Development of an experimental setup for microwave assisted drying of PET; a feasibility study

by

Ernst Tobias Meijer

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Student number:	4748344	
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Thesis committee:	Dr. ir. M. H. F. Sluiter,	TU Delft, supervisor
	Prof. dr. ir. S. J. Picken,	TU Delft
	Dr. ir. S. T. Abrahami,	TU Delft

TU Delft

Sustainability (*noun)*

/səˌsteɪnəˈbɪlɪti/

“Meeting the needs of the present without compromising the ability of future generations to meet their own needs.”

- United Nations Brundtland Commission, 1987

Abstract

With high global ambitions for sustainability and circularity and increasing energy prices, the recycling industry keeps innovating to keep up with political goals and customer demand. Reducing the energy consumption in the recycling process reduces environmental impact and reduces financial costs for the customer. A energy intensive process step in recycling is oven drying the plastics to an acceptable moisture content. The energy cost can be reduced by using microwave irradiation as heat source. This research investigates the moisture content and the intrinsic viscosity (IV) of polyethylene terephthalate (PET) during and after the drying process. The measurements of the PET dried in an experimental setup employing microwave irradiation as heat source is compared to the drying process in a oven at 80, 100 and 150 °C. The IV of the virgin PET pellets did not decrease significantly after drying in the experimental setup, but it did decrease after drying in the oven at 150 °C. The moisture content decreases more rapidly in the oven at all temperatures than in the experimental setup. Scaling up the experimental setup should increase the drying rate. No indications of unfeasibility of microwave assisted drying are found.

Keywords: PET (polyethylene terephthalate), Circular economy, Plastic recycling, Intrinsic viscosity (IV), Moisture content

Preface

Before you lies the master thesis “Development of an experimental setup for microwave-assisted drying of PET; a feasibility study”. It has been written to fulfill the graduation requirements of the Master Material Science and Engineering at the Technical University Delft. I did research and writing on this topic, on both this thesis and the preceding literature study, from November 2022 to March 2024.

During this graduation project, I worked three days per week at a contracting company. This left three days per week to work on this thesis and demanded me to switch between work and study frequently, which turned out to be harder than I anticipated. Some other personal challenges arose during the process as well, teaching me that I cannot do everything all at once. Sometimes I have to focus on a certain priority, which means letting go of other opportunities.

Here I would like to thank my supervisor from the TU Delft, Dr. Ir. Marcel Sluiter, for the outstanding support and guidance during the project. At some moments during the project I could not see a way out of a challenge. Whether this challenge was technical, relational or procedural, you gave me a different perspective that made the challenge surmountable. I could not have asked for better guidance.

I would like to express my gratitude to Urban Mining Corporation for making this thesis project available for me. I would also like to thank Piotr Glazer and Ouaffa Khaledkhodja for the daily supervision. Your insights in the processes around plastics recycling and your knowledge of the possibilities at Urban Mining Corporation and GETEC elevated this thesis to a higher level.

Finally, I want to thank all my friends and family members for their support, faith and patience. Thank you for always being there for me.
Here I want to make a special remark; my tertiary education lasted almost 10 years, during which I encountered numerous challenges. I could have never completed this without the extensive help from my dad, Wouter Meijer, who never ceased to believe in me and tirelessly supported me with advice and action. I am eternally grateful for his efforts and the sacrifices he and the people in his immediate surroundings have made for me. Thank you.

I hope you enjoy your reading.

Contents

1 Introduction

Plastics are everywhere in the modern world, for very good reasons. Plastics are cheap, easy to produce and durable. There are very few products that do not contain any plastic at all. All these plastic products need to be dealt with once they reach their end of life. The majority of plastic waste is dumped in the environment or burned when they reach their end of life. These are the least sustainable waste management solutions, thus plastic pollution around the world is ever increasing. One well-known example of this plastic pollution is the so-called plastic soup; a large area in the Pacific Ocean where the plastic waste has accumulated due to ocean currents to form a large garbage patch in the ocean of approximately 1.400.000 km^2. There are sustainable alternatives to deal with this plastic waste.

Recycling

To identify better waste management methods, the waste hierarchy is defined, as depicted in figure 1. Here the most preferable option is to refuse to use the material at all, avoiding the creation of waste in the first place. The second best option to reuse the product. This option is rarely feasible, because mostly when a product reaches the end of life, the material in the product is needed for a different product, in a different shape. Using the same material in a different shape is called recycling. Recycling is the only circular waste management option that provides flexibility in the shape of the material, making recycling essential in a circular economy.

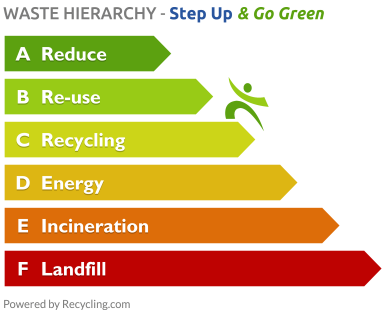


Figure 1: Waste hierarchy used by `http://www.recycling.com`

Even though reducing, re-using and recycling are the desired waste disposal methods from the perspective of a society striving for a circular economy, only 14-18% of all the waste plastics is recycled and a negligible amount is re-used. The remainder is partially incinerated and mainly disposed of in landfill [36] [34].

Plastics recycling starts with collection. Most plastics are still collected in residual waste and are hardly recycled. In the Netherlands, PET (polyethylene terephthalate) bottles are collected separately with the nudge of a deposit per bottle, resulting in a higher flow of recyclable PET and a higher purity of the PET inflow. Because of the developed character of the recycling stream of PET, this material is chosen for this research.

The process of recycling of plastics involves many steps, amongst which are several washing and drying steps to rid the recyclate from contaminants. Conventionally, drying is performed in a hot oven where forced convection of air dried with a desiccant dries the recyclate to the desired moisture content. This procedure consumes a large amount of energy. A possible energy saving improvement could be to use microwave irradiation to heat the recyclate.

Microwave dryer

The idea to apply microwave irradiation to heat PET recyclate originates from 1985 in a report by Lightsey, G.R. and Russell, L.D. with the title “Low Temperature Processing of Plastics Utilizing Microwave Heating Techniques”. This report is only known because it is used as a reference in a book from 1994, the report itself has never officially been published [21].

After 1985 subsidized research in 2006 and 2013 investigated the feasibility of using microwaves for this application [20] [31]. These papers shows promising results, potentially revolutionizing the plastic dryer industry. Some patents were filed and a few industrial machines are on the market. The expected revolution and dominance of this novel drying technique remains absent. One company, Bierther GmbH had a microwave dryer in their catalogue of 2023, but removed this product from their 2024 catalogue.

Important quality indicators in drying PET are the moisture content and the intrinsic viscosity. None of the found product specifications of the industrial microwave dryers show the effect of microwave drying on these quality indicators. Measurements of these quality indicators would be expected on such a product specification sheet. Therefore, a hypothesis to explain the unpopularity of this novel type of drying is a decrease in quality of the recyclate.

Research

To verify the hypothesis that there is a decrease in quality due to microwave assisted drying, the following research questions are posed:

- Can an experimental setup be built dries PET using microwave radiation as an energy source?
- What are the characteristics of the drying process in both a conventional oven and the experimental setup?
- Does the intrinsic viscosity change with respect to virgin PET after drying in a conventional oven or in the experimental setup?

To answer these questions, first some relevant theory is discussed in chapter 2, which facilitates a deeper understanding of the research. Then the materials and methods used to answer the research questions are explored in chapter 3. The results are given in chapter 4, followed by the discussion in chapter 5, the conclusion in chapter 6 and the recommendations in chapter 7. In the appendices additional information supporting this research can be found. Appendix I elaborates on the safety considerations for this research, Appendix II shows a separate research which was conducted to acquire insight in the production processes and Appendix III describes building the experimental setup.

2 Theory

In this section, on several relevant fields the underlying theory is given. First the chemical reactions for synthesis and degradation of polyethylene terephthalate (PET) are discussed. Then relevant material properties are elaborated upon. Some background information on microwave radiation is given, after which the heat transport is investigated by formulating the heat balance equation. The mass transport of water through a flake is modeled and the governing formula of evaporation is given. In the final section some prior developments around using microwaves to dry plastics are shown. These subjects provide an understanding of the thesis project presented.

