspect does not influence the benefits of the Pachuca concept (Kleijntjens et al., 1989). Practical benefits of air agitated slurry tanks are the low construction and maintenance costs in combination with the absence of corrosion sensitive parts.

From previous work on air agitated suspension reactors (mainly bubble columns), it is known that turbulence is the driving force behind particle suspension (Narayanan, 1969). It was stated that, to keep a solid phase in suspension, "the turbulence must be non-decaying and the viscous dissipation of the particles must be replenished by the turbulence" (Soo, 1956). According to this condition, slurry motion has to replenish the energy which is lost by the particles due to settling. If particle suspension is seen as a dynamic process in which settling particles are lifted by the turbulent motion, the origin of the viscous dissipation is found in the loss of potential energy of the particles. To quantify the amount of work required to keep particles in suspension, the following general expression has been derived (Kleijntjens et al, 1991b)

\[
P_{\text{in}} = \frac{1}{\eta_{\text{sus}}} \int_V \left[ g \Delta \rho \mathbf{u} \cdot \mathbf{e} \epsilon_s(x,y,z) \right] dV_r \tag{1}
\]

In this equation, the power input, \( P_{\text{in}} \), is determined by the product of the total amount of solids kept in suspension, \( \int_V \epsilon_s(x,y,z) dV_r \), and the rate at which the solids settle. Due to hindrance effects, this settling rate can be written as a function of the terminal settling velocity and the spatial
solid hold-up distribution: \( u^\infty_p f(\epsilon_s(x,y,z)) \) (Barnea and Mirzahi, 1973). The efficiency of suspension, \( \eta_{sus} \), gives the ratio between the amount of power used to sustain the solids and the power required to maintain the turbulent motion. Assuming \( \eta_{sus} \) to be a constant, the power input needed for suspension is determined by

\[
* \text{total solids volume, } \int_0^V \epsilon_s(x,y,z) \, dV_r
* \text{particle size, by way of } u^\infty_p
\]

Concerning these two parameters, only the latter can be used for optimization purposes since in most slurry processes (including the soil process) the solid hold-up is maximized for capacity reasons. Process optimization (power minimization) therefore focuses on the second parameter, the particle size. In mining industries, the ores are grained in a pretreatment section to the appropriate diameter, mostly \( d_p <100 \mu\text{m} \) (Hallet, 1981). In the processing of excavated polluted soils grinding is not an appropriate pretreatment (for cost reasons), therefore the heterogeneous, polydisperse character of soil has to be dealt with.

**Figure 1** Composition of the soil used as feed in the process.
In figure 1, a typical particle size distribution of soil, to be treated in such a slurry process is depicted. Although the densities of the soil fractions are somewhat different (≈2100 kg/m³ for clay and ≈2500 kg/m³ for sand and gravel), soil is considered to have a density of 2500 kg/m³ in this paper (worst case approach). The upper limit \( d_p = 4 \text{ mm} \) is determined by the mesh of the finest sieve used in the preprocessing of excavated soil, the lower limit is due to the smallest clay and humic particles \( d_p < 1 \mu\text{m} \) that are present. As shown in figure 1, the soil has significant contributions of each fraction in between 0 and 4000 \( \mu\text{m} \).

Due to different chemical/physical properties of each fraction (Brady, 1984), the pollutant is not equally distributed over the different fractions. From previous work (Kleijn et al., 1989) and related literature (Werther and Wilichowski, 1990) it was derived that more than 90 \% of the pollutant is preferentially adsorbed to the soil fraction smaller than 100 \( \mu\text{m} \). The inhomogeneous distribution of the pollutant offers the possibility to lower the power input needed in the reactor by removing the coarse fraction. Since the coarse fraction contains a small amount of the pollution, the overall decontamination level is hardly affected by the separation.

Aim of the research presented in this paper, is the development of a turbulent slurry reactor for soil processing. As starting point of the development a Pachuca-like reactor was used. In the reactor the biological treatment of the fines is combined with the separation of the coarse fraction. An integrated reactor/seperator is aimed for to keep the process as simple as possible (minimization of unit operations by integration). In the integrated reactor, the coarse fraction is treated (washed) before it leaves the system, the pollution present in the coarse material is thus removed. In the development of the integrated reactor, two stages are distinguished.

1) In this paper (part 1), the suspension behavior of a polydisperse solid phase in a turbulent flow is theoretically described by means of a particle response number. Based on this description, a criterion is developed for the design of the reactor/seperator. Experiments have been carried out to investigate the suspension
and separation performance of the integrated system at pilot scale (4.0 m³). In addition, the geometry (height/width ratio) of the reactor was optimized by means of model experiments.

2) In part 2 (Kleijntjens et al., 1991-B), the scale-up of the separator/reactor is treated. By means of the extrapolation of model results and experimental data (derived at two scales: 0.4 and 4.0 m³), the power input at a large scale is predicted.

THEORETICAL INVESTIGATION OF TURBULENT PARTICLE SUSPENSION

In turbulent suspension the fluid-particle interaction is regarded as crucial, since it is the fluid motion which supports the solid phase (Hetseroni, 1982; Davis 1986). Two conditions with respect to the suspension can be formulated

1) each particle must be able to follow the turbulent liquid motion (Hinze, 1972)

2) the turbulence must be non-decaying and sufficient in intensity to replenish the continuous energy dissipation of suspended particles (Narayanan et al., 1969; Soo, 1956)

The motion of particles in turbulent flows has been studied intensively in the last decades (Hetseroni, 1982; Squires and Eaton, 1990). In turbulent flows, particle suspension is due to the turbulent liquid motion which exerts a drag force on the particle, a force which frequently changes in direction. Therefore, the interaction between fluid and particle will only result in a complete suspension if the particle is able to follow the liquid motion to a large extent. Being a crucial aspect of turbulent suspension reactors, the mobility of a particle in a turbulent flow is taken as starting point for the design of the slurry reactor/separator.

Particle response number
An indication of the ability to follow turbulent motion is found in the ratio of a particle response time and a characteristic time for turbulent flow. This ratio (denoted as the particle response number), facilitates the hydrodynamic description of turbulent particle suspension since the two mechanisms dominating the interaction between fluid and particle are captured in one parameter. In theoretical studies focusing on particle behavior under turbulent conditions, the particle response number is frequently used as an inertia parameter in computer simulations (Squires and Eaton, 1990; Fung and Perkins, 1989).

The particle response time can be calculated from a force balance over a discrete particle in a turbulent flow according to Newton's law of motion (Hinze, 1972)

\[
\left( \rho_p \dot{V} + \beta \rho_f \dot{V} \right) \cdot \frac{d}{dt} \left( u_p - u_{fi} \right) - \Sigma F_i = 0
\]  

Equation 2 expresses the change in time of the particle velocity relative to the bulk velocity \( u_p - u_{fi} \). In the acceleration, both the particle mass and the virtual mass of the displaced liquid are considered. The term \( \Sigma F \) contains the flow resistance, gravity, pressure forces, rotational forces and other forces acting on the particle. From these forces, only the flow resistance (drag force) is taken into account, being the most important term.

Following Hinze (1972), the particles are assumed to react according to the creeping flow (Stokes) regime. This gives a drag coefficient of \( C_d = 24/Re_p \).

Using this expression the drag force becomes

\[
F_{drag} = 24/Re_p \cdot 0.25 \cdot \pi \cdot d_p^2 \cdot 0.5 \cdot \rho_f \cdot (u_p - u_{fi})^2
\]  

If this drag force is substituted for \( \Sigma F_i \) in the right hand side of equation 2, the equation can be written in a differential form having the relative Reynolds particle number as a variable. The solution of this equation relates the change in the relative particle Reynolds number to a specific time interval (Hinze 1972). Taking a time interval in which the Reynolds particle number changes by a factor 2, the following relation results for the response time \( \tau_p \)
\[ \tau_p = 0.04 \times \frac{(\rho_p + \beta \rho_f) \times d_p^2}{\eta_f} \]  \[4\]

In equation 4, \( \eta_f \) is the fluid dynamic viscosity. The term between brackets refers to the amount of mass considered in the response time, \( \rho_p \) accounts for the particle and \( \beta \rho_f \) for the virtual added mass (amount of fluid displaced as the particle moves). The particle response time can be seen as a measure for the particle inertia in a liquid flow.

It must be noted that only particles smaller than 100 \( \mu m \) are in the, above assumed, creeping flow regime. In view of figure 1, this means that Hinze's derivation is only valid for the fines in the soil. To derive the response number for the soil particles outside the creeping flow regime, the standard correlation curve for \( C_d - Re_p \) is used to correct the results (Bird et al., 1960).

For the definition of a characteristic time of turbulence, the rate determining process in the turbulent power conversion process must be taken into account. Two parameters specifying a class of eddies at the top of the turbulent energy spectrum are considered, the length of the energy containing eddy in the system, \( l_e \), and its turbulent intensity, \( u' \). In an isotropic turbulent flow, the following equation for the kinetic energy transfer rate per unit of mass (\( \epsilon_t \)) holds

\[ \epsilon_t = \frac{u'^3}{l_e} \]  \[5\]

Equation 5 is regarded as "the first law of turbulence" governing the turbulent power conversion (Batchelor, 1953). The two crucial parameters, \( u' \) and \( l_e \), are also considered in many other turbulent processes such as the eddy viscosity (Hinze 1959). Based on these two parameters, the characteristic time for the rate of change in a turbulent flow can be estimated as

\[ \tau_t = \frac{l_e}{u'} \]  \[6\]
Equation 6 is derived from dimensional analysis (Hinze, 1972). In order to estimate values for \( r_t \), it is necessary to substitute \( u' \) and \( l_e \) by measurable reactor parameters. For this purpose, the ratio between the eddy size and the characteristic length of the system (\( T \)) is considered. From previous work (Kleijnens et al., 1991a), it was derived that in the suspension reactor, the eddy length is both a function of scale and solid hold-up. The eddy length varied from \( l_e = 0.15* T \) (lab-scale) to \( l_e = 0.06* T \) (4.0 m³ reactor) for a slurry density of 10 wt%. For a slurry density of 40 wt%, the ratio \( l_e/T \) varied from 0.05 to 0.017 (4.0 m³). Based on these results, it was considered appropriate to vary \( l_e \) with scale. Using an average slurry density of 20 wt%, \( l_e/T \) was taken equal to 0.08 at small scale and 0.03 at pilot and at a large scale.

To substitute \( u' \), the specific turbulent energy transfer rate, \( \epsilon_t \), is approximated by the power input per volume \((P/V)\) divided by the fluid density. Equation 5 therefore reads as

\[
\frac{(P/V)}{\epsilon_f} = \frac{u'^3}{l_e} \tag{7}
\]

In the derivation of the characteristic turbulent time, for the small reactor scales the \( l_e/T \) ratio of 0.08 was used. At larger scale, a value of 0.03 for the ratio \( l_e/T \) was used. The general expression for the turbulent time now reads as a combination of equations 6 and 7

\[
r_t = \alpha \left( \frac{T^2 \ast \rho_f}{P/V} \right)^{1/3} \tag{8}
\]

In equation 8, \( \alpha \) is a function of the reactor scale. Combining equations 4 and 8, the final expression for the response number now reads as \((\gamma \text{ is a function of scale and particle size})\)

\[
r_p/r_t = \gamma \left( \frac{\rho_p + \beta \rho_f}{\eta_f \ast \rho_f^{1/3}} \right) \ast \left( \frac{d_p^2 \ast P/V}{T^2/3} \right)^{1/3} \tag{9}
\]

In equation 9, the response number is split into two parts. The first part contains the slurry properties. In the second part parameters such as reactor size, particle size and specific power input are present.
The significance of $r_p/r_t$ for the soil reactor design is demonstrated by model calculations showing the number as a function of the particle diameter. The following parameter values were used: $\rho_p = 2500$ kg/m$^3$ (sand), $\rho_l = 1000$ kg/m$^3$ (water), $\beta = 0.66$ (Hinze, 1972) and $\eta_r = 0.001$ Pa.s. The response numbers have been calculated for two small scale reactors, having $T$ equal to 0.01 and 0.1 m, and two large scale reactors with $T$ equal to 1.0 and 5.0 m. The eddy length was determined as $l_o = 0.08T$ (smallest scales) and $l_o = 0.03T$ (larger scale). Since only low power inputs are of interest, calculations were carried out for a specific power input of 50 W/m$^3$. Scale effects (normally P/V is larger for smaller scales) were not taken into account. The resulting response numbers are plotted as a function of particle size in figure 2.

![Diagram](image)

**Figure 2** Theoretical curves of the particle response number.

For values of $r_p/r_t = 1$, the particle reaction time is equal to the characteristic turbulent time. In line with the statement made by Hinze (1972) that "$r_p$ must clearly be smaller than $r_t$ to obtain a high degree of particle diffusion", the criterion for the turbulent suspension regime is
taken as \( \tau_p/\tau_t < 0.1 \). For these values, the mobility of the particle is large enough to allow for a fast adjustment to changes in the turbulent flow. For values of \( \tau_p/\tau_t > 0.1 \), the inertia of the particle is not negligible, meaning that the mobility of the particle and the turbulent flow are not identical. It therefore is concluded that for practical slurry flows, the settling regime corresponds to \( \tau_p/\tau_t > 0.1 \).

The model calculations clearly show the influence of the particle diameter and reactor scale on the response number. At small reactors scales, the change of regime takes place at a particle diameter of 150-400 \( \mu \text{m} \). At larger scales the regime change can be expected for particles with a diameter of 900-5000 \( \mu \text{m} \). It may therefore be concluded that even at large scale, the coarse soil fraction can most likely not be kept in suspension (settling regime).

Finally, it is important to note that in the particle response number, the mobility aspect of one particle in suspension is expressed. Although it is not a bulk parameter such as \( P/V \), the response number is a useful design parameter since it reveals that the coarse soil fraction is in the settling regime under turbulent conditions. Based on this result, a new concept for a soil slurry reactor is introduced.

**DUAL INJECTED TURBULENT SEPARATION REACTOR**

As demonstrated above, turbulent suspension can be used as tool for the discrimination between different soil fractions. Combining turbulence as separation mechanism and the low operating power of the Pachuca reactor, a new type of slurry reactor was developed. In this reactor, coarse soil material is separated from the fines without disturbing the aerobic decontamination of the latter.

A reactor was designed in which the driving force behind the separation, turbulence, is integrated with the efficient removal of settled material from the reactor bottom. Simultaneous injection of air and slurry at the bottom of a tapered vessel was adopted for this purpose. In the Dual Injected Turbulent Separation (DITS-) reactor, two compartments can be distinguish (figure 3).
* a zone with fluidized coarse material at the bottom
* an air-agitated three-phase suspension of fine material in the bulk of the reactor

![Diagram of the DITS-reactor](image)

**Figure 3** Schematic representation of the DITS-reactor.

Because of the tapered shape, the upward superficial slurry velocity rapidly decreases at increasing height. Therefore fluidization only is achieved for the settled particles at the bottom. Without disturbing the turbulent character of the gas agitated bulk, fluidization is carried out at the point where it is mostly needed. Due to the fluidization of the settled particles, blockage of the injector is prevented (air and slurry injection proceeds without difficulty). Having established two compartments, slurry can be withdrawn from these compartments independently. In this way, the coarse fraction can continuously be separated from the fines. The latter are for further treatment transported from the DITS-reactor to a cascade of slurry reactors. Since settlement problems are not to be expected, the second stage is equipped with air agitation only. In figure 4 a simplified flowsheet is shown of the integration of the DITS-reactor and cascade.
Considering the large scale of the air agitated cascade reactors (several thousands cubic meters), a series of large conical tanks with a considerable height does not seem optimal. Therefore a tapered "channel-like" cascade reactor was designed with a relative low height/width ratio. Air is injected by way of a line of injection manifolds at the bottom, covering the length of the reactor. Line injection was also considered beneficial for the DITS-reactor. Having distributed the fluids before injection, a more homogeneous dissipation can be expected.