2.1 The material PET

PET is a type of plastic mostly used for packaging and for clothing. This is mainly due to the low price, the ease of production and the suitable material properties of PET. In this chapter, the synthesis of PET will be discussed and the degradation mechanism of hydrolysis.

2.1.1 Synthesis of PET

The process of making PET starts with fossil resources. The process of making pure chemicals from crude oil is an elaborate process and the details of this process are out of the scope for this paper. Only the last chemical reactions in the production process are deemed interesting. For this paper, the starting materials for PET production will be ethylene glycol (EG), dimethyl terephthalic acid (DMT) and terephthalic acid (TPA). From these materials, there are two steps involved in PET sythesis. The first step can be either with DMT and EG in a transesterification or with TPA and EG in an esterification reaction. The result of this first step is the di-ester bis(2-Hydroxyethyl) terephthalate (BHET). BHET is then used to form PET in a polycondensation polymerization reaction that produces EG [59]. These steps are shown in more detail below.

Transesterification reaction of DMT and EG
The first path to form PET is by using DMT and EG as starting materials, these are shown visually in figure 2. In the transesterification reaction of DMT and EG, the ester DMT is reacted at low temperature with EG, producing the ester BHET and methanol as a by product.

The BHET can then be used in another transesterification reaction to produce PET. In this last step also EG is produced, which is not shown in the figure. The EG that is produced in the second transesterification reaction can be recovered and reused in the first transesterification reaction.

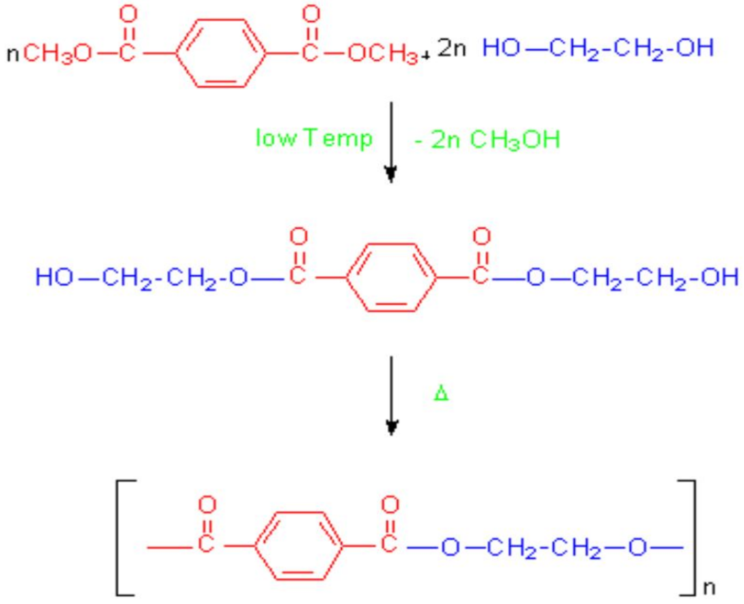


Figure 2: Reactions in transesterification of PET synthesis [29]

Esterification reaction of TPA and EG

The second path to form PET is by first reacting EG and TPA to form BHET in an esterification reaction as shown in figure 3. This reaction produces water, so it is also called a condensation reaction. It is performed at high temperature. The result of this reaction is the molecule BHET, similar to the transesterification reaction of DMT and EG.

The BHET is then used in a transesterification reaction to form PET and EG. This second step is the same reaction as seen in figure 2. Here too, the EG is separated from the PET and can be reused to form BHET in the first step.

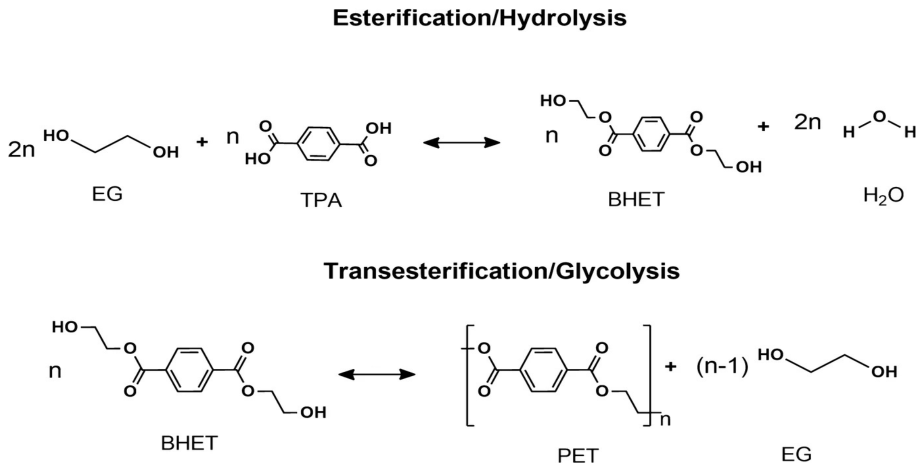


Figure 3: Reactions in esterification of PET synthesis [32]

2.1.2 Hydrolysis

Hydrolysis of PET is a reaction with water to produce the monomers terephthalic acid (TPA) and ethylene glycol (EG). This process is the reverse process of esterification as shown in figure 3. This reaction can occur in an acid, alkaline or neutral environment [19].

The process of hydrolysis accelerates at elevated temperature and pressure, as may be expected from chemical reactions. PET is injection moulded at high temperature and pressure, making hydrolysis an important failure mechanism in this process. To prevent hydrolysis during this process, the amount of water present in the PET should be minimized.

The acidity of the environment dictates the reaction path for depolymerization. In an alkaline environment, with a high pH value, the reaction path is shown in figure 4 to depolymerize the polymer. Here NaOH is used as the alkaline molecule PET reacts with. In industrial recycling processes, the PET is washed in an alkaline environment to get rid of contaminants like glue.

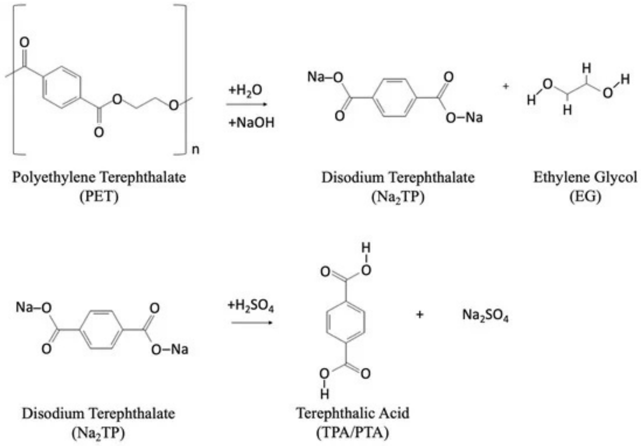


Figure 4: The alkaline hydrolysis reaction shown graphically, with NaOH reacting with the PET [22]

PET also degrades in an acidic environment, the reaction is shown in figure 5. Here the PET reacts with sulfuric acid to produce TPA and EG. The EG may react further with the acid to produce oxalic acid.

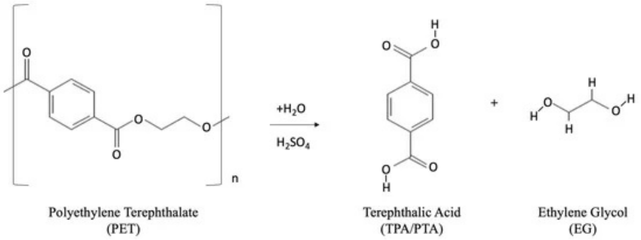


Figure 5: The acidic hydrolysis reaction shown graphically, with sulfuric acid reacting with the PET [22]

Finally hydrolysis can also occur in an environment with a neutral pH. This reaction is shown in figure 6. When hydrolysis is desired, in chemical recycling for instance, it is usually carried out at temperatures of 250-300 degrees Celsius and a pressure of 1-4 MPa. Despite the seemingly simplicity of the process, the precise chemical kinetics are not well understood. Hydrolysis in a neutral pH environment has the benefit that the produced TPA and EG do not react with the environment. However, the reaction is very slow so catalysts should be employed. Common catalysts include inorganic salts and alkali metal acetates. Also the carboxyl end-groups of TPA can have a catalytic effect. Since there are more carboxyl end-groups as the reaction progresses, the process accelerates over time [23] [52].