![Diagram of the slurry process](image)

**Figure 4** Simplified flowsheet of the slurry process.

The DITS-reactor, as shown schematically in figure 4, is designed as a tapered shaped unit-operation in which separation and microbial processing are integrated. It becomes clear from the flowsheet that the performance of the DITS-reactor largely determines the power input needed in the second stage. An efficient removal of coarse material minimizes the power input for the cascade of air agitated reactors.
EXPERIMENTAL

Two sets of experiments will be described below. First, optimization of the height/width ratio of the tapered reactor, which was carried out by means of suspension experiments in a model system. Secondly, the design and separation performance of the DITS-reactor at pilot scale (4.0 m³) is discussed.

1) Optimization of the reactor geometry (model system)

In order to determine the optimal reactor geometry, suspension experiments were carried out in a gas agitated flat scale model of a tapered channel (figure 5). A flat scale model with a small depth was considered appropriate since the major bulk flow takes place in the vertical reactor plane (Kleijntjens et al, 1991-A). To be able to study suspension phenomena in an unambiguous manner a model solid phase (quartz sand, \( d_p = 260 \) μm and \( \rho_s = 2460 \) kg/m³) was used.

![Figure 5 Experimental set-up of the air agitated reactor.](image-url)
In order to investigate the suspension behavior at different geometrical configurations and volumes, the reactor was equipped with two movable vertical plates (figure 5). Different geometries could be created by varying the slurry height (H) in combination with the reactor width (T). The depth, D, of the reactor was fixed in each experiment and equal to the length of the injection groove (0.45 meter). Gas was introduced at the bottom by means of an injection manifold with equidistant injection points made of vertical tubes with an inner diameter of 2 mm and a mutual distance of 2 cm. In order to circumvent maldistribution in the manifold, the cross sectional area of the manifold was chosen 3 times larger than the total injection surface area, as given by the sum of the injection openings (Dow and Shreveport, 1950).

The optimal geometry was considered to be the geometry requiring the lowest power input to maintain a state of minimal suspension. The state of minimal suspension was detected by visual observation of the (mono-disperse) particles near the reactor bottom. Minimal suspension was defined as the state of suspension in which a collar of slow slipping sand appeared just above the injection manifold. The critical gas input was determined by a stepwise decrease in gas input, starting from complete suspension. Using this method the standard deviation in the detected critical gas input was about 5 %.

The gas flow was measured at the manifold with a rotameter. The pressure registered in the rotameter was used to express the gas flow for atmospheric conditions (Coleman, 1956). Together with the slurry height and density, the gas flow is used in the work equation for rising bubbles in order to calculate the power input in the system (Lamont, 1958).

\[
p_{\text{in}}^{\text{gas}} = p^o \cdot \phi_{\text{gas}} \cdot \ln \left[ \frac{p^o + \rho_{\text{sl}} \cdot \text{g} \cdot H}{p^o} \right]
\]  \[10\]

It can be shown that the kinetic contribution \( \phi_v \cdot \rho \cdot \text{v}^2 \) is negligible (Lamont, 1958). Pressure losses in pipes and the injector itself are not considered, only the power input present in the gas phase at the moment of entrance is accounted for in equation 10.
To determine the optimal reactor geometry, particle suspension in a 10 wt% (0.041 v/v) slurry was studied at four different volumes, varying from 0.040 m³ up to 0.160 m³. Within each volume four H/T ratios (1, 2, 3 and 4) were tested.

Results

For the sixteen experiments carried out to determine the best geometry, the specific power input is given in figure 6. The figure shows that at H/T=1 the specific power input is minimal for each volume. This geometry corresponds with the trends in the response numbers (table 1). In this table the reactor characteristics are given in combination with the specific power input and the response number. The numbers are calculated according to equation 9. For all the geometries examined, the maximum reactor width is used as the characteristic length.

Figure 6 Measured specific power input (P/V) in the gas agitated model system.
At higher H/T ratios, larger values for the particle response number are found. At these higher ratios, the smaller characteristic reactor lengths induce the presence of smaller eddies \((l_e \approx 0.08T)\), resulting in a state in which particles follow the turbulence less easily. H/T = 1 obviously approximates the optimal geometry since the emerging flow pattern allows for the existence of larger eddies compared to other geometries. For the same reasons (larger eddies), particle suspension proceeds more efficiently at a larger scale.

Table 1 Data used in the optimization study to determine the particle response number.

<table>
<thead>
<tr>
<th>Reactor volume (m³)</th>
<th>H/T</th>
<th>T (m)</th>
<th>(l_e) (m)</th>
<th>Power input (W/m³)</th>
<th>(t_p/t_c) (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.04</td>
<td>1</td>
<td>0.40</td>
<td>0.040</td>
<td>179</td>
<td>0.05</td>
</tr>
<tr>
<td>0.04</td>
<td>2</td>
<td>0.24</td>
<td>0.024</td>
<td>210</td>
<td>0.08</td>
</tr>
<tr>
<td>0.04</td>
<td>3</td>
<td>0.19</td>
<td>0.019</td>
<td>269</td>
<td>0.10</td>
</tr>
<tr>
<td>0.04</td>
<td>4</td>
<td>0.16</td>
<td>0.016</td>
<td>283</td>
<td>0.12</td>
</tr>
<tr>
<td>0.08</td>
<td>1</td>
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<td>0.056</td>
<td>151</td>
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<tr>
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<td>0.34</td>
<td>0.034</td>
<td>206</td>
<td>0.06</td>
</tr>
<tr>
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<td>3</td>
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<td>0.026</td>
<td>217</td>
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</tr>
<tr>
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<td>0.022</td>
<td>259</td>
<td>0.09</td>
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<tr>
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<td>0.069</td>
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<td>0.032</td>
<td>200</td>
<td>0.06</td>
</tr>
<tr>
<td>0.12</td>
<td>4</td>
<td>0.27</td>
<td>0.027</td>
<td>217</td>
<td>0.08</td>
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<tr>
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<td>0.79</td>
<td>0.079</td>
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<tr>
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<td>0.037</td>
<td>179</td>
<td>0.06</td>
</tr>
<tr>
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<td>4</td>
<td>0.32</td>
<td>0.032</td>
<td>203</td>
<td>0.07</td>
</tr>
</tbody>
</table>

In Table 1 it is shown that \(r_p/r_t < 0.1\) for all experiments, except two. These values of the response number show that, although \(r_p/r_t < 0.1\) is regarded as transition criterion, in practice the transition takes place at values somewhat lower.
2) Design of the DITS-reactor

To test the principle of the DITS-reactor and its separation performance, a 4.0 m³ pilot plant was designed according to the optimal geometry (H/T=1). The slurry height and width in the pilot plant were 2.7 meter, the reactor depth was 1 m. To operate the air-slurry injected reactor, two devices were added to the experimental set-up shown in figure 5

* liquid/solid separator in the slurry recirculation
* dual (g-l) injection manifold

Treatment of the slurry circulation flow is neccessary since direct slurry injection (as indicated in figure 4), will block the dual injection manifold.

2a) Design of the liquid/solid separator in the recirculation slurry loop

The design of the liquid/solid separator is based on theoretical estimations and trial and error. Two aspects were crucial

* required circulation flow rate
* size of the particles to be separated

First, an estimation of the circulation flow is based on the minimal upward slurry velocity necessary to fluidize the coarse soil fraction (estimated diameter about 2 mm) at the deepest point of the tapered reactor. For the upward slurry velocity equal to the settling velocity (0.18 m/s) (Perry and Chilton, 1973), and a cross sectional surface, just above the manifold, of 0.01 m * 1 m, the injection flow rate needs to be 110 l/min. Estimating a settler efficiency of about 50 % (ratio of the slurry injected and the slurry withdrawn from the top), a slurry flow of about 220 l/min has to be treated in the settling unit.
Second, the size of the particles which are to be removed from the slurry before injection is considered. For this purpose, it was empirically established that soil particles above 100 μm cause blockage in the injector and therefore must be removed.
Settlers and hydrocyclones were considered as possible solid/liquid separators. For the relative small flow rate needed, a hydrocyclone would not be practical because of the small diameter. In addition, the operating pressure of such a hydrocyclone (several bar) is a hindrance (energy costs). An upward plug flow settler was chosen as the most appropriate system. In this settler an upward velocity smaller than the settling velocity of the 100 μm fraction must be realized.

To estimate the minimal settler volume, the characteristic settling time of the 100 μm fraction is determined from the settler height (about 1 meter) and the settling velocity (about 0.01 m/s). The characteristic settling time \( r_{set} = H_{set}/u_p \) was determined as 100 seconds. Regarding the plug flow character of the settler, the total volume is 0.360 m³ \( V_{set} = r_{set} \Phi_V \).
In addition to the removal of soil particles, measures for the removal of organic soil material appeared to be necessary, since fibrous floating material also blocked the injector. The use of a sieve was considered appropriate for this purpose.

The final shape of the liquid/solid separator was determined by the fact that it had to be placed on top of the pilot reactor. This settler position was chosen in order to return the settled fraction (which would not yet be completely decontaminated) to the reactor under gravitational force. The final design was a small and deep, two compartment, settler (height 1.2 m) with a total volume of 0.530 m³. Three sections can be recognized, shown schematically in figure 7:

1) Vertical sieve plate (mesh 1 mm, length 0.7 m, width 0.4 m, angle with the vertical 5 °), removing floating fibrous and organic material; the removed material is directed back to the reactor.

2) Inertia settler, this section is designed as a pretreatment of the upflow section, removing the heavier sand fraction; the underflow of this section directly opens to the reactor; the volume of this tapered section is 0.160 m³.

3) Upflow settler removing the remaining soil particles with a diameter above 100 μm; the underflow is also directed to the reactor; the volume of this section was 0.370 m³.

The slurry flow in the liquid/solid separator, is schematically depicted in figure 7. Slurry from the reactor is pumped to the top (a) of the vertical sieve plate (1). Fibrous material, removed by the sieve (1 mm) is returned to the reactor (b), while the sieved slurry flows continuously to the inertia settler (c). In this settler (2), the larger particles are removed by way of flow d, while the fines are further transported (e) to a second settler. In (3) particles not sustained by the flow will settle and leave the system (flow f). To maximize the settler efficiency, both flows d and f are minimized. The top product of (3) is pumped straight to the slurry injector (g). In figure 8 a three dimensional representation of the settler section and the DITS-reactor is given.
2b) Injector design

To be able to inject air and recycled slurry simultaneously at the bottom of the tapered reactor, a special injection manifold was developed. In the manifold, slurry is distributed among vertical injection points in a similar fashion as described for the gas injector of the 0.4 m³ (figure 5). Major problems with slurry injection were particle settlement and maldistribution. The latter was circumvented by choosing the cross sectional area 3 times larger than the total injection surface area (Dow and Shreveport, 1950). The slurry manifold was designed for a slurry feed in which all particles above 100 μm are removed.
The gas-liquid injector was developed with 40 equidistant injection points, each constructed as an inner air jet, surrounded by outer slurry jets. (Luyben and Kleijntjens, 1988). The amount of power introduced due to slurry injection is discussed in part 2 (Kleijntjens et al. 1991b)

Experimental strategy

To investigate the separation performance of the DITS-reactor, experiments were carried out at a scale of 4.0 m³. Since only high slurry densities were considered of interest for the process design, the reactor performance at 30 and 40 wt% soil slurries was investigated. First, it was empirically determined which combination of gas and slurry injection flows resulted in the simultaneous existence of a fluidized bed at the reactor bottom, and a turbulent suspension of fines in the bulk. Having achieved two compartments, the gas and slurry injection flow rates were further minimized to obtain a maximum size bottom compartment (visual observation). Subsequently, samples were taken from the bed and the bulk. Particle size distributions were established by wet sieving of the slurry samples, using a set of sieves with decreasing mesh (see figure 9).

RESULTS AND DISCUSSION

Performance of the DITS-reactor, pilot-scale

Three items concerning the reactor performance were studied

*) the injection flow rates and the injector performance
*) power input due to slurry and gas injection
*) separation performance
*) the solid/liquid separator

The gas and slurry injection flows, necessary to sustain the 30 wt % and 40 wt % soil slurries, were 0.47 and 0.43 m³/min respectively for the gas, and 0.14 and 0.15 m³/min for the slurry. The measured slurry injection flows
were somewhat higher than the estimated value of 0.11 m³/min used in the design of the equipment.

From the measured flow rates, the power input was established. For 30 wt%, the power input due to the gas phase (see equation 10) was 191 W and for the slurry input 14 W (see part 2). Taking the gas input as responsible for the turbulent agitation in the bulk, the specific power input (P/V) for this compartment is 48 W/m³. For the 40 wt% slurry for the gas and slurry input respectively 206 W and 20 W were measured, resulting in a P/V bulk compartment equal to 51 W/m³.

Besides the flow rates, a major parameter relevant to the performance of the injector was the pressure, measured at the manifold (see figure 5). Both the gas and slurry manifolds were monitored separately. The gas injector was operated at a pressure of about 0.4 bar. To inject the slurry, a pressure build-up of about 0.7 bar was required. Taking the pressure build-up due to the slurry height in the reactor as 0.3 bar, the pressure loss in the gas injector was about 0.1 bar. In the slurry injector, the pressure loss was about 0.4 bar. Since the pressure in the slurry manifold itself did not increase during operation, both the removal of the particles and the fibrous materials was in accordance with requirements. If the reactor is operated without a sieve section, pressure increased within 2 hours up to 3-5 bar, followed by complete blockage of the manifold.

To investigate the separation capacity with respect to the coarse fraction in the 4.0 m³ pilot plant, from the bottom and the bulk compartment slurry samples were taken and analyzed for their particle size distribution. In figure 9 the distributions of the bottom and bulk compartment for the 30 wt% and 40 wt% slurry densities are depicted. For both slurry densities, the distribution in the bulk samples differs from the distribution in the bed samples. The coarse fractions 7 (710-1000 µm) and 8 (1-4 mm) are absent in the bulk samples.
Figure 9 Measured particle size distributions in the bottom and bulk compartment of the 4 m³ DITS-reactor.

To determine the settler efficiency, the slurry flows pumped to the settler are considered (see point a in figure 7). For the 30 and 40 wt% slurry these
flows were 0.273 and 0.402 m³/min respectively. The settler efficiencies (injected flow rate/withdrawal flow rate) varied from 0.51 to 0.37, coming close to the estimated value of 0.5 used in the design of the equipment. With regard to the removal of the solids, the settling performance was as expected; no particles above 100 μm were detected in the overflow at point g (figure 7). The latter demonstrates that the upflow pattern in the second settler suited its purpose.