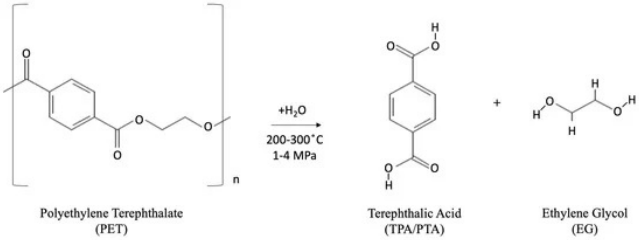


Figure 6: The hydrolysis reaction shown graphically in a neutral environment[22]

2.2 Material properties

2.2.1 Molecular weight

Molecular weight is a measure of the degree of polymerization. More polymerization means heavier molecules, thus the degree of polymerization is proportionate to the molecular weight. The degree of polymerization is the mass of the chains, divided by the weight of a monomer. Since the mass of a monomer PET (molecular formula $C_{10}H_8O_4$) is $M_0 = 192,17 g/mol$, the degree of polymerization of PET is given in equation 1.

$$DP = \frac{M_n}{M_0} = \frac{M_n}{192,17} \tag{1}$$

In the above formula, $M_n$is the number average molar mass, given in equation 2.

$$M_n = \frac{\Sigma M_i N_i}{\Sigma N_i} \tag{2}$$

There are material properties that depend on the chain length, so molecules with a higher molecular mass will have a comparatively higher contribution. This is quantified by the weight average molar mass, given in equation 3

$$M_w = \frac{\Sigma M_i^2 N_i}{\Sigma M_i N_i} \tag{3}$$

The melt elasticity is dependent on the Z-average, as given in equation 4. [64]

$$M_z = \frac{\Sigma M_i^3 N_i}{\Sigma M_i^2 N_i} \tag{4}$$

The viscosity molar mass, given in equation 5, can be determined through viscometry.

$$M_v = [\frac{\Sigma M_i^{1+a} N_i}{\Sigma M_i N_i}]^{1/a} \tag{5}$$

Here a is the exponent in the Mark-Houwink equation that relates intrinsic viscosity to number average molar mass, as described in the next paragraph.

Mark-Houwink equation

The Mark-Houwink equation is given in equation 6 [38].

$$\eta_{int} = K * M_n^a \tag{6}$$

Here η_{int} is the intrinsic viscosity of the fluid, K and a are the Mark-Houwink parameters, depending on the polymer, the solvent and the temperature and M_n is the number average molar mass. For PET dissolved in meta-Cresol at $25°C$, the values for K and a are $0,77 mL/g$ and $0,95$ respectively. [38].

2.2.2 Viscosity

Viscosity is the resistance to shear stresses in a fluid. The viscosity is one of the most important indicators of quality for polymers, for two reasons. Firstly, the machinery in factories are adjusted to exert a specific pressure to press the plastic into a product. Small deviations in the viscosity can cause the plastic to behave unpredictably, creating defects in the products. Secondly, viscosity relates to the molecular weight according to the Mark-Houwink equation from the previous paragraph. The molecular weight is an important indicator for material properties, because it is closely related to properties like strength, ductility, melt and rubber temperature and crystallinity.

For the theoretical background presented this paper, we restrict ourselves to determining the viscosity using U-tube viscometers. The experiments are conducted with an Ubbelohde, which is a specific type of U-tube viscometer. The Ubbelohde measures the time it takes for a fluid to pass through a constriction in the flow canal, this time is called the efflux time. When fluids with different concentrations of the diluted specimen are used, the measurements can be compared to find the intrinsic viscosity. The precise working mechanism of an Ubbelohde is discussed in paragraph 3.3.1.

Finding intrinsic viscosity [41] Three calculations to find the intrinsic viscosity are given. The first method involves extrapolation of the reduced viscosity and the second involves the extrapolation of the inherent viscosity. The third method calculates the intrinsic viscosity from a single concentration. The intrinsic viscosity can be used to find the number average molecular weight in the Mark-Houwink equation, given in equation 6.
In paragraph 3.3.1 a software program calculates the viscosity. These calculations are based upon the calculations given in this paragraph to find the intrinsic viscosity.

Extrapolation of the reduced viscosity
To find the reduced viscosity, first the relative viscosity and the reduced viscosity have to be found. Relative viscosity is the ratio of the viscosity of the solution and the viscosity of the solvent, which makes it a dimensionless number. This ratio can also be found by dividing the efflux time of the solution by the efflux time of the solvent, as shown in equation 7.

$$\eta_{rel} = \frac{\eta_{solution}}{\eta_{solvent}} = \frac{t_{solution}}{t_{solvent}} \tag{7}$$

The ratio of the dimensionless relative viscosity increment, more commonly known by specific viscosity, given in equation 8, and the mass concentration of the polymer in the solution is called the reduced viscosity, given in equation 9, where c is the mass concentration.

$$\eta_{spec} = \frac{\eta_{solution} - \eta_{solvent}}{\eta_{solvent}} = \frac{t_{solution} - t_{solvent}}{t_{solvent}} \tag{8}$$

$$\eta_{red} = \frac{\eta_{solution} - \eta_{solvent}}{\eta_{solvent} c} = \frac{t_{solution} - t_{solvent}}{t_{solvent} c} \tag{9}$$

The reduced viscosity has units m^3/kg and can be plotted on a straight line for different concentrations. Extrapolating the measurements for the reduced viscosity for a hypothetical solution with a concentration of 0% gives the intrinsic viscosity.

Extrapolation of the inherent viscosity

The ratio of the natural logarithmic of the relative viscosity, given in equation 7, and the mass concentration c is called the inherent viscosity, as shown in equation 10.

$$\eta_{inh} = \frac{ln(\eta_{solution})}{c} \tag{10}$$

The inherent viscosity has units m^3/kg and is linear with the concentration. Similar extrapolation as done for the reduced viscosity gives the intrinsic viscosity for a hypothetical solution with a concentration of 0%.[41] [49]

Single concentration measurement

Since every measurement costs money, the least amount of measurements to find the intrinsic viscosity is desired. A method to determine the intrinsic viscosity by performing one measurement, is by using the Billmeyer equation given in equation 11 [57]. This equation gives an approximation of the intrinsic viscosity, which forms the basis for the numerical approach for determining the intrinsic viscosity.

$$\eta_{int} = \frac{1}{4}(\frac{\eta_r el - 1 + 3ln(\eta_r)}{c}) \tag{11}$$

Values for the intrinsic viscosity of PET

Depending on the desired characteristics of the final product and the production method, PET has an intrinsic viscosity of 0,40-1,00 dl/g, as shown in table 1.

PET grade	Intrinsic Viscosity (in dL/g)
Fiber grade	0,40 – 0,70
Film grade	0,70 – 1,00
Bottle grade	0,70 – 0,78
Water and soft drink bottle grade	0,78 – 0,85

Table 1: Intrinsic viscosities of different PET grades [14]

2.2.3 Hygroscopicity

The final important material property for this research is hygroscopicity, the behaviour of a solid substance absorbing moisture from the surroundings. The more hygroscopic a material is, the more water that material attracts at a certain ambient humidity. The polar ester group can form a hydrogen bond with water molecules. This makes PET a very hygroscopic material. Moisture in the material is unwanted because this enables hydrolysis during processing. A moisture content by weight of around 50 ppm to 100 ppm is allowed. The hygroscopic properties of PET originate from the local polarity in the ester groups of PET. Water forms hydrogen bonds at these local polar regions, as shown in figure 7[61].

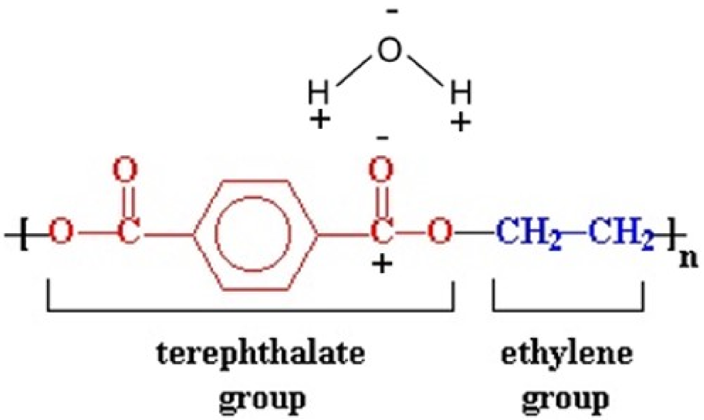


Figure 7: Local polarity of PET allows for hydrogen bond [49]

2.3 Microwave radiation

The discovery of microwave radiation as a source of heating is credited to Percy Spencer. Whilst doing tests on a new vacuum tube called a magnetron for a radar project in 1945, the chocolate bar in his pocket melted. After this initial discovery, some improvements on this technology were made until the microwave oven with a turntable was available as a countertop kitchen device in 1967.

Microwave radiation is a region of electromagnetic radiation with a wavelength in between 1 millimeter and 1 meter. This corresponds with a frequency domain of 300 MHz to 300 GHz and this puts microwave radiation in between infra red and radio waves, as shown in figure 8. There is a stubborn myth that for commercial microwave oven applications, the frequency of 2.45 GHz is mostly used because this is the frequency at which water molecules resonate, when in reality this is just the frequency approved by the FCC [60].

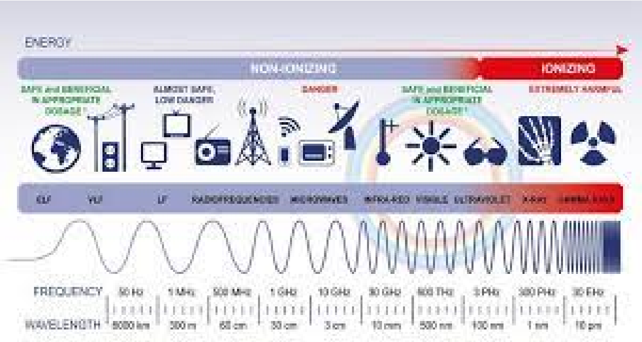


Figure 8: Electromagnetic spectrum showing a microwave between 1 and 10 GHz. Infograpic by Polina Kudelkina `http://www.polinakudelkina.com`

Electromagnetic radiation influences all dipoles and forces them to align with the radiation. It also exerts a force on ions, as shown in figure 9. The movement of these dipoles and ions is also transferred to neighbouring molecules and this kinetic energy is turned into heat. Microwave radiation is the frequency range where the influence of the radiation on molecules and ions is exploited to transfer energy and heat a product [63].

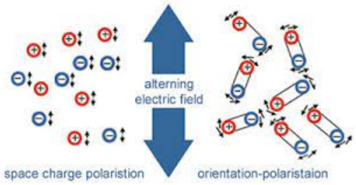


Figure 9: Excitation of dipoles and ions under microwave radiation [45]

2.4 Heat transport

To gain some insight in the heat transfer mechanisms, two systems are considered, heating in the microwave and in the oven. Each of these systems had their own characteristics, shown in table 2. The accompanying parameters for these systems are given in paragraph 3.6.

	Microwave	Oven
Type of heating	Volumetric irradiation heating	Surface convection heating
Situation at t=0	Uniform temperature	Temperature gradient
Situation at t=end	Temperature gradient	Uniform temperature
Temperature gradient	$T_{specimen} > T_{Surroundings}$	$T_{specimen} < T_{Surroundings}$

Table 2: Table with the characteristics of microwave and oven side-by-side

2.4.1 Heat balance equation

These characteristics give rise to the heat balance equation given in equations 12 and 13.

$$\begin{aligned}\dot{Q}_{Microwave} = \dot{Q}_{Microwave\_irradiation} + \dot{Q}_{Convection} \\ + \dot{Q}_{Temperature\_radiation} + \dot{Q}_{Latent\_heat\_evaporation}\end{aligned} \tag{12}$$

$$\dot{Q}_{Oven} = \dot{Q}_{Convection} + \dot{Q}_{Temperature\_radiation} + \dot{Q}_{Latent\_heat\_evaporation} \tag{13}$$

In the following paragraphs, the microwave , convection and temperature radiation will be elaborated upon. In section 2.5 the mass transport will be discussed in more detail. Here calculations are given that show that $\dot{Q}_{Latent\_heat\_evaporation}$ does not contribute significantly to the heat balance.

2.4.2 Microwave

To find the absorbed microwave power, the contributions of the PET and the water absorption are found and added together proportionally. In this paragraph calculations on the radiation losses in wet PET will be done at microwave frequencies of 100 MHz and 1 GHz. The amount of energy absorbed by the wet PET can be determined by evaluating the radiation losses in the material under microwave irradiation.

To find the radiation losses, the dielectric loss has to be calculated. The dielectric loss refers to the energy dissipated as heat when an alternating electric field is applied. The dielectric loss can be found by evaluating the difference between the power of the electromagnetic radiation incident on the material and the power of the electromagnetic radiation exiting the material. This can be calculated by using an equation derived from the Beer-Lambert law, as given in equation 14 [42].

$$P(d) = P_0 e^{-k*tan(\delta)d} \tag{14}$$

This equation can be rewritten to find the ratio of the power incident and exiting the material. This ratio is useful to find the power loss in the material for different amounts of electromagnetic power incident on the material. This equation is given in equation 15.

$$\begin{gathered} P(d) = P_0 e^{-k*tan(\delta)d} \\ P(d)/P_0 = e^{-k*tan(\delta)d} \end{gathered} \tag{15}$$

Here P_0 is the power incident on the material, k is given by $k = 2 * \pi * f/c$, d is the thickness of the material and $tan(\delta)$ is the ratio between the imaginary and the real part of the permittivity of the material. This means that $tan(\delta)$ is the ratio between power dissipated and power lost in other ways, like reflection.

To find the $tan(\delta)$, first the $tan(\delta)$ is found for both PET and tap water at 25 and 100 degrees at 100 MHz and 1 Ghz. Then the volume weighted average, as shown in formula 16 is taken between $tan(\delta)_0$ and $tan(\delta)_1$, which are the $tan(\delta)$ values for the wet PET at 25 degrees Celsius and the dried PET at 100 degrees Celsius respectively. To find the $tan(\delta)$ for tap water, figures 10 and 11 and the equation $tan(\delta) = \omega\epsilon" + \sigma/\omega\epsilon'$ is used. Since σ is a factor 12 to 14 smaller than $\omega\epsilon"$, the equation simplifies to $tan(\delta) = \epsilon"/\epsilon'$. The $tan(\delta)$ for PET can be found in figure 12. Here it must be noted that these values for $tan(\delta)$ are found at room temperature, no data for PET at 100 degrees Celsius could be found. The calculated values of $tan(\delta)$ for tap water are similar to the values of $tan(\delta)$ for deionized water. The found values for $tan(\delta)$ are summarized in table 3.

Frequency of radiation [Hz]	Temperature (Celsius)	$tan(\delta)_{water}$	$tan(\delta)_{PET}$
$1 * 10^8$	25	0.051	0.017
	100	0.27	0.017
$1 * 10^9$	25	0.032	0.017
	100	0.080	0.017

Table 3: Table of $tan(\delta)$ of water and PET at 25 and 100 degrees Celsius at 100 MHz and 1 GHz as found in figures 12 and 10 [35] [46]

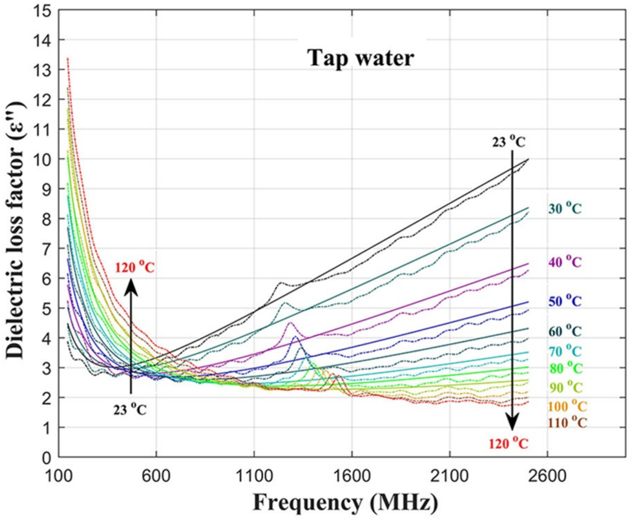


Figure 10: Dielectric loss of tap water in the microwave spectrum, with the temperature range of 23 - 120 degrees Celsius [35]