Experimental suspension criterion

To compare the experimental suspension behavior in the pilot-plant with the theoretical prediction, the results from figure 9 are compared with the curves given in figure 2. This comparison can be made since the experimental P/V (48 and 51 W/m³) is close to the P/V used in the model calculations (50 W/m³).

To calculate the theoretical regime change (r_p / r_t < 0.1) for the pilot-plant, first the characteristic length of the tapered reactor has to be established. Based on the flow pattern in the tapered system (Kleijntjens et al., 1991a) the characteristic length of the pilot plant is taken equal to the width at half height (about 1 meter for the 4 m³ pilot-plant). Using a length of 1 meter, the regime change in the pilot-plant is predicted for d_p = 1000 μm (see the curve for the 1 meter reactor scale in figure 2). This prediction is in reasonable agreement with the experiments showing the regime change to take place at d_p = 700 μm (figure 9). Regarding the difference between the theoretical and the measured transition particle size, it should be recognized that the theory was derived for one single particle in a turbulent flow, while experiments were carried out for dense slurries.

An additional support for the statement that the regime change (suspended-settling) can be expected at values of the response number of about 0.1 is found in literature (Squires and Eaton; 1990). In theoretical simulations a response number equal to 0.075 was used as lower limit.

CONCLUSIONS
From the theoretical and experimental results derived in this paper the following can be concluded

* Due to negligible amount of pollution adsorbed to the coarse soil fraction, it is beneficial to separate this fraction from the polluted fines \((d < 100 \ \mu m)\) at the beginning of the slurry process.

* Integrated separation and soil processing can be achieved in a tapered Dual Injected Turbulent Slurry (DITS-) reactor. Pilot scale experiments \((4.0 \ m^3)\) showed that, for a 30 and 40 wt% slurry density the coarse soil fraction \((d_p > 700 \ \mu m)\) can be separated from the fines.

* Assuming that "a high degree of particle diffusion can only be obtained if the particle response time is smaller than the turbulence time" (Hinze, 1972), the theoretical regime transition from turbulent suspension to settling is taken at \(\tau_p/\tau_t \approx 0.1\). This criterion is confirmed by the experiments carried out in the 4 m\(^3\) DITS-reactor.

* For a 40 wt% slurry density, the measured power input for slurry and gas in the DITS-reactor were 20 W and 206 W respectively. The latter results in the economically attractive low P/V for the reactor bulk of \(= 50 \ W/m^3\).

* As optimal geometry for the tapered reactors used in the soil slurry process, H/T-1 was found.

Acknowledgement

The authors are grateful to prof. F.T.M. Nieuwstadt for his theoretical support, and to Michiel Bosse, Mark de Groot, Hilda van de Laar, Rene Langevoort and Yvonne van Voorst for their cooperation in the project. The authors also are grateful to G. Van der Toolen and P. Vetter for their indispensable technical assistance.
LIST OF SYMBOLS

\( d \) = diameter, m
\( E_{\text{pot}} \) = potential energy of the solids in suspension, Joule (kg.m\(^2\)/s\(^2\))
\( F \) = force, N
\( g \) = gravitational constant, 9.8 m/s\(^2\)
\( H \) = reactor height, m
\( H_{\text{set}} \) = height of the settler, m
\( l_e \) = length of energy containing eddy, m
\( p \) = pressure, Pa
\( P \) = power, W
\( P_{\text{in}} \) = power input, W
\( \frac{F}{V} \) = specific power input, W/m\(^3\)
\( \text{Re}_p \) = relative Reynolds particle number \( d \cdot \rho_f \cdot (u_{\text{pi}} - u_{\text{f}})/\eta_f \)
\( t \) = time, s
\( T \) = maximal width of tapered reactor, m
\( u \) = velocity, m/s
\( u' \) = turbulent liquid fluctuation velocity, m/s
\( V_R \) = reactor volume, m\(^3\)
\( x \) = coordinate in the depth of the reactor, m
\( y \) = coordinate in the width of the reactor, m
\( z \) = coordinate in axial direction, reactor height, m

Greek symbols

\( \alpha \) = scale dependent coefficient
\( \beta \) = virtual coefficient
\( \gamma \) = scale and particle size dependent coefficient
\( \epsilon_{\text{xyz}} \) = local solid hold-up, m\(^3\) solids/m\(^3\) reactor
\( \epsilon_t \) = turbulent kinetic energy transfer rate, W/kg
\( \eta \) = dynamic viscosity, Pa.s
\( \eta_{\text{sus}} \) = suspension efficiency
\( \phi \) = volumetric flow rate, m\(^3\)/s
\( \rho \) = density, kg/m\(^3\)
\( \Delta \rho \) = density difference between solid and liquid phase, kg/m\(^3\)
\( \tau_p \) = particle response time, s
\[ \tau_t = \text{characteristic turbulent fluctuation time, s} \]
\[ \tau = \text{residence time, s} \]

Super- and subscripts

drag = drag force action at particle

e = eddy

f = fluid phase, water

G = gas phase

gas = gas phase, air

i = specific direction

inj = injector

L,1 = liquid phase

o = atmospheric conditions

p = particle

S = solid phase

set = settler

sl = slurry

\( \infty \) = in infinite medium, used for terminal settling velocity

LITERATURE


Three phase slurry reactor for soil processing

Part 2 Scale-up

R.H. Kleijntjens, R.G.J.M. van der Lans, K.CH.A.M. Luyben
Kluyver Laboratory for Biotechnology, Delft University of Technology
Julianalaan 67, 2628 BC Delft

ABSTRACT

To decontaminate soil efficiently in a slurry process, the coarse particle fraction is treated and separated in a newly developed reactor type, the so-called Dual Injected Turbulent Separation (DITS) reactor. Since the feasibility of the process largely depends on this reactor, its scale-up is studied on the basis of a model and experiments, carried out at two scales (0.4 and 4.0 m$^3$). It was found that the DITS-reactor can be described as a two compartment system. Coarse particles (particle diameter larger than 700 µm) are fluidized in the bottom zone as a result of liquid injection, while fines are suspended in the bulk. For scale up, the specific power input per unit of volume (P/V) in both compartments was determined. It is predicted that for a slurry density of 40 wt%, and a scale of 40 m$^3$ the P/V in the bottom compartment is around 1000 W/m$^3$. For the bulk compartment a P/V of about 50 W/m$^3$ is needed.

Keywords Slurry reactor, soil, scale-up, pilot-plant, power input
INTRODUCTION

In process development the stage of scale-up is often a difficult one. Not only technological problems are met in this stage, but also logistical, financial and planning issues have to be dealt with (Van de Berg and De Jong, 1980; Peters and Timmerhaus, 1981). For these reasons the choice of an appropriate pathway is important to achieve successful scale-up (Rose, 1981; Johnstone and Thring, 1957; Hudson, 1967).

The most straightforward scale-up methodology is the empirical "trial and error" approach (Cooper et al., 1965). In this practical approach, by means of a stepwise scale enlargement, the full scale design is achieved. Although this strategy is money consuming, in industry this method is often used since it is fast, the equipment can be tested and the product (produced in mini or pilot-plants) is available in an early stage for marketing and other purposes (Conn, 1971).

In many cases however, the trial and error approach should be avoided. For example, in the development of new environmental remediation technologies, where "new" types of multiphase reactors are used originating from industrial applications (Schugerl, 1989; Shah, 1979). If sufficient theoretical support is available, a more mechanistic scale-up can be carried out. Using this approach, the rate limiting phenomena in kinetics, mass- or heat transport are mathematically modelled. Experiments at laboratory scale and pilot scale are carried out to establish the scale dependency of model parameters. Scale-up is achieved by means of the extrapolation of trends observed in the model parameters and in the experimental data (Berty, 1979; Rose, 1981; Constan, 1984).

Advantages of the mechanistic approach, are found in the deeper understanding of the mechanisms leading to greater confidence, which in turn allows greater scale-up factors to be used (Johnstone and Thring, 1957; Rose, 1981). Moreover, due to the insight in the mechanisms the operation can be optimized more easily, and contrary to the trial and error approach, it is possible to develop flexible scale-up criteria which can be used for similar systems.

Scale-up is also a crucial step in the development of an economic biotechnological slurry process for the decontamination of excavated polluted soils (Kleijntjens et al., 1990). Design and scale-up problems in the slurry process were not found in the pre- and after treatment of the
soil, but in the slurry reactor that forms the core of the process. In this case, the so-called DIIT-reactor needed special attention. In the Dual Injected Turbulent Separation reactor, the crucial separation and treatment of the coarse soil fraction (particle diameter larger than 700 μm) is achieved (see part 1, Kleijnjens et al., 1991b). After separation, the fine, polluted, soil fractions are further treated. The feasibility of the slurry process largely depends on the performance of this DIIT-reactor. Therefore, research focussed on the suspension behavior of the solid phase (polydisperse soil with a particle diameter between 1 and 4000 μm, see figure 1), and the power input needed for suspension.

In this paper, scale-up of the DIIT-reactor is described in four steps

* description of reactor system
* mechanistic suspension model
* experimental work and results
* prediction of the power input at large scale

![Graph showing particle size distribution](image)

**Figure 1** Typical composition of the soil used as reactor feed.
DESCRIPTION OF THE DITS-REACTOR

Considering the particle size distribution of the soil (figure 1), the following can be noted. The upper limit of the distribution \( d_p = 4 \text{ mm} \) is determined by the mesh of the finest sieve used in the preprocessing of the excavated soil. The lower limit \( d_p < 1 \mu m \) is formed by the smallest clay and humic particles present. To get an indication of the suspension behavior of these fractions, the suspension hydrodynamics were investigated. In line with the results presented in part 1, the soil was considered to consist of three hydrodynamical classes:

1) a fine soil fraction \( (1 \mu m < d_p < 300 \mu m) \), this fraction is completely in suspension \( (\tau_p/\tau_t < 0.01) \)

2) a mid-size soil fraction \( (300 \mu m < d_p < 700 \mu m) \), critical with respect to suspension \( (0.01 < \tau_p/\tau_t < 0.1) \)

3) a coarse soil fraction \( (700 \mu m < d_p < 4000 \mu m) \), this fraction is in the settling regime \( (\tau_p/\tau_t > 0.1) \)

In figure 2 a schematic representation of the DITS-reactor is given. Two compartments are indicated, namely:

* a slow slipping, fluidized bed compartment at the reactor bottom, containing the, settling, coarse fraction.
* a turbulent air agitated bulk compartment containing the fine and the mid-size fraction.

Due to this stratification, a separate removal of the coarse fraction from the DITS-reactor is possible by means of slurry withdrawal from the reactor bottom.
MECHANISTIC SUSPENSION MODEL

To provide a theoretical framework for the scale-up of the DITS-reactor, a mechanistic suspension model is derived. The model derived in this article, is meant to give a first impression of scale effects, rather than to provide a fully accurate description of the complex suspension behavior of the solid phase at the various scales.

As basis for the suspension model, the difference in (turbulent) suspension behavior of the soil fractions is taken. As discussed above, the polydisperse soil can be divided into three hydrodynamical classes. Since the hold-up distribution in slurry systems is also characterized by the suspension behavior of the solids (Kato et al., 1972), there will be a relationship between the suspension behavior and the axial solid hold-up profile. The suspension model used for scale-up is based on power dissipation of the solid phase (Kleijntjens et al., 1991c). Since this model uses the axial solid hold-up distributions, the first stage in the derivation of the scale-up model concerns this profile.
Axial solid hold-up distribution

In accordance with the hydrodynamic partitioning in a coarse, mid-size and fine fraction, the axial solid hold-up in the DITS-reactor is considered to consist of three zones, schematically depicted in figure 3 (z is the dimensionless height)

1) a fluidized bottom zone \((0 < z < z_1)\), containing the coarse fraction; as can be found in dense fluidized beds, the hold-up can become as high as \(\varepsilon_s \approx 0.4-0.5 \ (\text{v/v})\) (Richardson and Zaki, 1954).

2) a gas agitated suspension zone \((z_1 < z < z_2)\), containing the mid-size soil fraction; in agreement with the sedimentation-dispersion model the axial profile shows an exponential course running from \(\varepsilon_s \approx 0.4\) down to \(\varepsilon_s < 0.1\) (Kato et al., 1972; Deckwer, 1985).

3) a top zone \((z_2 < z < 1)\), here the fine soil fraction is distributed homogeneously, resulting in a flat axial profile: \(\varepsilon_s \approx 0.05-0.1\)

![Figure 3](image)

**Figure 3** Theoretical curve of the axial solids hold-up distribution at the central axis of the DITS-reactor.
Following this simplified representation, the three zones are assumed to have each a characteristic particle size. The solid phase is thus modelled as three individual monodisperse fractions each with their characteristic position in the system. Based on the particle size distribution and the hydrodynamic classification, the following characteristic diameters have been chosen (between brackets, the corresponding terminal settling velocities for sand particles in water according to Perry and Chilton (1973))

* fine fraction : \( d_p = 100 \ \mu m \) \( u_p^\infty = 0.01 \ \text{m/s} \)
* mid-size fraction : \( d_p = 300 \ \mu m \) \( u_p^\infty = 0.03 \ \text{m/s} \)
* coarse fraction : \( d_p = 900 \ \mu m \) \( u_p^\infty = 0.10 \ \text{m/s} \)

Model assumptions

The relationship between the axial profile and the power input is based on the interaction between the fluid and the particles. Therefore, the statement "in order to sustain a solid phase in a fluid flow, work must be done by the liquid motion" (Hetseroni, 1982; Narayanan et al., 1969), serves as a starting point for the model assumptions. For the bottom (bed) compartment of the DITS-reactor, this is obvious. Due to the upward injected slurry flow, coarse material is kept in fluidization.

To achieve turbulent suspension in the bulk compartment, "the turbulence must be non-decaying and the viscous dissipation of the particles must be replenished by the turbulence" (Soo, 1956). In line with these two different mechanisms, the following model assumptions are made

* the power needed for the bottom compartment comes solely from slurry injection
* the power needed for the bulk compartment comes solely from gas injection
* the injected slurry and gas have no mutual influence
* the axial solid hold-up distribution is, in line with figure 3, partitioned in three zones

These assumptions are in line with the fact that the superficial upward slurry velocity rapidly decreases as a function of height due to the tapered shape (figure 2). Only at the deepest reactor part, where the cross section
of the reactor is smallest, the injected slurry flow provides enough superficial velocity to fluidize the settled fraction. Fluidization takes place where it is mostly needed (see part I).