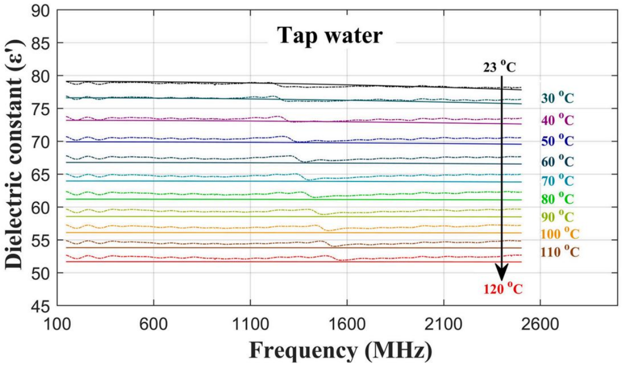


Figure 11: Dielectric constant of tap water in the microwave spectrum, with the temperature range of 23 - 120 degrees Celsius [35]

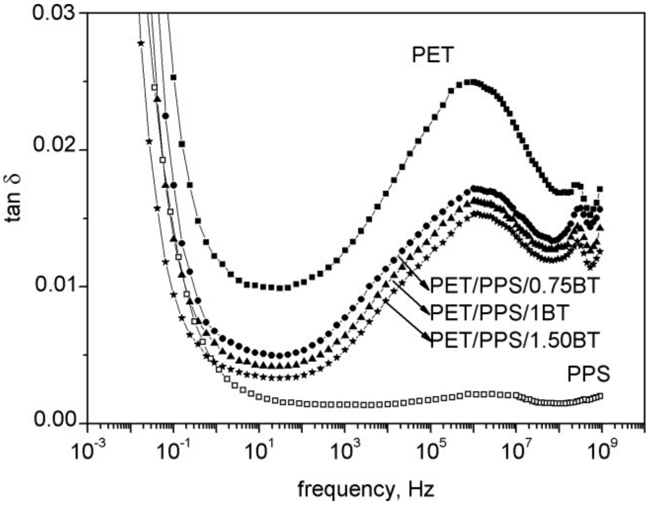


Figure 12: Loss tangent of PET at room temperature in the microwave spectrum, shown as the bottom line in this graph [46]

The volume weighted average will be taken, as described in equation 16. Using the values of table 3, table 4 can be filled.

$$tan(\delta)_{\#} = \frac{tan(\delta)_{water}V_{water} + tan(\delta)_{PET}V_{PET}}{V_{tot}} \tag{16}$$

Frequency of radiation [Hz]	$tan(\delta)_0$	$tan(\delta)_1$	$tan(\delta)$
$1 * 10^8$	0.018	0.017	0.018
$1 * 10^9$	0.017	0.017	0.017

Table 4: Calculated values for $tan(\delta)$ at 100 MHz and 1 GHz. Here it should be noted that $tan(\delta)_0$ is at 3% water and 25 degrees Celsius and $tan(\delta)_1$ is at 0.1% water and 100 degrees Celsius

With the found values for $k = 2 * \pi * f/c$ and $tan(\delta)$, equation 15 can be solved for different thicknesses of the material. It is solved for the thicknesses of 0.01 m, 0.1 m, 1 m and 10 m to gain insight in the heating process. The results are shown in table 5.

In table 5 also $1 - P(d)/P_0$ is given, because this gives the ratio between the power incident on the material and the power dissipated in the material. It is observed that for a microwave frequency of 100 MHz the PET is mostly transparent. Even for a thickness of 10 meters only 31% of the radiation is absorbed by the material. For a frequency of 1 GHz 97% of the radiation is absorbed by a material with a thickness of 10 meters. Here it should be noticed that these found values are not to be interpolated or extrapolated. As can be seen from figure 10, the $tan(\delta)$ of tap water does not behave predictably as the

f [Hz]	$tan(\delta)$ [ ]	d [m]	$P(d)/P_0$ [ ]	$1 - P(d)/P_0$ [ ]
$1 * 10^8$	0.018	0.01	1	0
		0.1	1	0
		1	0.96	0.04
		10	0.69	0.31
$1 * 10^9$	0.017	0.01	1	0
		0.1	0.96	0.04
		1	0.70	0.30
		10	0.03	0.97

Table 5: Calculated values for E_{avg}/E_0 at 100 MHz and 1 GHz with thicknesses of 10^{-2} m, 10^{-1} m, 10^0 m and 10^1 m

frequency increases or decreases. The observed behaviour is expected to be due to the ionic response of the ions still present in the tap water at lower frequencies. At higher frequencies and for deionized water, the $tan(\delta)$ is expected to behave more predictably.

Based on these calculations an assumption is made on the amount of microwave absorbed by the PET in the bowl. The results are not extrapolated, rather it is observed in figure 10 that the microwave absorbance of water increases at higher frequencies. The assumption on the amount of microwave is most likely within one order of magnitude inaccurate, future experiments could evaluate this hypothesis. A $1 - P(d)/P_0$ factor of $0,01$ is assumed for microwave energy absorption at $2,45GHz$ with the penetration depth present in the bowl. With a microwave power of $100W$, this leads to an estimated average microwave power absorption of $1W$.

2.4.3 Convection

The experiments in this research are conducted in an oven without forced convection. In industry, the PET is heated in an oven with forced convection, allowing all sides of the PET flake or pellet to be exposed to the hot air.

Airflow In both the oven and the microwave, the airspeed is estimated at $0,05 m/s^2$, see paragraphs 3.6.3 and 3.6.2. At low air speeds, it is not clear if natural convection or forced convection is the dominant mode. Characterization of the type of convection is necessary to find the governing equations for the system. Natural convection is characterized by the Grashof number Gr_L, which represents the ratio of buoyant forces to viscous forces, given in equation 17.

$$Gr_L = \frac{g\beta|T_s - T_\infty|L_c^3}{v^2} \tag{17}$$

Where
g = gravitational acceleration in m/s^2
β = coefficient of volume expansion in $1/K$. (β = 1/T for ideal gases)
T_s = temperature of the surface in $^\circ C$
T_∞ = temperature of the fluid sufficiently far from the surface in $^\circ C$
L_c = characteristic length of the geometry in m
v = kinematic viscosity of the fluid in m^2/s

The ratio of the Grashof number to the Reynolds number squared characterizes the airflow as forced, mixed or natural convection, as shown in equation 18 [24].

$$Gr_L/Re_L^2 \begin{array}{ll} << 1 & \text{, forced convection} \\ \approx 1 & \text{, mixed convection} \\ >> 1 & \text{, natural convection} \end{array} \tag{18}$$

Combining equations 17 and 18 gives equation 19.

$$Gr_L/Re_L^2 = \frac{g\beta|T_s - T_\infty|L_c}{u^2} \tag{19}$$

Using the parameters from paragraph 3.6 in equation 19, we find that for a microwave the dimensionless number Gr_L/R_L^2 is approximately 26,7 and for oven temperatures ranging from $80-150^\circ C$ this number ranges from $20,0-36,2$. Since these values for Gr_L/R_L^2 all exceed 10, the convection is best described by forced convection [25].

Forced convection Only the values for the oven at 80° C are used in this paragraph to gain insight in the convection mechanism for the situation with the smallest temperature difference. External forced convection is governed by the Reynolds, Prandtl and Nusselt numbers. The maximum Reynolds numbers in the microwave and in the oven, using the values from paragraph 3.6 are 390 and 500 respectively. Since this is well below the onset of turbulent airflow at a Reynolds number of $5 * 10^5$, the flow is laminar. Since the Prandtl number for the oven and the microwave are $0,69$ and $0,68$ respectively, the average Nusselt number is then found by equation 20 to be $11,6$ and $13,1$ respectively. These Nusselt numbers are associated with slug, laminar flow, which is in accordance with the experimental setups [12]. With the lengths of $0,16m$ and $0,15m$ and values of k from paragraph 3.6, gives convection heat transfer coefficients of $2,18$ and $2,64$ $W/(m^2K)$ [26].

$$Nu_{avg} = \frac{hL}{k} = 0,664 Re_L^{0,5} Pr^{1/3} \tag{20}$$

Using the found values for the convection heat transfer coefficient and Newton's law of cooling given in 21, the heat transfer through convection is $-2,5W$ and $2,4W$ for the microwave and the oven respectively. The small difference in convective heat transfer is expected, because in both systems forced convection with a temperature difference of approximately $60°C$ occurs.

$$\dot{Q}_{Convection} = hAdT \tag{21}$$

2.4.4 Temperature radiation

Assuming blackbody radiation, heat transfer through infrared radiation is dictated by the temperature difference, as given in equation 22 [27].

$$\dot{Q}_{Temperature\_radiation} = \sigma A(T_1^4 - T_2^4) \tag{22}$$

Using the Stefan-Bolzmann constant $\sigma \approx 5,670 * 10^{-8}$ and the values given in paragraph 3.6, the heat transfer through temperature radiation is $-8,9W$ and $6,9W$ for the microwave and the oven respectively. The negative value of the heat transfer for the microwave means that the specimen loses heat through convection, whereas the specimen gains energy through convection in the oven.

2.4.5 Conclusion

The calculations presented show that irradiation contributes more to the heat transfer than heat transfer through convection. This surprising conclusion is mainly due to the low airflow resulting in laminar flow. With a higher airspeed, the airflow would be turbulent, leading to higher convective heat transfer. The combined heat loss in the microwave is approximately $-2,5W - 8,9W = -11,4W$. This is an order of magnitude larger than the expected microwave absorption of $1W$. Possible explanations are discovered in chapter 5.

2.5 Mass transport

In the drying process, two types of mass transport are expected. Firstly the the diffusion of the water through the PET and secondly evaporation of the water in the air. These describe the mass transport of the absorbed and the adherent water respectively.

2.5.1 Diffusion

In general, nonsteady-state diffusion is described by Fick's second law of diffusion 23. This describes the change in concentration over time $\frac{\delta C}{\delta t}$ as a function of the change in the spatial concentration gradient over space $\frac{\delta^2 C_A}{\delta x^2}$, multiplied by the diffusion coefficient $D_{AB}(inm2/s)$. The change in concentration over time can be used to find the mass transport.

$$\frac{\delta C}{\delta t} = -D_{AB}\frac{\delta^2 C_A}{\delta x^2} = D_{AB}\nabla^2 C \tag{23}$$

Here the diffusion coefficient D_{AB} is highly dependent on the crystallinity of the PET [56]. Based on values found in literature, a diffusion coefficient of $5 * 10^{-12} m^2/s$ is assumed [48] [39].

To gain insight in the diffusion mechanism, the differential equation is modeled for a flake. The flake is modeled to have the same thickness as a PET bottle, which is assumed to be $5 * 10^{-4} m$. The flakes are so thin compared to their other dimensions, that the differential equation reduces to a one-dimensional problem. Diffusion in other directions than this dimension is negligible. Due to symmetry, $x = 0$ at the middle of the flake and $x = x_{max}$ at the surface. It is assumed that there is no water in the surrounding air. Initially, the specimen is assumed to be saturated with absorbed water, at 3000 ppm. These boundary conditions and the initial condition are given in 24.

$$\begin{aligned} \text{Boundary condition 1:} \quad & \frac{\delta C(0,t)}{\delta x} = 0 \\ \text{Boundary condition 2:} \quad & C(x_{max}, t) = 0 \\ \text{Initial condition:} \quad & C(x, 0) = C_i = 0,003 \end{aligned} \tag{24}$$

Equation 23 with boundary conditions presented in 24 is modeled for a time range of $7200s$, see figure 13.

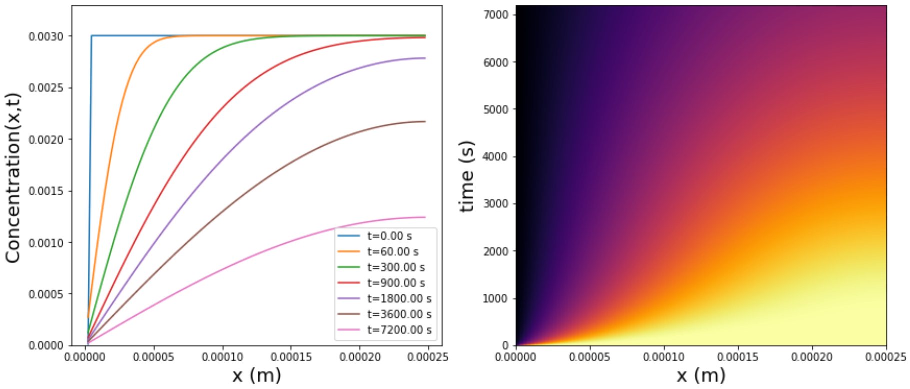


Figure 13: Model of the mass diffusion through the flake with a thickness of 0,5 mm

2.5.2 Evaporation

The evaporation of water is an endothermic process, requiring energy for the latent heat of vaporization. The amount of heat needed to evaporate water is given in equation 25. Here M is the molar mass of water of $18,0 kg/mol$ and Δh is the enthalpy of vaporization for water. In figure 14 the enthalpy of vaporization at $80°C$ ($\approx 350K$) is approximately $42 * 10^3 J/mol$.

$$\dot{Q}_{Latent\_heat\_evaporation} = \frac{\dot{m}}{M} \Delta h \tag{25}$$

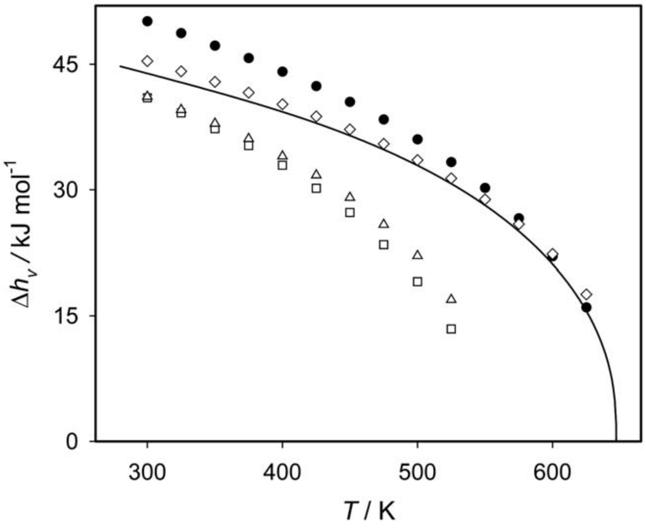


Figure 14: Enthalpy of water evaporation at different temperatures

Per kg PET $3g$ of water is absorbed. Assuming all water is evaporated at $80°C$, the total energy required to overcome the latent heat of evaporation is $7J$. Assuming all water evaporates over a time of $2hours$, the average power needed for evaporation of the water in $1kg$ PET is $0,001W$, which is negligible with respect to the powers in the heat balance equation for the microwave as can be seen in paragraph 2.4.5. Also, since the evaporated water has the same temperature as the bulk, there is no heat transfer through mass transport, as can be seen in the heat equation 26, where $\Delta T = 0$. For these two reasons, the evaporation is no factor in the heat balance equation

$$\dot{Q}_{Heat\_energy} = mc_p\Delta T \tag{26}$$

It should also be noted that when water evaporates, the water content of the specimen is lowered. This decreases the amount of microwave irradiation absorbed by the bulk, because there is less absorbent material. The amount of energy transferred by microwave irradiation is calculated in 2.4.2, where assumptions are made to accommodate for this loss in absorbent material.

2.6 Prior developments

The use of microwave radiation to dry PET is interesting, because using microwaves instead of hot air would decrease the energy required to dry the PET flakes and it would also speed up the process. These possible benefits have been identified as early as 1985 in a report by Lightsey, G.R. and Russell, L.D. with the title “Low Temperature Processing of Plastics Utilizing Microwave Heating Techniques”. This report is only known because it is used as a reference in a book from 1994, the report itself has never officially been published [21]. Since this principle has first been mentioned, multiple efforts have been made to integrate this technology in the market. First the projects that directly or indirectly received funding from the EU are discussed. Then the patents on this subject were investigated.