**Bulk compartment**

In the gas agitated bulk, turbulent liquid motion has to replenish the (potential) energy which is lost by the particles due to settling. If particle suspension is seen as a dynamic process in which settling particles are lifted by the turbulent motion, the amount of work done by the liquid is quantified by the rate of potential energy loss of the solid phase (Kleijntjens et al., 1991a)

\[
\frac{dE_{\text{part}}}{dt} = \int_0^R \Delta \rho \cdot g \cdot \epsilon_s(x,y,z^*) \cdot u_p(x,y,z^*) \, dV_r \quad [1]
\]

In this equation, the total rate of loss in potential energy is determined as the integration over the reactor of the product of the relative specific solid phase weight, \( \Delta \rho \cdot g \cdot \epsilon_s(x,y,z^*) \), and the settling velocity, \( u_p \). However, in a suspension of particles, the actual settling velocity at a specific location is determined by hindrances of the surrounding particles. In order to calculate the correct rate of loss of potential energy these hindrances must be incorporated. Therefore a literature correlation was used which relates the settling velocity in a swarm of particles to the solid hold-up. For the polydisperse soil system, the correlation proposed by Barnea and Mirzahi (1973), covering a wide range of experimental results, was considered appropriate. This correlation is as follows

\[
u_p(x,y,z^*) = \frac{u_p \cdot (1 - \frac{S \cdot \epsilon(x,y,z^*)}{3(1-S\cdot\epsilon(x,y,z^*))})}{\exp\left[\frac{5 \cdot \epsilon(x,y,z^*)}{3(1-S\cdot\epsilon(x,y,z^*))}\right]} \quad [2]
\]

To determine the total rate of energy loss, the integral at the right side of equation 1 must be solved by substitution of \( u_p(x,y,z^*) \) according to equation 2. What results after substitution, is an integral which is completely determined by the spatial hold-up distribution.
In agitated systems, not only the particle suspension has to be sustained, also turbulent motion itself must be maintained. Incoming power is thus divided between two processes (Kleijntjens et al., 1991a)

\[
P_{\text{in}} = \frac{dE_{\text{Part}}}{dt} \left[ \text{tot} \right] + \epsilon_t \cdot V_r \cdot \rho_{\text{sl}}
\]

[3]

In equation 3 the generated turbulence is assumed to be isotropic, therefore it is allowed to use the specific turbulent energy conversion rate \( \epsilon_t \) (Hinze, 1959).

From the power balance given in equation 3, the efficiency of suspension can be estimated. This estimation is based on the phenomenon that, although particles are transported by the turbulence in all directions, only work in the vertical upward direction will be effective in counteracting gravity. The "suspension efficiency" is defined as the ratio of the power effectively used, and the total amount of power introduced in the system (Kleijntjens et al., 1991c)

\[
\eta_{\text{sus}} = \frac{[dE_{\text{Part}}/dt]_{\text{tot}}}{P_{\text{in}}}
\]

[4]

From experiments with a monodisperse solid phase (quartz sand with \( d_p = 260 \) \( \mu \)m) in a turbulent gas agitated tapered reactor, it was determined that \( \eta_{\text{sus}} = 0.2 \). A suspension efficiency of about 20% implies that most of the power introduced in the system, is used to maintain the turbulent motion. Only some 20% is used to counteract the gravity acting on the solids.

From a combination of equations 1 and 4, a direct relation between the amount of work necessary to sustain the solid phase (the power input), and the solid hold-up distribution can be established (Kleijntjens et al., 1991c)

\[
P_{\text{in}} = \frac{1}{\eta_{\text{sus}}} \cdot V_r \cdot 2g \Delta \rho \cdot \rho_{\text{sl}} \left[ \int u_p(z) \ast - \epsilon_s(z) \ast z \right] dz
\]

[5]

In this equation the axial height is replaced by the dimensionless axial height \( z^* / H \). To simplify the integration, the radial averaged solid hold-up is taken as \( \epsilon_s(z) = q(z) \ast \epsilon_s(0,z) \), where \( q \) is the radial shape factor. In this way, the radial shape of the profile at height \( z \) is incorporated in
the central axial profile (Kleijntjens et al. 1991c). The model expression for the specific power input in the bulk compartment due to gas agitation reads as

$$\frac{p}{V_{\text{bulk}}} = \frac{1}{\eta_{\text{sus}}} \cdot 2 \cdot e^* \Delta \rho^* \int_{z_1}^{z_2} \left( u_p(z) \cdot q \cdot \epsilon_s(0,z) \cdot z \right) dz$$

In equation 6 the transition from bed to bulk takes place at a dimensionless height $z_1$. Since in the bulk both the mid-size and the fine fractions are suspended, the integration in equation 6 contains two parts (see figure 3)

1) integration from $z_1$ to $z_2$, in this part of the integration an average particle size of 300 $\mu$m is taken.

2) integration from $z_2$ to $z = 1$, in this part an average particle size of 100 $\mu$m is taken.

For both fractions, a radial shape factor of $q = 0.6$ was used (Kleijntjens et al. 1991c). The power input necessary to keep the mid-size and the fine fraction in turbulent suspension, then can be determined from the axial measured profile.

Bottom compartment

Near the deepest point of the reactor coarse solids are kept in fluidization by means of an upward injected slurry flow. In fluidization, the frictional drag executed by the fluid, is equal to the apparent weight of the solids (Coulson and Richardson, 1978). The work done by the drag force is related to the loss in potential energy, therefore the power input can be described in terms of the mechanistic model derived above.

However, the mechanism in which the solid phase is sustained differs from that in the gas agitated bulk compartment. While turbulence was shown to be a rather inefficient way of sustaining the solid phase in the bulk compartment ($\eta_{\text{sus}} = 0.2$), it is assumed that in the fluidized zone the solid phase can be sustained very efficiently. The "efficiency of fluidization" is taken equal to unity. In addition, the axial distribution in both compartments differs significantly (see figure 3). In analogy with fluidized beds, the axial distribution in the bottom zone is taken to be uniform. In
addition, radial distributions are assumed to be negligible in the bottom zone. Incorporating this, the power input for the bottom compartment becomes

\[ P/V_{\text{pro}}^{\text{bed}} = 2g*\Delta\rho*\int_{0}^{z_1} (u_p^{\text{bed}} * \epsilon_s^{\text{bed}} * z) \, dz \]  \[7\]

In analogy with equation 6, an expression for the settling velocity in equation 7 is needed in order to solve the integration. For this purpose, the well known relation of Richardson and Zaki (1954) is used here

\[ u_p^{\text{bed}} = u_p^\infty * (1 - \epsilon_s^{\text{bed}})^{2.4} \]  \[8\]

From the measured axial solid hold-up and the transition height, the specific power input for the bed compartment can now be calculated.

EQUIPMENT AND EXPERIMENTAL METHODS

Experimental conditions

Experiments were carried out in a three phase (s-l-g) tapered slurry reactor with an angle of the tapered part equal to 60°. In figure 4 a three dimensional representation of the DITS-reactor is shown. Particle suspension was studied at laboratory scale (0.4 m³) and at pilot scale (4.0 m³). Both reactors were designed according to the principle of geometrical similarity, with the experimentally established optimal height over width ratio of unity (see part 1). The height, width and depth of the 4.0 m³ reactor are a factor 10^{1/3} larger than at the 0.4 m³ scale. The liquid height, H (and T) was 1.27 m at the smaller scale, and 2.70 m at the pilot scale. The reactor depth was equal to 0.45 and 1 meter respectively. The treatment section in the slurry recycle loop and the gas-slurry injector are described in part 1 (Kleijnjens et al., 1991b).
Having established a steady state suspension in the reactor, first the power input was determined at minimal gas and slurry flow rates. For this purpose, the gas input was decreased to the point where a slow moving fluidized bed of coarse particles could be observed visually. Second, the axial hold-up distribution was measured at the central axis of the reactor.

Performance of the soil was studied at the two (DITS-) reactor volumes with a total solids hold-up of 10, 20, 30 and 40 wt%, corresponding to a fractional volume hold-up of 0.041, 0.088, 0.140 and 0.210 v/v respectively.

**Power input in the bulk compartment**

Gas (air) is introduced at the bottom of the reactor by means of an injection manifold with equidistant injection points. The injection points were designed as vertical tubes with an inner diameter of 2 mm and a mutual distance of 2 cm. The gas flow is measured at the manifold with a rotameter. The measured pressure was used to express the gas flow at atmospheric conditions (Coleman, 1956). The ratio of the total injection surface, given
by the sum of the tube openings, and the cross sectional internal diameter of the manifold pipe, was chosen equal to 3 in order to circumvent maldistribution (Dow and Shreveport, 1950).

The standardized gas flow is used in the work equation for rising bubbles in order to determine the power input in the system

\[ p_{\text{inj}}^{\text{bulk}} = p^0 \cdot \frac{\phi_0}{\text{gas}} \cdot \ln \left[ \frac{p^0 + \rho_{\text{sl}} \cdot g \cdot H}{p^0} \right] \]  

In equation 9 the average bulk density is used, defined as

\[ \rho_{\text{sl}} = (1 - \varepsilon_s) \cdot \rho_\text{l} + \varepsilon_s \cdot \rho_s \]  

The power input due to gas injection is assumed to dissipate completely in the bulk compartment. The experimentally determined specific power input is calculated as \( P/V_{\text{inj}}^{\text{bulk}} \), in which \( V_{\text{bulk}} \) is determined from the transition height between the bulk and the bed: \( V_{\text{bulk}} = V_r \cdot (1 - z_1^2) \).

**Power input in the bottom compartment**

In addition to air, slurry is injected simultaneously. For this purpose, the dual injection manifold was developed (see part 1). In order to determine the power input due to slurry injection, both the potential and kinetic contributions have to be considered. The kinetic term, \( (0.5 \cdot \phi_{\text{inj}}^{\text{inj}} \cdot \rho_{\text{sl}} \cdot v_{\text{inj}}^2) \), is calculated from the measured injection flows reported in part 1. It was calculated that the kinetic energy added maximal about 3 W to the reactor system (at 4 m³).

To determine the potential energy contribution, first the amount of power present in the injected slurry flow has to be considered. Within the framework discussed, as potential power is taken that amount of work which needs to be done by the slurry to overcome the hydrostatic pressure at the moment of entrance. In the DITR-reactor the hydrostatic pressure is a combination of the pressure due to the height of the dense slurry in the bottom compartment, \( \rho_{\text{fb}} \cdot g \cdot H \cdot z_1 \), and the height of the slurry in the bulk, \( \rho_{\text{sl}} \cdot g \cdot H \cdot (1 - z_1) \). In addition to the amount of power injected at the bottom, power is withdrawn from the system due to the slurry flow leaving the system over the top (the overflow rate equals the injection flow rate). Assuming
that the slurry overflow contains no particles, the following relation holds for the amount of power dissipated in the reactor

\[ \phi_{\text{bed}}^{\text{inj}} - \phi_{\text{v}}^{\text{inj}} \propto H \times g \times (\rho_{fb} \times z_1 + \rho_{s1} \times (1-z_1) - \rho_1) \]  \[ \text{(11)} \]

From the measured flows reported in part 1 (Kleijntjens et al., 1991b) it was derived that the power introduced according to equation 11 is about 10 times larger than the kinetic term. It was therefore concluded that the kinetic contribution could be neglected.

According to the model assumptions, the power input due to slurry injection dissipates completely in the fluidized bed compartment. The specific power input in the bed, \( P/V_{\text{bed}}^{\text{inj}} \), is calculated by dividing the amount of power according to equation 11 by the bed volume.

**Measurements of the axial hold-up distributions**

To determine the specific power input from the axial profiles (equations 6 and 7), the axial particle hold-up distributions were determined, at the state of experimentally minimized power input. The profiles were measured with a solids sampling probe (figure 5). To measure the hold-up, the open probe is brought to the desired location, then, after several seconds, the probe is closed and withdrawn from the slurry. Subsequently, the probe content is analyzed for its dry mass. From the dry mass and the sampling volume of the probe (9.42 ml), using the densities of the solid phase and the liquid phase, the solid volume fraction can be calculated.
Figure 5 Solids sampling probe.

To determine the axial distribution, at regular intervals, three samples were taken within a period of several minutes. In line with previous work (Kleijn et al., 1991a) the presence of radial profiles (y-directions) is incorporated into the axial distribution by way of the radial shape factor (q). Profiles in the x-direction (reactor depth) were considered negligible. The standard deviations in the measured solid contents varied from 5 to 30% of the mean value. This variation was a result of turbulent fluctuations.

EXPERIMENTAL RESULTS

Once the solid/liquid separation in the circulation unit operated at certain flow capacity, it was difficult to alter the flow rate of the liquid injected at the reactor bottom. Therefore it was the air flow which really determined the state of suspension in the system. Given a certain injection flow rate (in figure 6 the gas flows (m³/min) are given), a fluidized bed zone is obtained by minimizing the gas injection flow.
For the empirically established slurry flow and the minimized gas flow, the axial hold-up distributions at the central axis of the system were measured. In figure 6 the axial profiles are depicted. It must be noted that due to the polydispersity of the solid phase, the composition of the sampled slurry is different at each height. An indication of the height dependency of the slurry composition is given in part 1 (Kleijntjens et al., 1991b).

![Figure 6](image)

**Figure 6** Measured axial solids hold-up distribution at the axis of the 0.4 and 4.0 m³ DITS-reactor.

**DISCUSSION**

**Determination of transition heights**

To be able to determine the specific power input for the bulk and bed compartments, both the power input as well as the compartment volumes have to be determined. To establish the latter, it is necessary for each experiment to define the transitions heights ($z_1$ and $z_2$). In line with figure 3, the most straightforward determination is offered by the measured
axial profiles. However, the experimental results in the DITS-reactor are not as unambiguous as the theoretical curve. Especially for the transition between bed and bulk, \( z_1 \), the difference between the theoretical curve and the practical curves (figure 6) is significant. From the experimental curves, no clear transition point from the bottom compartment to the bulk can be identified. Therefore another method is used to estimate the transition height, \( z_1 \).

Based on the separation performance of the DITS-reactor, it is concluded that 25 wt% of the soil material (the coarse fraction with \( d > 700 \mu m \)) is present in the bottom compartment as a fluidized bed. In addition, it is assumed that \( \varepsilon_{\text{bed}} = 0.5 \). Since, for each experiment, it is exactly known how many kilograms of soil were added to the system, the volume of the bottom compartment can be calculated. Finally, from the calculated volume of the fluidized bed, \( z_1 \) can be determined from the following relation:

\[
V_{\text{bed}} = V_r (1-z_1^2).
\]

Using this method, for the 0.4 m\(^3\) reactor the transition height \( z_1 \) was estimated at 0.22, 0.33, 0.47 and 0.512 for the four slurry densities respectively. For the 4.0 m\(^3\) reactor \( z_1 \) is found at heights of 0.07, 0.10, 0.13, and 0.16 respectively. In figure 6, the upward arrows at the left side denote the position of \( z_1 \).

In agreement with the model curve, the transition from the mid-size to the fine fraction, \( z_2 \), could be determined from the measured profiles due to the clear transition from exponential to homogeneous zone. The downward arrows at the right side locate these transitions.

**Determination of P/V in the bottom and bulk compartment**

To determine the total power input due to the injected slurry and gas flows in the 0.4 and 4.0 m\(^3\) DITS-reactor, equations 9 and 11 were used. Subsequently, from this total power input, the specific power input in both compartments, \( P/V_{\text{bed}} \) and \( P/V_{\text{bulk}} \), was derived using the actual volume of the bottom and bulk compartments. The compartment volumes were determined according to the method discussed above. In figure 7 the results are given for the experimental specific power input.
The specific power input from the axial profiles, $P/V_{\text{bed}}^{\text{pro}}$ and $P/V_{\text{bulk}}^{\text{pro}}$, was determined according to the model equations 6 and 7. In figure 7 these inputs are depicted.

![Graphs showing specific power input in bottom and bulk compartments for 0.4 m$^3$ and 4.0 m$^3$ DITS-reactor.](image)

**Figure 7** Specific power input in bottom and bulk compartment for the 0.4 and 4.0 m$^3$ DITS-reactor.

As shown, in the 0.4 m$^3$ DITS-reactor, the $P/V_{\text{bed}}^{\text{pro}}$ ranges from 100-200 W/m$^3$, while $P/V_{\text{bed}}^{\text{inj}}$ is found to be in between 30 and 20 W/m$^3$. The values of $P/V$ based on the solid hold-up distributions are about a factor 5 larger. At a volume of 4.0 m$^3$, $P/V_{\text{bed}}^{\text{pro}}$ is a factor 2-3 larger than $P/V_{\text{inj}}^{\text{bed}}$, 300-600 W/m$^3$ and 100-200 W/m$^3$ respectively. For the bulk compartment, the specific power input calculated according to the injection flow rates are in reasonable
good agreement with those determined from the hold-up profiles. At 0.4 m$^3$, the $P/V_{\text{pro}}$ bulk is still somewhat larger than the $P/V^\text{bulk}_{\text{inj}}$, but at a scale of 4.0 m$^3$, the $P/V$ values do not differ significantly.

The deviations between the results of these two calculation methods are due to the assumptions made. Both methods suffer from the fact that in practice, the transitions from bed to bulk will be not so unambiguous as assumed in the model curve (figure 3). There will be entrainment from the bed to the lower part of the bulk. Consequently, an exact determination of $z_1$ and therefore of the compartment volumes is hard to make. For the $P/V_{\text{pro}}$ holds that the assumed partitioning in three mono-disperse fractions with each a characteristic settling velocity is not in agreement with practice. In the slurry reactor a part of the mid-size fraction is present in the bottom compartment, while some of the coarse particles are in the bulk (see part 1). Since the partitioning largely determines the amount of power needed to sustain the solid phase (equations 6 and 7), this assumption largely contributes to the uncertainty in $P/V_{\text{pro}}$.

Concerning $P/V_{\text{inj}}$, the assumption that both compartments are agitated independently is questionable. The injected slurry will not dissipate all its power in the bottom compartment, some of the power will be dissipated in the bulk compartment. However, it can be calculated that most of the slurry power is used for fluidization, the power input in the bulk is negligible. Similarly, the contribution of the gas injection to the $P/V_{\text{bed}}$ also will be small. The gas phase performs as a spout in the bottom thereby not contributing to the fluidization. Therefore it was concluded that the $P/V$ determination from the injection flows are more reliable than the values derived from the profiles. Scale-up of the DITS-reactor will be based on the $P/V$ values derived from the injection flows.
Scale-up

Based on the fact that the DITS-reactor is a new type of operation, a factor 10 enlargement (compared to the 4 m$^3$ pilot-plant) is considered appropriate (Peters and Timmerhaus, 1981). The 40 m$^3$ reactor is taken geometrically identical to the 4.0 m$^3$ pilot plant. The resulting depth is taken equal to 2 m (which also is the length of the injector), the H/T ratio is taken equal to unity (Kleijntjens et al, 1991b) with H equal to 6.3 m. A slurry density of 40 wt% was assumed.

To predict the power input for the 40 m$^3$ DITS-reactor, the trend in the ratios of $P/V_{inj}^{\text{bulk}}$: $P/V_{inj}^{\text{bed}}$ will be used. In figure 8 the values of these ratios are shown for the two scales examined.

![Figure 8](image-url)  
**Figure 8** Ratio of $P/V_{inj}^{\text{bulk}}$: $P/V_{inj}^{\text{bed}}$ for the 0.4 and 4.0 m$^3$ DITS-reactor.

As shown in figure 8, the ratio $P/V_{inj}^{\text{bulk}}$: $P/V_{inj}^{\text{bed}}$ depends on scale and slurry density. At larger slurry densities the ratio increases, with $P/V_{inj}^{\text{bulk}}$ increasing relatively more than $P/V_{inj}^{\text{bed}}$. Increasing the scale from 0.4 to 4.0 m$^3$ results in a decrease of the ratio from values in between 1-10 down to 0.03-0.2. Extrapolation of this trend would result in about 0.05 for the 40 m$^3$ reactor. This indicates that at large scale the specific power input in the fluidized bed is 20 times higher than in the bulk. Using a conservative extrapolation for the bulk ($P/V_{\text{full}} = P/V_{\text{pilot}}$), the specific power input in
the bulk is 50 W/m³, resulting in P/V equal to 1000 W/m³ for the fluidized bed.

To estimate the injection flows, \( z_1 \) at 40 m³ is, in line with the trend at 0.4 and 4.0 m³, defined at \( z=0.1 \), resulting in a bed volume of about 0.4 m³. Knowing the P/V and the volume of the two compartments, from equation 9 and 11 the injection flows can now be calculated. The required gas and slurry injection flows for the 40 m³ DITS-reactor are 2.1 m³/min and 1.4 m³/min respectively.

CONCLUSIONS

From the experiments and the model the following has been concluded

* The two compartments (bottom and bulk) of the Dual Injected Turbulent Separation (DITS-) reactor can be modelled on the basis of a different hydrodynamic behavior of the soil fractions in the slurry.

* The values for \( P/V_{\text{bed}}^{\text{pro}} \) and \( P/V_{\text{bulk}}^{\text{pro}} \) were larger than the \( P/V_{\text{bed}}^{\text{inj}} \) and the \( P/V_{\text{pro}}^{\text{bed}} \) for both the 0.4 and 4.0 m³ reactor. For the bottom compartment (bed) the P/V from the profiles were a factor 3-5 larger, for the bulk a factor 1-2.

* Since the determination of the specific power input from the injection flows (\( P/V_{\text{inj}}^{\text{pro}} \)) was more direct than those obtained from the model (\( P/V_{\text{pro}}^{\text{inj}} \)), it was concluded that the injected power input was more suitable for scale-up purposes.

* The injected power needed for a 40 m³ large scale DITS-reactor (40 wt% hold-up), is about 1000 W/m³ for the bottom compartment and around 50 W/m³ for the bulk. To achieve this power input, the injection flow rates for slurry and air are 1.4 m³/min and 2.3 m³/min respectively.

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List of symbols

\[ d \] = diameter, m  \
\[ F_{\text{pot}} \] = potential energy of the solids in suspension, kg\(m^2/s^2\)  \
\[ F \] = force, kg\(m/s^2\)  \
\[ g \] = gravitational constant, 9.8 m/s\(^2\)  \
\[ G \] = gas phase  \
\[ H \] = reactor height, m  \
\[ H_{\text{set}} \] = height of the settler, m  \
\[ L \] = liquid phase  \
\[ l_e \] = length of energy containing eddy, m  \
\[ p \] = pressure, kg/m\(s^2\)  \
\[ P \] = power, W  \
\[ P_{\text{in}} \] = power input, W  \
\[ P/V \] = specific power input, W/m\(^3\)  \
\[ q \] = radial correction factor  \
\[ \text{Re}_p \] = relative Reynolds particle number  \
\[ \frac{d \times \rho_f \times (u_{\text{pi}} - u_{\text{fi}})}{\eta_f} \]  \
\[ S \] = solid phase  \
\[ t \] = time, s  \
\[ T \] = maximal width of tapered reactor, m  \
\[ u \] = velocity, m/s  \
\[ u' \] = turbulent liquid fluctuation velocity, m/s  \
\[ V_r \] = reactor volume, m\(^3\)  \
\[ v_l \] = liquid velocity, m/s  \
\[ x \] = coordinate in the depth of the reactor, m  \
\[ y \] = coordinate in the width of the reactor, m  \
\[ z \] = coordinate in axial direction, reactor height, m  \
\[ z^* \] = coordinate in axial direction  \
\[ z \] = dimensionless coordinate in axial direction

Greek symbols

\[ \epsilon_{\text{tot}} \] = total solid hold-up in, m\(^3\) solids/m\(^3\) reactor
\( \varepsilon_{(xyz)} \) - local solid hold-up, m\(^3\) solids/m\(^3\) reactor

\( \varepsilon_{(z)} \) - radial averaged solid hold-up (axial), m\(^3\) solids/m\(^3\) reactor

\( \varepsilon_t \) - specific turbulent kinetic energy transfer rate, W/kg

\( \eta \) - dynamic viscosity, kg/m.s (Pa.s)

\( \eta_{sus} \) - suspension efficiency

\( \phi \) - volumetric flow rate, m\(^3\)/s

\( \rho \) - density, kg/m\(^3\)

\( \Delta \rho \) - density difference between solid and liquid phase, kg/m\(^3\)

\( r_p/r_t \) - particle response number, ratio of the particle response time \( r_p \) and the characteristic turbulent time \( r_t \)

Super- and subscripts

bed - bottom (bed) compartment

bulk - bulk compartment

drag - drag force action at particle

e - eddy

f - fluid phase, water

gas - gas phase, air

i - specific direction

in - incoming

inj - experimentally established injection flows

l - liquid phase

pro - established from the axial profiles

o - atmospheric conditions

p - particle

sl - slurry

t - turbulent

1 - transition height from fluidized zone to mid-size suspension zone

2 - transition height from mid-size to fine suspension zone

\( \infty \) - in infinite medium, used for terminal settling velocity
LITERATURE


PROCESS DESIGN AND FEASIBILITY
Feasibility study of a biotechnological slurry process for soil decontamination

K.Ch.A.M. Luyben, R.H. Kleijntjens, T.A. Meeder
Kluver Laboratory for Biotechnology, Delft University of Technology
Julianalaan 67, 2628 BC Delft, the Netherlands

ABSTRACT

The feasibility of a biotechnological slurry process for soil decontamination was investigated. This comprised of an integration of previous work concerning a number of physical, chemical and practical aspects of the process. The integration also involved the newly designed slurry reactor for soil processing, the DITS (Dual Injected Turbulent Separation)-reactor. For the complete process a flowsheet was designed, which formed the basis for a cost estimation. Following a scale-up strategy, costs were estimated for three scales of operation, the pilot scale (16 m³), the demonstration scale (160 m³) and the full scale (2680 m³). It was estimated that, if decontamination down to the Dutch B-level (1 g/kg) is appropriate, both the demo-plant and the full scale can operate for a price below the 250 Dfl per ton soil, which is economically feasible.

KEYWORDS

Soil decontamination, slurry process, full scale, economic feasibility scale-up
INTRODUCTION

In most industrialized countries, a large number of polluted sites have been discovered during the last decade. In the Netherlands, for instance, over 100,000 polluted sites have been estimated. Of these sites, 6000 are qualified as urgent and are planned to be decontaminated in the next decade (estimated costs 5.10^9 Dfl, Keuzenkamp et al., 1990). The development of economically and environmentally feasible soil decontamination processes is therefore needed (Sanning et al., 1990; Eijsackers, 1989).

For the treatment of decontaminated soil, distinction must be made between in-situ and ex-situ techniques. In urban areas, where it is not always possible to excavate the polluted site, mostly in-situ techniques are used (Staps, 1990). Groundwater, containing dissolved pollutant, is pumped from the polluted site to an waste-water treatment plant. Due to the continuous withdrawal of groundwater, the pollutant concentration is decreased (Booth et al., 1990)). Another example of in-situ reclamation is biorestoration, here the microbial activity at the polluted site is stimulated by injection of organisms, nutrients and oxygen. Advantages of in-situ techniques are their simplicity and the low cost (about 100 Dfl/ton). Major difficulties in in-situ techniques are adhesion of the pollutant to the solid matrix, the hydrogeological situation, the ambiguity of the decontamination level reached, and the long treatment times (periods over 1-2 year, Werner and Brauch, 1988).

In ex-situ techniques, the polluted soil is excavated and transported to an installation located either at the site or off-site. If the installation is located on site, the treated soil can be directly used to refill the excavated site. Otherwise, the excavated site is generally refilled with clean soil or sand. The size of the contamination, transport costs, technical and socio-political factors determine whether such an installation can be constructed on site. Major advantage of ex-situ treatment installations is the fact that decontamination takes place under controlled and optimized reactor conditions.

Recently, feasibility studies have been carried out to compare different ex-situ remediation techniques (Sanning et al, 1990; Hinsenveld et al, 1990).
It was reported that, due to the variety of polluted sites different techniques are emerging in parallel. It depends on the origin and type of the pollution, the (hydro) geological state of the site and on social factors, which technique is most suitable. In general, the following five aspects can be considered to judge the merits of ex-situ remediation techniques:

- range of pollutants
- soil types
- grade of decontamination
- end product quality
- costs

Considering these aspects, it can be concluded that physico-chemical treatment, such as thermal soil decontamination, can obtain a high level of decontamination for organic soil pollutants. Costs are mostly in between 150 and 250 Dfl/ton (Reintjes, 1990). Disadvantages of thermal soil processing are related to the treatment of wet silty soils and the bad quality (infertility) of the treated soil.

For ex-situ biological techniques, such as landfarming, different types of organic pollutants can be treated at low costs. Although the end product quality is good (fertile soil), landfarming suffers from long treatment times and low conversion rates. In addition clay/silty soils are difficult to treat (agglomerates).

To take more advantage of the possibilities that biological soil decontamination can offer (mineralization, good end product quality), a biotechnological soil slurry process has been developed (Kleijnjens et al., 1990). Regarding the five criteria given above, the following aims for the soil slurry process were set:

- treatment of excavated soils, polluted by oil/fuel
- a large variety of polydisperse soil types (including clay/silt soils) as feed
- reduction of the pollutant concentration below 1 g/kg (dutch B-level for oil) and, if possible, below the A-level (50 mg/kg)
- healthy soil as end product
* an economic process design (low cost)

To be able to judge the biotechnological slurry process in terms of these aims, the process outline is given by way of the following points

**design**
- flowsheet
- scale-up

**parameters**
- soil composition
- solids hold-up
- power input
- kinetics

Crucial in this paper is the integration of previous work on the slurry process. From the integrated process design, with the help of a proper scale-up strategy, an estimation of the process economics subsequently is made.

**DESIGN OF THE SLURRY PROCESS**

**Flowsheet**

Most *ex-situ* techniques can be divided in three sections (figure 1). First, the excavated soil is pre-treated by means of grinding, crushing and sieving. The actual conversion or removal of the pollutant takes place in a reactor section, and finally after-treatment completes the process. Also for the slurry process this partitioning holds.
Figure 1 The three sections of soil treatment processes.

First stage in the slurry process is the soil pre-treatment (figure 2). Vibrating grizzly bars (S2) are used in order to remove debris (e.g. stones and roots larger than 60 mm). Subsequently, the underflow of the bar screen is used to feed a micro-pulverizer (M1). After grinding, the soil is transported (belt B6) to a sieve where the coarse gravel fraction ($d > 4$ mm) is separated. In this wet screening section (S10), both recirculation fluid and the nutrient addition are used as washing water.

The first reactor unit (R12) is fed with polydisperse soil, added as slurry (particle size between 1 $\mu$m and 4000 $\mu$m). This is a tapered suspension reactor with a height/width ratio equal to unity. This reactor also serves as a separator, according to the DITS (Dual-Injected-Turbulent-Separation) reactor principle (Luyben and Kleijntjens, 1988). The soil is separated into a coarse (settling) fraction, fluidized by means of an upward injected slurry flow at the bottom of the reactor, and a fine fraction which is kept in turbulent suspension. In the two phase (g-l) line injector, air (originating from compressor C3) is simultaneously injected with slurry provided by the recirculation unit. In this unit, slurry from the top of the reactor is pumped (P7) to a solid-liquid separator (S9) in which particles above 100 $\mu$m are removed. Subsequently, the treated slurry is injected at the reactor bottom (pump P8) (see figure 2).