2.6.1 Subsidized research

The first EU backed research was under the name “Polydry” and took place from 2004 until 2006. Here the goal was “to develop a new low cost, rapid, energy efficient microwave based moisture measurement and drying system” [20]. The final product was a working concept model and a recommendation for further research and investigation. Most of the participating companies still exist, some of them apply microwaves in drying processes, other make products for drying polymers, however none of them apply microwave-based drying for the drying of polymers. No remarks towards this research could be found on any of the websites. The website of the research itself has been pulled offline and was unreachable on 20th February 2023. No follow-up on this research could be found.

The second EU backed research in this domain was done under the name “HiPerDry” and took place from 2013 until 2016. This project had two aims, as given below [31];

- to extend the knowledge to overcome technical barriers associated with drying of hygroscopic polymers, especially those derived from bio-based materials, establishing sustainable production patterns across the European plastic processing industry
- to develop a novel microwave-enhanced superheated steam process in continuous operation for high performance drying of hygroscopic polymers, which has a promising potential to help strengthen the competitiveness of European plastics processing SMEs through the improved energy efficiency permitting up to 50% reduction in energy consumption compared to the current state-of-the-art systems.

Even though the report states “A widespread dissemination of the project was ensured mainly through the SME-AGs.”, HiPerDry is mentioned nowhere else, besides one publication in a Spanish journal [50]. Also a deliverable with the title “Best Practice Guideline to Drying of Hygroscopic Polymers” is said to be published, but could not be found. The website of the research itself has been pulled offline and was unreachable on 20th February 2023. On none of the participating member's website anything related to this project could be found. However, one of the participating members of this consortium, Bierther, has produced a microwave dryer which is commercially available. After this project has finished, three research papers have been published by utilizing the equipment made in this project [44] [43] [58].

Here it should also be noted that there is a third EU-backed research in the field of drying polymers using microwaves, however this research focuses on drying elastomers after the products were made, to get rid of the water that has been produced in the condensation reaction in the production process. This research is not directly subsidized by the EU, it is funded by a cluster of companies which receives support from the EU [47]. Very limited information on this project has been found, no affiliated people or participating organizations have been identified. A company by the name of "Motzener Kunststoff- und Gummiverarbeitung" has been linked to this research. This company is said to "make considerable investments into microwave technology", however it shows no evidence of these investments on their website.

2.6.2 Patents

The first patent on the subject of drying polymers using microwaves was published in 1990 with patent number EP0312741 [53]. Two designs proposed by this patent are shown in figure 15. Interestingly, this patent is deemed to be withdrawn before taking effect in the European database, because "[the] reply to the communication from the examining division [was] not received in time, [date] [1992/23]". This patent appears to be a basic design and no use of the patent in this time could be found.

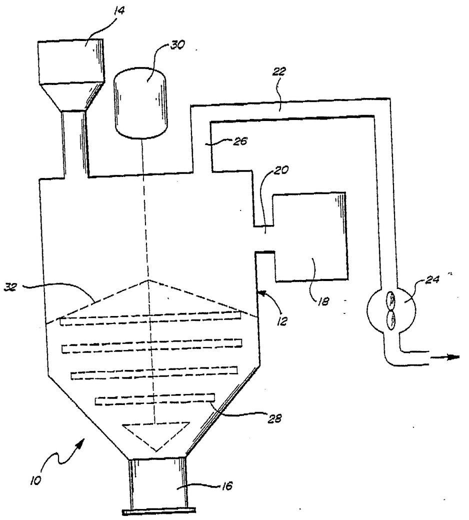


FIG. 1

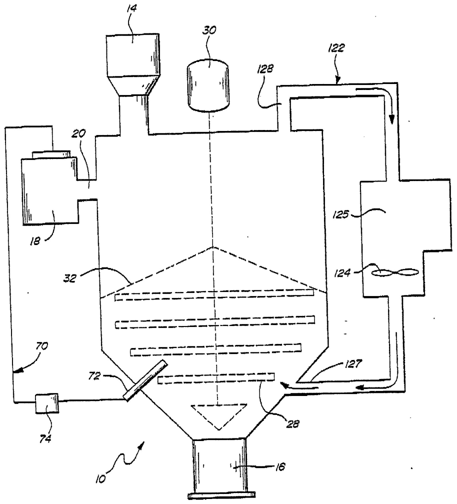


FIG. 2

Figure 15: Figures of two proposed designs as proposed by the first patent using microwaves to dry plastics with patent number EP0312741 [53]

In 2022 SMC patented a microwave based dryer, with patent number IT202000005476U1 [54]. the accompanying figure is shown in figure 16. The big difference between this patent and the patent from 1990 is that here the microwave is placed outside the hopper instead of inside the hopper. SMC has four different models utilizing this technique in their catalogue, when the website was visited on the 20th of February 2023. Here it is interesting to note that SMC has not participated in any of the EU-backed research.

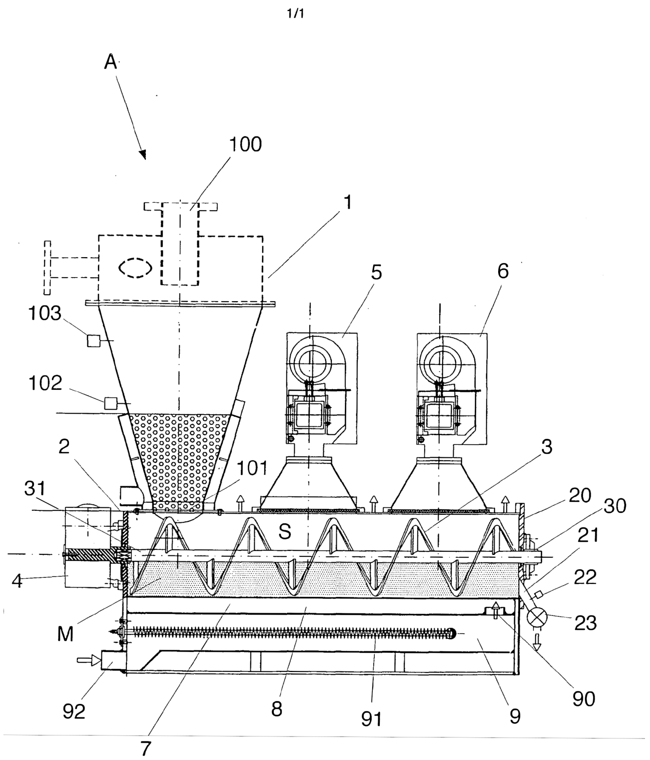


FIG 1

Figure 16: Figure of the patent from SMC with patent number IT202000005476U1, applying microwaves outside of the hopper [54]

3 Materials and methods

3.1 Experimental setup

To test the effect of microwave irradiation on wet PET, a research setup was designed, built, tested and used which allows drying of PET with microwave irradiation. In this section the essential decisions in building, testing and using are summarized. For insight in the lessons learned in the iterative process, see appendix III.

3.1.1 Building the setup

The setup had to fulfill certain demands;

- All electronics work on the standard power outlet of 230 V
- Controllable amount of microwave irradiation on the specimen
- Microwave has a timer that automatically turns the microwave off
- The microwave must be closed off from ambient air
- Possibility to saturate the microwave inlet with dry air, by using a desiccant
- Desiccant can be replaced easily when it is saturated with water
- Closed-loop and open-loop configuration are easily interchangeable
- Thermo-hygrometer to measure the temperature and humidity of the air at both the air inlet of the microwave and the air outlet
- Setup must be robust, only minimal or easily repairable damage are acceptable after careful transport
- Heat and fire resistant building materials must be used to minimize the effect of a potential calamity
- Setup must be safe to use (no sharp edges, minimal exposure to microwave radiation...)

Wishes for the setup were identified too:

- The setup should be cheap
- The setup should be made of off-the-shelf materials
- The setup should be sustainable in design and use
- The setup should be usable for future research
- The setup should be easily modifiable
- The setup should look and feel professional, no “duct tape and cardboard”

- Data collected from the thermo-hygrometer should be easily accessible and easy to read out
- The setup should be a table-top model
- The setup should be easy to build

The above demands and wishes resulted in the design shown schematically in figure 17. Some optimizations on this schematic design are performed in the final design. The microwave used is the AEG MSB2547D-M [1]. The thermo-hygrometer is the TFA 009 Thermo-hygrometer [17], which is placed inside the dryer, where it can either be placed at the air inlet or the air outlet. The desiccant chosen in the dryer is silica gel [15].