From the bottom of the first reactor, the fluidized coarse fraction is withdrawn (extrusion screw B13) and directly fed to the dewatering section (S17). The fines are given a further treatment in a cascade of suspension reactors. Approximating plug flow, a cascade of three equal mixed tanks was selected as the optimal reactor configuration (Levenspiel, 1962) for the
first order degradation of the pollutant (see below). Due to the removal of the coarse fraction at the beginning of the process, no settling problems are expected in the cascade reactors (R15, R16 and R18), that are agitated by means of air.

Figure 2 Flowsheet of the biotechnological slurry process.

After being treated in the cascade, the slurry is dewatered by means of filtration (S17). As a filter aid, the coarse fraction from the DITS-reactor is used. From the dewatering section, percolated process water is recycled to the beginning of the process (S10). The recycle ratio is set at about 3, indicating that 75% of the fluid flow through the system is recycled. The additional liquid flow originates from the nutrient flow. Despite its benefits in terms of reducing the waste water stream, it should be
recognized that, if toxic compounds are formed in the process, recycle ratios as high as 3 can result in the accumulation of toxic compounds in the system.

To prevent the pollutants from leaving the reactors in the gas phase, the air outlet is filtered using a biofilter (F19). Recycling water is heated with excess heat from the the compressor (H5), the reactor operation temperature is 30 °C.

In essence, the flowsheet is based on the inhomogeneous distribution of pollutants in soil. From the literature and previous experiments, it is known that the coarse soil fraction contains a relatively small amount of pollutant, whereas more than 90 % is adsorbed to the fines \( d_p < 100 \, \mu m \) (Werther et al., 1990). In addition, the suspension behavior of the coarse fraction under turbulent conditions is problematic (settling, Kleijnjens et al., 1991a). Due to the removal of the coarse material in the beginning of the process (DITS-reactor), the process capacity is improved and settling problems in the largest part of the reactor volume (the cascade) are circumvented. The reactors are efficiently used for the decontamination of the polluted fines, instead of being used for the transport of inert mass.

By integrating the first reactor and the separation, the number of unit operations is minimized. In addition, the coarse fraction is washed before it leaves the system. Due to the attrition which takes place in the fluidized bottom compartment, pollutants enclosed in the coarse fraction are removed and treated. The residence time for the coarse fraction is selected in line with the required washing time (several hours). To enhance the conversion rate of the microbial degradation in the bioreactors, process conditions are optimized in terms of power input, aeration rate, pH, temperature and nutrients.

Data on the above described process, were gathered from a 40 liter mini-plant to determine the kinetics (Kleijnjens et al., 1991b). At several scales (up to 4 m³) hydrodynamics and suspension efficiencies were studied for the reactor design (Kleijnjens et al., 1991a). Crucial for the integrated process design therefore is scale-up.
Scale-up

In scale-up, several pathways can be distinguished depending on the availability of experience, rules of thumb or scale-up theory (Johnstone and Thring, 1957). When accurate data are not available, or when past experience does not give an adequate design basis, pilot tests are generally necessary (Peters and Timmerhaus, 1981). In modern scale-up, pilot plant experiments are combined with mathematical models. Experimental results are translated into model parameters which, in turn, are used to support the extrapolation made in the scale-up procedure (Rose, 1981). In addition, the pilot plant serves a marketing purpose. Since a considerable amount of material is processed, the feasibility of the process is demonstrated.

On the basis of the experience with this particular process (Kleijntjens et al., 1991b), a stepwise approach was considered to be most appropriate. First, a 16 m³ pilot-plant was designed according to the flowsheet (4 * 4 m³ reactors). This stage was intended to test the degradation kinetics at a scale 400 times larger than the mini-plant. In essence, the 16 m³ pilot plant can be used to translate laboratory experience into practice.

The second stage is the construction of a demonstration plant (4 * 40 m³). Using this, optimization of the DITS-reactor and the process conditions in the cascade can be carried out at a scale 10 times that of the pilot plant. The dimensions of the 40 m³ reactors are 6.3 m for the height and width (tapered geometry with H/T=1), and 2 m in depth. Again the flowsheet according to figure 2 can be used.

Based on the results achieved on the demonstration scale, a full scale system (2680 m³) should be constructed as two parallel reactor cascades of 1340 m³ each. The volume of the DITS-reactor at full scale can be taken equal to that in the demonstration scale (40 m³). The additional cascade volume of 1300 m³ is achieved by means of 3 tanks, each with a volume of 435 m³. Also the height and width of the cascade reactors are taken to be equal to those of the 40 m³ DITS-reactor (6.3 m, for a geometry of H/T=1). The depth of each cascade reactor is 22 m, giving the 435 m³ reactors a channel-like appearance. Due to the large capacity of the cascade reactors at full scale, the smaller DITS-reactor at the head of each cascade mainly performs as a separator.
To get a feeling of the magnitude and character of the unit operations used in the slurry process, a three dimensional impression of the demo-plant (160 m³) is depicted in figure 3. As is shown, the tapered shape (H/T=1) results in a compact installation (maximum height is about 15 m). The most complex unit-operation is the DITS-reactor at the beginning of the process. Once the separation of the coarse fraction is achieved, the slurry handling of the fines proceeds relatively easy in the cascade.

Figure 3 Three dimensional representation of the demonstration plant (160 m³).

An important aspect in the feasibility study of the slurry process is the mobility of the operation. By constructing the 4 reactors as individual units which are assembled at the polluted site, transport by truck is possible for the demo-plant. However, it should be recognized that due to the weight of the installation (full weight = 200 ton), the foundation requires proper attention.
From an economic point of view, the mobility of the installation might be crucial. For large sites or large countries, the transportation costs of the soil from the site to the installation, can easily disrupt the economics of ex-situ soil processing. For the sake of simplicity, the economics of the slurry process will be considered in terms of the pure treatment costs per ton soil. Transportation costs are not included (for the Dutch situation these are about 50 Bfl/ton).

PROCESS PARAMETERS

Soil composition

Table 1 gives the composition of the soil feed to the slurry process.

Table 1 Soil composition.

<table>
<thead>
<tr>
<th>70 wt% net particle mass</th>
<th>4% gravel</th>
<th>8% debris</th>
<th>18% water</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 &lt; d &lt; 700 μm</td>
<td>700 &lt; d &lt; 4000 μm</td>
<td>4 &lt; d_p &lt; 60 mm</td>
<td>d_p &gt; 60 mm</td>
</tr>
<tr>
<td>75% fines</td>
<td>25% coarse</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Based on this soil composition, 1 ton (1000 kg) of excavated soil corresponds to a net polluted particle mass of 700 kg. In figure 4 the particle size distribution of the polluted soil mass is given. It can be seen that the distribution is rather uniform. The transition from fine to coarse fractions is determined by the settling behavior of the larger particles and the hydrodynamics in the reactor (experimental value d_p > 700 μm) (Kleijnj Jens et al., 1991a). For the soil density, an average of 2200 kg/m³ was taken, being the average of the fines (2050 kg/m³) and the coarse fraction (2600 kg/m³).
Figure 4 Particle size distribution of the soil feed.

**Solids hold-up**

Although the kinetics in the mini-plant were determined with a solids hold-up of about 15-20 wt%, the loading in the cascade reactors was made to be 40 wt% (0.24 v/v). In the mini-plant the slurry density was limited for practical reasons, and not because of kinetic arguments or oxygen mass transfer problems. Furthermore, in the 4 m³ pilot plant, a 40 wt% slurry density could be suspended without difficulty (Kleijntjens et al, 1991c). Taking the slurry density in the cascade reactors to be equal to 40 wt%, the average solids hold-up in the DITS-reactor becomes 51 wt% (0.32 v/v). This is due to the fact that an extra 25% of coarse material is present in the bottom zone of the first reactor. Comparing the particle loading in this process design and the solids hold-up used in the mining industry, it can be concluded that the densities chosen are realistic. In literature, slurry systems with densities as high as 60-70 wt% can be found (Hallet, 1980; Parthen et al, 1990).
Power input

To estimate the power input, separation must be made between the 4 m³ and 40 m³ DITS-reactors and the 4 and 40 m³ cascade reactors. For the pilot scale, the power inputs were determined experimentally, while for the 40 m³ reactors, extrapolations were used (Kleijntjens et al., 1991c). In Table 2, the power input needed for the various reactors is given.

<table>
<thead>
<tr>
<th></th>
<th>4 m³ DITS-reactor</th>
<th>40 m³ DITS-reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>P/V (W/m³)</td>
<td>φ (m³/min)</td>
<td>P/V (W/m³)</td>
</tr>
<tr>
<td>bulk 50</td>
<td>0.43 (air)</td>
<td>bulk 50</td>
</tr>
<tr>
<td>bed 250</td>
<td>0.15 (slurry)</td>
<td>bed 1000</td>
</tr>
<tr>
<td></td>
<td>hold-up = 51 wt%</td>
<td>hold-up = 51 wt%</td>
</tr>
</tbody>
</table>

**Cascade reactors**

<table>
<thead>
<tr>
<th></th>
<th>P/V = 50 W/m³</th>
<th>hold-up = 40 wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total gas input for the cascade</td>
<td>3 * 4 m³</td>
<td>3 * 40 m³</td>
</tr>
<tr>
<td>φ (m³/min)</td>
<td>1.3 (air)</td>
<td>6.3 (air)</td>
</tr>
</tbody>
</table>

These power input data were calculated according to the methods described in Kleijntjens et al., (1991c).

**Kinetics**

The soil treated in the process is assumed to contain oil with an average pollutant concentration of 2-3 g/kg (see figure 5). Two different degradation rates will be compared, an estimated best case degradation rate, giving a short residence time, and the experimentally measured degradation rate (Kleijntjens et al., 1991b). For the measured kinetics, two soil
residence times are regarded, first the residence time required for the b-level, and second, the residence time to reach the a-level. This latter residence time is an extrapolation based on estimated desorption kinetics of the pollutant.

![Chromatograms](image)

**Figure 5** Chromatograms before and after processing.

In the economic study described below, all three residence times will be considered. This leads to the following residence times for soil in the reactor section
\* \( \tau_{\text{b-level}} = 2 \) days (best case)
\* \( \tau_{\text{b-level}} = 6 \) days (mini-plant)
\* \( \tau_{\text{a-level}} = 28 \) days (desorption)

The residence time in the pre- or after treatment sections are not taken into account.

**ECONOMICS OF THE SLURRY PROCESS**

Considering the economic feasibility of the slurry process, the different scales have to be studied. With regard to the 16 m³ pilot plant, the investment costs are of prime importance. The purpose of such a pilot operation is to prove the technical feasibility. In this plant, no significant amounts of polluted soil can be processed. It can therefore be expected that the costs at this small scale are relatively high. The pilot plant is seen as a necessary step in the process development (Johnstone and Thring, 1957).

For the demonstration plant the economical situation is different. Having proven the process to be operational at pilot scale, significant amounts of polluted soil can be treated in the demo-plant. Since larger investments are required to construct this plant, at least return of the investments should be obtained. The capacity, power input and end product quality are to be investigated in relation to the costs. Due to the financial risks which are taken in this stage, it is necessary to have potential polluted sites for which the slurry process can be considered a serious remediation technology. For the full scale plant, a reasonable return on investment is needed. Marketing becomes more important at this stage. Due to the economy of scale, the price per ton of soil will be lower than for the smaller scales.

The economics of the slurry process are determined by the following three aspects
* process capacity
* investment costs
* variable costs

Table 3 shows the capacities of the three plants for each of the three residence times. It can be seen that the capacity of the pilot plant is too small to be used for remedial action purposes. For small sites the demo-plant has an appropriate capacity. From pilot to demonstration scale the capacity increases by a factor of 10. From demonstration to full scale, the capacity increases 17-fold. With a residence time of 6 days, the capacity of the full scale plant comes close to that of thermal treatment installations (250 ton/day).

Table 3 Treatment capacities of the three plants.

<table>
<thead>
<tr>
<th>soil</th>
<th>Pilot-plant (16 m³)</th>
<th>Demo-plant (160 m³)</th>
<th>Full scale plant (2680 m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 days</td>
<td>5.04</td>
<td>50.4</td>
<td>801</td>
</tr>
<tr>
<td>6 days</td>
<td>1.63</td>
<td>16.3</td>
<td>268</td>
</tr>
<tr>
<td>28 days</td>
<td>0.36</td>
<td>3.6</td>
<td>58</td>
</tr>
</tbody>
</table>

Investment costs

In table 4, the investment costs are given for the three scales. The investments are divided into mechanical, electrical and civil engineering costs. For each item three prices are given. The first holds for a soil residence of 2 days, the second for 6 days and the third for 28 days. The data in table 4 are estimates with an inaccuracy of about 30 %. As is shown in the table, the major investment costs are formed by the equipment (reactors, piping, pumps etc.). The electrical engineering make up about 10-20 % of these costs. The civil engineering for the pilot plant is
negligible, for the demonstration plant they are in the order of the electrical engineering, and for the full scale they are in the order of the equipment costs. The reason for the latter is that the full scale cascade reactors are made of concrete, and therefore these reactors are considered as civil engineering works.

Table 4 Investment costs.

<table>
<thead>
<tr>
<th></th>
<th>Pilot-plant (16 m³)</th>
<th>Demo-plant (160 m³)</th>
<th>Full scale (2680 m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>investment costs (in 10⁶ Dfl)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>mechanical engineering</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>steel</td>
<td>1.20</td>
<td>3.50</td>
<td>8.50</td>
</tr>
<tr>
<td>reactors, piping, pumps</td>
<td>1.10</td>
<td>2.70</td>
<td>5.70</td>
</tr>
<tr>
<td></td>
<td>1.10</td>
<td>2.30</td>
<td>4.40</td>
</tr>
<tr>
<td>electrical equipment</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(power supply control etc.)</td>
<td>0.25</td>
<td>0.55</td>
<td>1.30</td>
</tr>
<tr>
<td></td>
<td>0.23</td>
<td>0.50</td>
<td>1.20</td>
</tr>
<tr>
<td></td>
<td>0.23</td>
<td>0.46</td>
<td>1.10</td>
</tr>
<tr>
<td>civil engineering</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(roads, foundation concrete etc.)</td>
<td>negl.</td>
<td>0.70</td>
<td>8.60</td>
</tr>
<tr>
<td></td>
<td>negl.</td>
<td>0.58</td>
<td>7.70</td>
</tr>
<tr>
<td></td>
<td>negl.</td>
<td>0.51</td>
<td>6.80</td>
</tr>
<tr>
<td>total</td>
<td>1.45</td>
<td>4.75</td>
<td>18.4</td>
</tr>
<tr>
<td></td>
<td>1.33</td>
<td>3.78</td>
<td>14.6</td>
</tr>
<tr>
<td></td>
<td>1.33</td>
<td>3.27</td>
<td>11.5</td>
</tr>
</tbody>
</table>

The total investment costs for the slurry process as a function of the scale (reference scale is the pilot-plant) are depicted in figure 6. The investment costs do not increase linearly with the capacity. For a scale-up factor of 10 (from pilot to demonstration) the investment costs rise about a factor 3. From demonstration to full scale (ratio of 17), the investment costs increase by a factor 4. Compared with the often used exponential rise by a power of 0.6 for the chemical industries (Perry and Chilton, 1973), the
exponent in this case is about 0.5. The investment costs for the slurry process thus increase somewhat slower. Due to the smaller soil flow rates, the investment costs for the longer soil residence time (a-level) are lower compared to the other cases, in which larger pre- and after-treatment sections are required.

![Graph showing investment costs vs. scale-up factor for different soil residence times (2, 6, and 28 days). The graph is labeled with different phases: full, demo, and pilot.]

**Figure 6** Investment costs.

**Variable costs**

In table 5 the variable costs are depicted, again with with an inaccuracy of about 30%. Maintenance and energy costs are about the same, and each some 15% of the total variable costs. Labour costs are based on the assumption that the daily influent soil flow is achieved within 8 hours (7 days a week). This assumption makes the process semi-continuous with respect to the soil flow. However, the continuous character of the bio-degradation is not altered by this semi-continuous soil intake. Labour costs are about 70% of the variable costs.
Table 5 Variable costs.

<table>
<thead>
<tr>
<th></th>
<th>Pilot-plant (16 m³)</th>
<th>Demo-plant (160 m³)</th>
<th>Full scale (2680 m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>variable costs (in 10^6 Dfl per year)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>maintenance (equipment, piping, etc.)</td>
<td>0.034</td>
<td>0.100</td>
<td>0.310</td>
</tr>
<tr>
<td></td>
<td>0.032</td>
<td>0.090</td>
<td>0.240</td>
</tr>
<tr>
<td></td>
<td>0.030</td>
<td>0.080</td>
<td>0.200</td>
</tr>
<tr>
<td>energy (compres. (pumps, etc.)</td>
<td>0.035</td>
<td>0.120</td>
<td>0.375</td>
</tr>
<tr>
<td></td>
<td>0.030</td>
<td>0.110</td>
<td>0.360</td>
</tr>
<tr>
<td></td>
<td>0.025</td>
<td>0.100</td>
<td>0.345</td>
</tr>
<tr>
<td>labour</td>
<td>0.130</td>
<td>0.275</td>
<td>0.515</td>
</tr>
<tr>
<td></td>
<td>0.130</td>
<td>0.275</td>
<td>0.515</td>
</tr>
<tr>
<td></td>
<td>0.130</td>
<td>0.275</td>
<td>0.515</td>
</tr>
<tr>
<td>total</td>
<td>0.199</td>
<td>0.495</td>
<td>1.20</td>
</tr>
<tr>
<td></td>
<td>0.174</td>
<td>0.475</td>
<td>1.11</td>
</tr>
<tr>
<td></td>
<td>0.185</td>
<td>0.455</td>
<td>1.06</td>
</tr>
</tbody>
</table>

In Table 6 the costs per ton of treated soil are calculated from the data given in the previous tables. The yearly capital costs are derived from an estimated technical life time of 10 years for the slurry plant. In addition, it was assumed that 10 years is the maximum operating period for the slurry process within the changing market of environmental remediation technology. Forecasts made by the Dutch government in relation to soil processing are also based on a period of 10 years (VROM, 1989). The yearly capital costs are based on a life time of 10 years and an interest rate of 8 %. The capital costs are about 50-60 % of the total yearly costs.

In Table 6 it can be seen that the costs per ton decrease as scale increases. This is illustrated in Figure 7. The estimated costs per ton soil with a soil residence time of 28 days (a-level) are significantly larger than the costs at lower residence times.
Table 6 Costs per ton soil treated in the slurry process.

<table>
<thead>
<tr>
<th></th>
<th>Pilot-plant (16 m³)</th>
<th>Demo-plant (160 m³)</th>
<th>Full scale (2680 m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>costs per year (in 10⁸ Dfl per year)</td>
<td>costs per year (in 10⁸ Dfl per year)</td>
<td>costs per year (in 10⁸ Dfl per year)</td>
</tr>
<tr>
<td>yearly capital costs</td>
<td>0.217</td>
<td>0.717</td>
<td>2.60</td>
</tr>
<tr>
<td></td>
<td>0.205</td>
<td>0.537</td>
<td>2.20</td>
</tr>
<tr>
<td></td>
<td>0.205</td>
<td>0.484</td>
<td>1.60</td>
</tr>
<tr>
<td>variable costs</td>
<td>0.199</td>
<td>0.495</td>
<td>1.20</td>
</tr>
<tr>
<td></td>
<td>0.174</td>
<td>0.475</td>
<td>1.11</td>
</tr>
<tr>
<td></td>
<td>0.185</td>
<td>0.455</td>
<td>1.06</td>
</tr>
<tr>
<td>treatment capacity</td>
<td>1200</td>
<td>11,751</td>
<td>200,000</td>
</tr>
<tr>
<td>(ton/year)</td>
<td>390</td>
<td>3900</td>
<td>64,510</td>
</tr>
<tr>
<td></td>
<td>87</td>
<td>864</td>
<td>13,824</td>
</tr>
<tr>
<td>costs per ton soil</td>
<td>346</td>
<td>102</td>
<td>19</td>
</tr>
<tr>
<td>treated (Dfl/ton)</td>
<td>1017</td>
<td>258</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>4482</td>
<td>1075</td>
<td>200</td>
</tr>
</tbody>
</table>

ECONOMIC FEASIBILITY OF THE SLURRY PROCESS.

Figure 7 summarizes the economics of the biotechnological slurry process. The horizontal line indicates the present price limit (excluding transport) of 250 Dfl/ton. If treatment comes above this price, polluted soil is considered not treatable. The costs are considered too high and the only solution in that case is long term deposition. If the costs per ton treated exceed this limit of 250 Dfl/ton, the slurry process is economically unfeasible.

As shown in figure 7, there is a direct relationship between the decontamination level of the treated soil, the scale of the operation and the economic feasibility of the slurry process. It can be concluded that the pilot plant can not be operated economically.
For the two larger scales, if the b-level is acceptable, both the demo-plant and the full scale plant are feasible since the costs are around or under the 250 DFL/ton limit. However, if the a-level is to be reached, only the full scale is estimated to be feasible. Scale-up and economics are closely linked.

Considering the accuracy of the cost estimations, it should be remarked that technical feasibility studies and optimization will influence the economic position of the slurry process. In addition, the market situation for new soil remediation technologies may seriously change in the future (e.g. restrictions in waste disposal). Finally, with respect to the required decontamination standards for soil treatment processes (the dutch a-b-c reference list), developments are expected which may alter the market of soil processing in favour of biological techniques (end product quality).

CONCLUSIONS
The data presented in this paper are based on results from previous work, carried out in the framework of the development of a soil slurry process (Kleijntjens et al., 1991b and c). The following conclusions can be drawn

* Integration of the flowsheet and experimentally determined parameters leads to an adequate description of the slurry process.

* To implement the slurry process at full scale, a step-wise strategy is suggested with scale-up in three stages

  - pilot-plant (16 m³)
  - demo-plant (160 m³)
  - full scale plant (2680 m³)

* From the cost estimates, it was concluded that the economic feasibility of the slurry process is determined by the

  - maximum costs allowed
  - end concentrations required (a or b-level)
  - scale of operation

* On the basis of the data presented, the costs per ton treated soil in the demo-plant and full scale plant look promising (costs under the 250 DFL/ton limit), if the b-level is acceptable as end concentration. With respect to the a-level, only the full scale plant can be operated economically.

* For a more accurate cost estimation and to demonstrate the technical feasibility of the slurry process, the next step should be the construction of a 16 m³ pilot-plant.

ACKNOWLEDGEMENT

The authors would like to thank J. Verheul, G. Broerkens, J. Bos and G. van de Streek from DHV-consultancy engineers for the practical advices and the
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VROM, "Ten Year Scenario Soil Clean-Up", Ministry of Housing, Planning and Environment (1989)
Appendix 1

A-B-C REFERENCE LIST FOR SOIL AND GROUNDWATER CONTAMINATION
(4th revised version of the Leidraad Bodemsanering, November 1988)

Reference value (A)

Soil is considered contaminated if compounds are present at a higher concentration than might be expected on a natural basis (for that location). The concentrations for the A-level reference values given in Table 1, 2, 3 and 4, can be taken as indicator levels. If analysis demonstrates that concentrations are above this reference value, the soil is contaminated. For the compound groups 1 and 2 (Table 1), the A-level reference values correspond to the average background concentrations in the Dutch soil. Related to the soil composition, the background concentration depends on the location. It therefore may be necessary to determine the true background concentration for the location considered since this value may deviate from the average.

The A-level reference values (soil) for the compound groups 3 to 7, correspond to the detection limits in soil. Current analytical techniques are considered, using concentration techniques if necessary.

Indicative value for further investigation (B-level)

The fact that soil contamination has been demonstrated by analytical results exceeding the A-level, does not automatically imply a further investigation of the polluted site (Interim Wet Bodemsanering). Further investigation only is required when the origin, location and concentration of the pollution is a potential threat for the human health or the environment. If one or more polluting compounds exceed the B-level, this threat is considered realistic. The potential threat is to be seen within the framework of the local situation and the future use of the site.

Analytical results proving the contamination to be above the B-level point in the direction of further investigation. Below the B-level, the total
framework of the soil pollution determines whether this investigation is required.

Indicative value for clean-up (C-level)

The fact that analytical results showed the contamination level to exceed the B-level, also does not imply that remediation is to be carried out directly. However, if concentrations exceed the C-level, it is necessary to carry out (as quickly as possible) the definite analytical mapping of the site and to decide whether and which remedial action is to be undertaken. In case the C-level is not exceeded, the execution of the final analytical mapping of the site may be postponed. Remediation is not considered very urgent. The remediation and thus the final analytical mapping can be delayed to future soil cleaning programmes. Again local factors and the use of the site is of importance for the decisions to be made.

The partitioning of the 7 compound groups mentioned in the table are based on the relationship between the compounds in one class. In addition, past experience with soil contamination is incorporated in the classification. The numerical values of the concentrations given for the A, B and C level are, if possible, in line with literature. Also the mutual relationship between the compounds in one group are taken into account. Considering the A, B and C concentrations for the soil and the water compartments, the partitioning between the solid and liquid phase was taken into account.

AIMS OF REMEDIAL ACTIONS (reference value for treated soils)

Basic principle for evaluation of treated soils, is the good quality of the soil. This principle leads to the concept of multi-functionality, which means that decontaminated soils should be usable for each purpose. In practice the principle of multi-functionality leads to the A-level as reference value for soil treatment. However, if the decontaminated soil is reused as construction material, the A-level is not suitable as reference value. As shown in table 5, concentrations from half the B-level up to the C-level may be acceptable according to the "concept resolution construction material decree" of 25 april 1989.
Examination framework for several soil pollutants: indicative values for concentration levels (4th revised version Leidraad Bodemsanering, November 1988)

### Indicative values

- **A**: reference value
- **B**: indicative value for (further) investigation
- **C**: indicative value for cleaning-up

<table>
<thead>
<tr>
<th>Occurrence in:</th>
<th>Soil (mg/kg dry matter)</th>
<th>Groundwater (µg/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td><strong>1. Metals</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cr (chromium)</td>
<td>*</td>
<td>250</td>
</tr>
<tr>
<td>Co (cobalt)</td>
<td>20</td>
<td>50</td>
</tr>
<tr>
<td>Ni (nickel)</td>
<td>*</td>
<td>100</td>
</tr>
<tr>
<td>Cu (copper)</td>
<td>*</td>
<td>100</td>
</tr>
<tr>
<td>Zn (zinc)</td>
<td>*</td>
<td>500</td>
</tr>
<tr>
<td>As (arsenic)</td>
<td>*</td>
<td>30</td>
</tr>
<tr>
<td>Mo (molybdenum)</td>
<td>10</td>
<td>40</td>
</tr>
<tr>
<td>Cd (cadmium)</td>
<td>*</td>
<td>5</td>
</tr>
<tr>
<td>Sn (tin)</td>
<td>20</td>
<td>50</td>
</tr>
<tr>
<td>Ba (barium)</td>
<td>200</td>
<td>400</td>
</tr>
<tr>
<td>Hg (mercury)</td>
<td>*</td>
<td>2</td>
</tr>
<tr>
<td>Pb (lead)</td>
<td>*</td>
<td>150</td>
</tr>
</tbody>
</table>

| **2. Anorganic compounds** |   |    |    |    |    |    |
| NH₃ (as N) |     |    |    |    |    |    |
| F (total)  | *  | 400| 2,000| * | 1200| 4,000|
| CN (total-free) | 1 | 10 | 100| 5 | 30 | 100|
| CN (total-complex) | 5 | 50 | 500| 10 | 50 | 200|
| S (total sulfides) | 2 | 20 | 200| 10 | 100 | 300|
| Br (total)  | 20 | 50 | 300| * | 500 | 2,000|
| PO₄ (as P) |    |    |    | * | 200 | 700|

| **3. Aromatic compounds** |   |    |    |    |    |    |
| benzene | 0.05 (d) | 0.5 | 5 | 0.2 (d) | 1 | 5 |
| ethylbenzene | 0.05 (d) | 5 | 50 | 0.2 (d) | 20 | 60 |
| toluene | 0.05 (d) | 3 | 30 | 0.2 (d) | 15 | 50 |
| xylene | 0.05 (d) | 5 | 50 | 0.2 (d) | 20 | 60 |
| phenols | 0.05 (d) | 1 | 10 | 0.2 (d) | 15 | 50 |
| aromatics (total) | . | 7 | 70 | . | 30 | 100 |

| **4. Polynuclear aromatic hydrocarbons (PAH)** |   |    |    |    |    |    |
| naphthalene | . , . | 5 | 50 | 0.2 (d) | 7 | 30 |
| phenanthrene | . , . | 10 | 100 | 0.005 (d) | 2 | 10 |
| anthracene | . , . | 10 | 100 | 0.005 (d) | 2 | 10 |
| fluoranthene | . , . | 10 | 100 | 0.005 (d) | 1 | 5 |
| chrysene | . , . | 5 | 50 | 0.005 (d) | 0.5 | 2 |
| benz(a)anthracene | . , . | 5 | 50 | 0.005 (d) | 0.5 | 2 |
| benzo(a)pyrene | . , . | 1 | 10 | 0.005 (d) | 0.2 | 1 |
| benzo(k)fluoranthene | . , . | 5 | 50 | 0.005 (d) | 0.5 | 2 |
| indeno(1,2,3-cd)pyrene | . , . | 5 | 50 | 0.005 (d) | 0.5 | 2 |
| benzo(g,h,i)perylene | . , . | 10 | 100 | 0.005 (d) | 1 | 5 |
| PAH (total) | 1 | 20 | 200 | . | 10 | 40 |

Table 1: Indicative values for concentration levels of several pollutants in soil.
### Occurrence In:

<table>
<thead>
<tr>
<th>Substance Level</th>
<th>Soil (mg/kg dry matter)</th>
<th>Groundwater (µg/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td>5. Chlorinated Hydrocarbons</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Halogenated CHC</td>
<td><em>.</em></td>
<td>5</td>
</tr>
<tr>
<td>Halogenated CHC</td>
<td>-</td>
<td>7</td>
</tr>
<tr>
<td>Chlorobenzenes</td>
<td><em>.</em></td>
<td>1</td>
</tr>
<tr>
<td>Chlorobenzenes</td>
<td>-</td>
<td>2</td>
</tr>
<tr>
<td>Chlorophenols</td>
<td><em>.</em></td>
<td>0.5</td>
</tr>
<tr>
<td>Chlorophenols</td>
<td>-</td>
<td>1</td>
</tr>
<tr>
<td>Chlorinated PAH</td>
<td><em>.</em></td>
<td>1</td>
</tr>
<tr>
<td>PAH</td>
<td><em>.</em></td>
<td>1</td>
</tr>
<tr>
<td>EOCI</td>
<td>0,1</td>
<td>8</td>
</tr>
</tbody>
</table>

### 6. Pesticides

<table>
<thead>
<tr>
<th>Substance Level</th>
<th>Soil (mg/kg dry matter)</th>
<th>Groundwater (µg/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td>Organic chlorinated</td>
<td><em>.</em></td>
<td>0.5</td>
</tr>
<tr>
<td>Chlorinated</td>
<td><em>.</em></td>
<td>1</td>
</tr>
<tr>
<td>Non chlorinated</td>
<td><em>.</em></td>
<td>1</td>
</tr>
<tr>
<td>Non chlorinated</td>
<td>-</td>
<td>2</td>
</tr>
</tbody>
</table>

### 7. Other pollutants

<table>
<thead>
<tr>
<th>Substance Level</th>
<th>Soil (mg/kg dry matter)</th>
<th>Groundwater (µg/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td>Tetrahydrogenarson</td>
<td>0.1</td>
<td>4</td>
</tr>
<tr>
<td>Pyridine</td>
<td>0.1</td>
<td>2</td>
</tr>
<tr>
<td>Tetrahydroethylylene</td>
<td>0.1</td>
<td>5</td>
</tr>
<tr>
<td>Cyclohexane</td>
<td>0.1</td>
<td>6</td>
</tr>
<tr>
<td>Styrene</td>
<td>0.1</td>
<td>5</td>
</tr>
<tr>
<td>Phthalates</td>
<td>0.1</td>
<td>50</td>
</tr>
<tr>
<td>Oxidized PAH</td>
<td>1</td>
<td>200</td>
</tr>
<tr>
<td>Mineral oil</td>
<td>*</td>
<td>1000</td>
</tr>
</tbody>
</table>

Table 1 (cont.): Indicative values for concentration levels of several pollutants in soil

* = Reference values for heavy metals, arsenic en fluor (table 2)
* = Reference values for organic compounds in soil (table 4)
d = Detection limit

For metals marked with *, the A-value relates to the clay content (C) and/or the organic matter (O) content of the soil according to:

\[
\text{Reference value (O} = 0-2 \%) = \frac{	ext{reference value (O} = 10 \}) \times 2}{10}
\]
\[
\text{Reference value (O} = 2-30 \%) = \frac{	ext{reference value (O} = 10 \}) \times O}{10}
\]
\[
\text{Reference value (O} = 30-100 \%) = \frac{	ext{reference value (O} = 10 \}) \times 30}{10}
\]

Table 2: Reference values for heavy metals, arsenic en fluor

For organic compounds marked with *, the A-value relates to the organic compound content of the soil according to:

- Reference value (O = 0-2 %) = reference value (O = 10) \times 2
- Reference value (O = 2-30 %) = reference value (O = 10) \times O
- Reference value (O = 30-100 %) = reference value (O = 10) \times 30
<table>
<thead>
<tr>
<th>Substance</th>
<th>Groundwater</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>nitrate</td>
<td>5.6 mg Na</td>
<td>For protection of poor areas, lower values may be required</td>
</tr>
<tr>
<td>phosphate</td>
<td>0.4 mg P/l sandy areas</td>
<td></td>
</tr>
<tr>
<td>phosphates</td>
<td>3.0 mg P/l clayey and boggy areas</td>
<td></td>
</tr>
<tr>
<td>sulphate</td>
<td>150 mg/l</td>
<td>In areas with naval influence, higher values occur naturally (salt and brackish groundwater)</td>
</tr>
<tr>
<td>bromides</td>
<td>0.3 mg/l</td>
<td></td>
</tr>
<tr>
<td>chlorides</td>
<td>100 mg/l</td>
<td></td>
</tr>
<tr>
<td>fluorides</td>
<td>0.5 mg/l</td>
<td></td>
</tr>
<tr>
<td>ammonium compounds</td>
<td>2 mg Na/l sandy areas</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10 mg Na/l clayey and boggy areas</td>
<td></td>
</tr>
</tbody>
</table>

Tabel 3: Reference values for other anorganic compounds

<table>
<thead>
<tr>
<th>Substance</th>
<th>Reference values at 10 percent organic matter (O=10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) Halogenated hydrocarbons en choline esterase inhibitors</td>
<td></td>
</tr>
<tr>
<td>hexachlorocyclohexane; endrin</td>
<td>less than 10 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>tetrachloroethane; tetrachloroethylene; trichloroethane; trichloroethylene; trichloromethane</td>
<td>less than 10 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>PCB IUPAC numbers 28 and 52</td>
<td></td>
</tr>
<tr>
<td>chloropropene; tetrachloroethylene; hexachloroethane; hexachlorobutadiene; heptachloropoxide; dichlorobenzene; trichlorobenzene; tetrachlorobenzene; hexachlorobenzene; monocloronitrobenzene; dichloronitrobenzene</td>
<td>less than 10 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>aldrin; dieldrin</td>
<td></td>
</tr>
<tr>
<td>chlordane; endosulfan; trifluralin; azinphos-methyl; azinphos-ethyl; disulfoton; fenitrothion; parathion (and -methyl); triazophos</td>
<td>less than 100 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>PCB IUPAC numbers 101, 118, 138, 153 and 180</td>
<td></td>
</tr>
<tr>
<td>DDD, DDE, pentachlorophenol</td>
<td>less than 100 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>b) Polynuclear aromatic hydrocarbons (PAH)</td>
<td></td>
</tr>
<tr>
<td>naphthalene; chrysene</td>
<td>less than 10 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>phenanthrene; anthracene; fluoranthene; benzo(a)pyrene</td>
<td>less than 100 μg/kg dry matter, per substance</td>
</tr>
<tr>
<td>benzo(a)anthracene</td>
<td>less than 1 mg/kg dry matter, per substance</td>
</tr>
<tr>
<td>benzo(k)fluoranthene; indeno(1,2,3-cd)pyrene; benzo(ghi)perylene</td>
<td>less than 10 mg/kg dry matter, per substance</td>
</tr>
<tr>
<td>c) Mineral oil</td>
<td></td>
</tr>
<tr>
<td>total</td>
<td>less than 50 mg/kg dry matter, per substance</td>
</tr>
<tr>
<td>octane, heptane</td>
<td>less than 1 mg/kg dry matter, per substance</td>
</tr>
</tbody>
</table>

Tabel 4: Reference values for organic compounds in soil
(Re)use of soil according to the "concept resolution construction materials decree" of 25 April 1989

<table>
<thead>
<tr>
<th>Category</th>
<th>Concentration of pollutants (indicative)</th>
<th>Leachability of pollutants (indicative)</th>
<th>measures to be taken</th>
</tr>
</thead>
<tbody>
<tr>
<td>N1</td>
<td>* reference value soil quality (A-value)</td>
<td>N/A</td>
<td>* non, the construction material is free applicable</td>
</tr>
<tr>
<td>N2</td>
<td>* for metals and anorganic compounds (about the B-value)</td>
<td>little</td>
<td>* duty of announcement to the authorities (registration)</td>
</tr>
<tr>
<td></td>
<td>* for the other compounds, at most half of the B-value</td>
<td></td>
<td>* application has to be retractable (no mixing allowed)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>* retraction has to be guaranteed</td>
</tr>
<tr>
<td>N3</td>
<td>* for metals half of the C-value up to several times the C-value</td>
<td>moderate</td>
<td>* as N2</td>
</tr>
<tr>
<td></td>
<td>* for the other compounds, much lower than the C-value</td>
<td></td>
<td>* minimum quantity of 5000 m³</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>* application at least 50 cm above the mean highest groundwater-level</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>* isolation by means of toplayer</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>* periodical controle of isolation-provisions and groundwater quality</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>* demanding financial assurances (banking guarantee) of f 500,- per m², covering environmental responsibility</td>
</tr>
<tr>
<td>V1</td>
<td>* reference value soil quality (A-value)</td>
<td>N/A</td>
<td>* non, the construction material is free applicable</td>
</tr>
<tr>
<td>V2</td>
<td>* from half of the B-value till several times the B-value</td>
<td>little</td>
<td>* non, the construction material is free applicable</td>
</tr>
<tr>
<td>V3</td>
<td>* from half of the C-value till some times the C-value, except for PAH (B-value)</td>
<td>moderate</td>
<td>* As N3</td>
</tr>
</tbody>
</table>

Table 5: Review of categories, standards, leachability and the to be taken measures according to the "concept resolution construction materials decree" of 25 April 1989
Appendix 2

Construction aspects of the 4 m$^3$ pilot-plant shown in cross sections

[Diagram of the pilot-plant with labeled parts: sieve, settlers, pump, slurry withdrawal, reactor, injection flow, slurry injector, gas injector, first settler, second settler, slurry withdrawal.]
Appendix 3

Construction of the solids sampling probe

adjustable between 14 and 50 m

spring Ø7x0.7

sampling volume (9,42 ml)

movable lids

bar with adjustable length

handle

0-ring Ø22.5x1.5
Appendix 4

Construction of the liquid velocity sensor

\[ \text{Flow Velocity} = \frac{\text{distance probes}}{\text{residence time pulse}} \]
Appendix 5

Derivation of the sedimentation dispersion model for a tapered reactor

In the sedimentation-dispersion model two counteracting forces are considered to act on the particles in suspension, a downward gravity force resulting in the particle settlement and an upward directed diffusivity force. The upward force originates from a gradient type of transport in the suspension, defined in analogy with Fick's law of molecular transport as

$$\phi_{\text{up}} = A * ( - E_s * \frac{d \varepsilon(h)}{dh} )$$  \[A1\]

The constant $E_s$ is defined as a turbulent diffusion or dispersion coefficient. If a steady state is considered in a batch operated system the following differential mass balance in axial direction is derived in the model:

$$\frac{d(A * (- E_s * \frac{d \varepsilon(h)}{dh} ))}{dh} = \frac{d(A * Up * \varepsilon(h))}{dh}$$  \[A2\]

In equation A2 it is assumed that the solid hold-up has no radial distribution, $\varepsilon_s$ therefore only is a function of $h$. For cylindrical systems like bubble columns, the surface $A$ is not a function of the reactor height and can thus be eliminated. The resulting differential equation can be solved without difficulty as shown in equation 1.

However, for a tapered shaped system, the surface $A$ is a function of the height and can thus not be eliminated from equation A2. For the system depicted in figure 1, the relation between surface and height is given by

$$A = C_1 * D * h$$  \[A3\]

Substitution of the surface $A$ results into a homogeneous linear second order differential equation with variable coefficients and a regular singularity at $h=0$

$$\frac{d(h * - E_s * \frac{d \varepsilon(h)}{dh} )}{dh} = \frac{d(h * Up * \varepsilon(h))}{dh}$$  \[A4\]
In equation A4 both the constant $C_1$ and the depth $D$ are eliminated. In line with the standard solution of the s-d model it is assumed that both the particle dispersion coefficient $E_s$ and the settling velocity $u_p$ are independent of the height. For a dimensionless height $z$ and with the use of the $Pe$ number according to equation 2, equation A4 can be rewritten as

$$\frac{d(z \cdot \epsilon_s(z))}{dz} = - Pe_p \cdot \frac{d(z \cdot \epsilon_s(z))}{dz}$$  \[A5\]

This second order differential equation has two independent solutions, the general solution may be written as a linear combination of these two solutions

$$\epsilon_s(z) = C_1 \cdot \epsilon_s^1(z) + C_2 \cdot \epsilon_s^2(z)$$  \[A6\]

By means of substution in equation A5 it was found that one of the independent solutions can be given as

$$\epsilon_s^1(z) = \exp(- Pe_p \cdot z)$$  \[A7\]

The other solution can now be found by a combination of equation A5 and a new function $u(z)$

$$\epsilon_s^2(z) = \exp(- Pe_p \cdot z) \cdot u(z)$$  \[A8\]

To determine $u(z)$ the equation given in A8 is substituted in the differential equation A5

$$\exp(-Pe_p \cdot z) \cdot \frac{du}{dz} - Pe_p \cdot z \cdot \exp(-Pe_p \cdot z) \cdot \frac{du}{dz} + z \cdot (-Pe_p \cdot z) \cdot \frac{d^2u}{dz^2} = 0$$  \[A9\]

To solve the differential equation A9 a reduction of order is carried out by taking

$$\frac{du}{dz} = v \quad \text{and} \quad \frac{d^2u}{dz^2} = \frac{dv}{dz}$$  \[A10\]

With this substitution equation A8 can be rewritten as
\[
\frac{1}{z} \cdot \text{d}z + \text{Pe}_p \cdot \text{d}z - \frac{\text{d}v}{v} \quad [A11]
\]

This differential equation is first solved for \( v \), the equation that results has to be solved for \( u \)

\[
\text{du} = \frac{\exp(\text{Pe}_p \cdot z)}{z} \cdot \text{dz} \quad [A12]
\]

Equation A10 can be solved using a table for standard integrals

\[
u(z) = C_3 \cdot \left[ \ln(z) + \sum \frac{\text{Pe}_p^2 \cdot z^2}{n \cdot n!} \right] + C_4 \quad [A13]
\]

The second independent equation now is given by substitution of A13 in equation A7

\[
\epsilon_s^2 (z) = \exp(-\text{Pe}_p) \cdot \left( C_3 \cdot \left[ \ln(z) + \sum \frac{\text{Pe}_p^2 \cdot z^2}{n \cdot n!} \right] + C_4 \right) \quad [A14]
\]

The general solution is now given by the combination of A14 and A6 according to equation A5. The two coefficients determining the linear combination \( C_1 \) and \( C_2 \) can be determined by the two boundary conditions at \( z=0 \). One of the boundary conditions is that at \( z=0 \) a finite value of \( \epsilon_s(z) \) is physical present, the second condition is that the slope of the function describing the axial profile has a finite value \( q \) at \( z=0 \)

\[
\text{for } z = 0 \quad \epsilon_s = \epsilon_s(0) \text{ and } \frac{\text{d} \epsilon_s}{\text{d}z} = q \quad [A15]
\]

With these two boundary conditions the singularity at \( z=0 \) is solved by making the coefficient \( C_2 \) equal to zero. The coefficient for \( \epsilon_s^1(z) \) is determined by the boundary condition at \( z=0 \) the hold-up is equal to \( \epsilon_s(0) \). The final solution of the differential equation A5 is given by

\[
\epsilon_s(z) = \epsilon_s(0) \cdot \exp\left( -\text{Pe}_p \cdot z \right) \quad [A16]
\]

The final solution presented in equation A16 has the same form as the standard solution for cylindric system, this is both due to the physical
boundary conditions as well as to the limitation of the sedimentation-dispersion model. The boundary conditions restrict the form of the axial profile in such a way that at z=0 a physical realistic hold-up value is reached, the model restrictions are given in the fact that only axial phenomena are taken into account, no shape effects therefore are incorporated in the modelling.