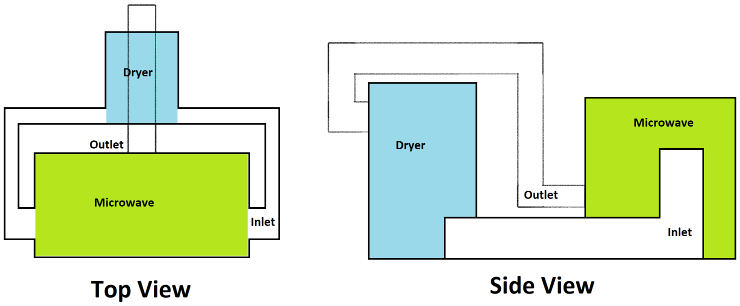


Figure 17: Schematic figure of the research setup

In building the setup, the appropriate Personal Protective Equipment (PPE) is used, as shown in appendix I. After disassembling the microwave, the desired path for the airflow was determined. The excessive holes were closed off and structures were placed inside the hull of the microwave to guide the airflow in the desired path. These structures were moulded from silicone putty Stensil ECO 70 Blue [16] and are shown in figure 18.

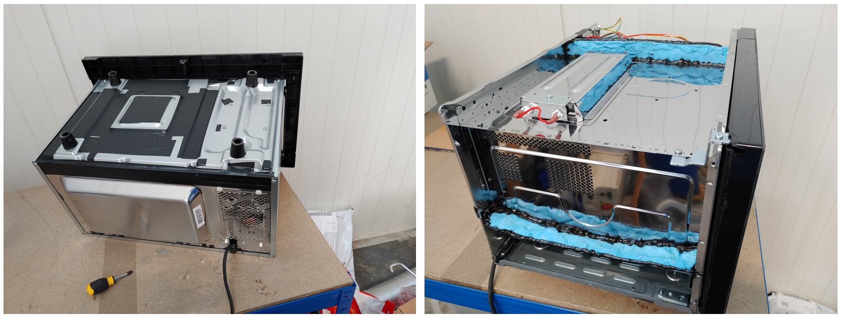


Figure 18: Blocking excessive holes in the microwave and blocking unwanted airflow with silicone putty structures

A large storage box is used to hold the desiccant. The large volume makes sure air remains in this desiccant dryer for an extensive amount of time. The removable lid allows for easy access to the desiccant and the hygrometer. Silica gel is put in bags made out of chicken wire, allowing the bags to be easily regenerated in the oven. These builds are shown in figure 19.

Figure 19: Building the dryer and the desiccant. In the first figure a different desiccant, Portland cement, can be seen, which was an earlier iteration of the design. See appendix III for more information

The microwave outlet is attached to one end of the dryer and the microwave inlet is attached to the other end, as shown in figure 20. An open-loop configuration can be achieved by opening the lid of the desiccant dryer. With the pipes attached to the microwave and the dryer, the research setup is complete.

Figure 20: Finished research setup from two angles, showing both the microwave plug and the thermo-hygrometer data cable

3.1.2 Testing the setup

In testing the setup, the appropriate Personal Protective Equipment (PPE) is used and the appropriate safety measures are taken, as shown in appendix I. The results of the tests are that the setup and all devices work as intended, it should be operated in closed configuration, the desiccant material should be silica gel and virgin PET pellets are used as specimen. In section III.2 of appendix III, the process of testing the setup is elaborated upon in more detail.

3.1.3 Conducting experiments

The specimens were made from virgin PET pellets. This ensured that the composition was known and did not contain any fillers. For more explanation on why virgin PET pellets were chosen, see appendix III.

Urban Mining Corporation experiences challenges with fines that are produced in the recycling process. These fines are lower in value than the bulk of the plastic and are therefore to be avoided. Additional research to the source of these fines and methods to avoid their production is conducted and is added to appendix II of this paper.

The setup is always used in closed configuration, as is explained in appendix III. Before each test, the dryer is opened to check if the coloured silica gel balls are still bright orange, indicating a saturation of less than 50 %.

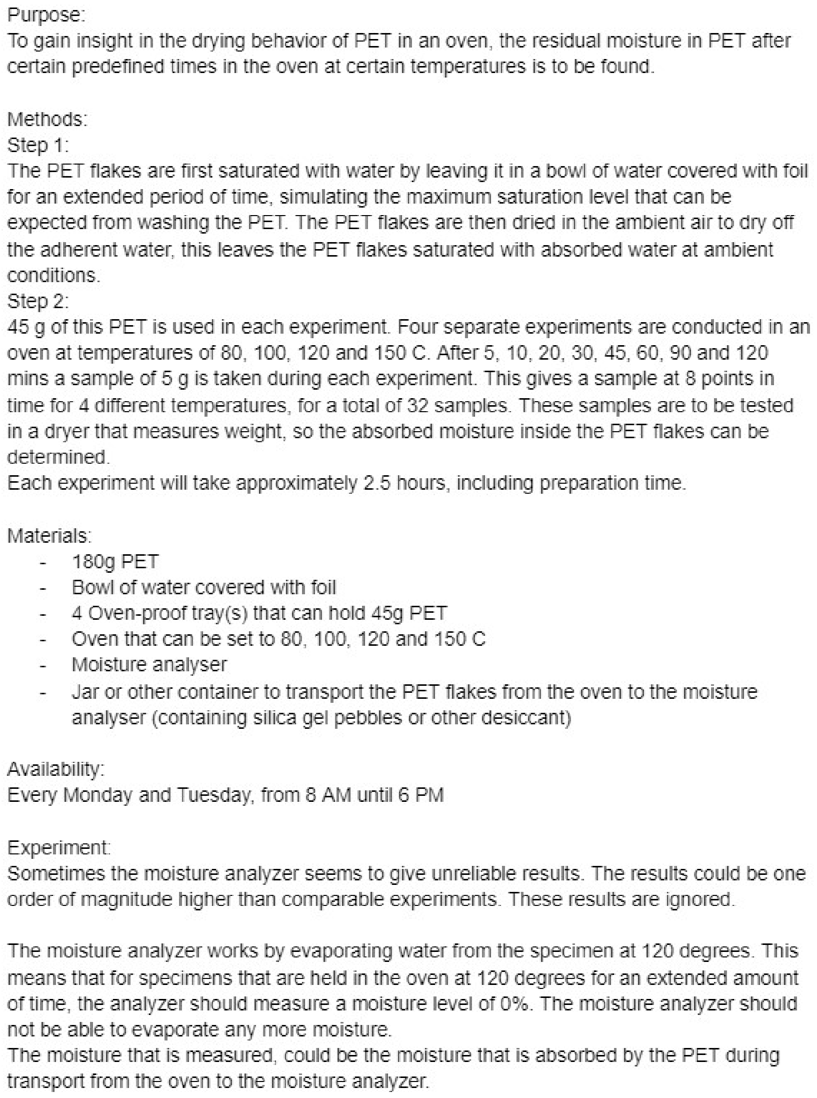
The microwave in the experimental setup has a power output of 100 W for 50 g (+/- 1g) of PET in a ceramic bowl, resulting in a specific power of ≈ 2000 W/kg PET, and a specific energy added to the system of $1,44 * 10^7$ J/kg PET after an experiment with a duration of 120 minutes. Two industrially available microwave dryers, by the companies Bierther and SMC, both have a throughput of 1000 kg/hour. The power output of these machines is 94 kW and 101 kW respectively. This results in a specific energy of $3,4 - 3,610^5$ J/kg PET. This means that the specific energy of the experimental setup is approximately 60x higher than the specific energy of an industrial dryer.

In the industry mostly dryers using hot air are used. To mimic these dryers, an oven is used to dry the PET at different temperatures, for different amounts of time.

3.2 Materials

3.2.1 Oven

The oven used has a temperature resolution of 1 °C. The oven can reach temperatures of 200 °C. The generic research plan for drying experiments in the oven is given in figure 21.



Purpose:
To gain insight in the drying behavior of PET in an oven, the residual moisture in PET after certain predefined times in the oven at certain temperatures is to be found.

Methods:
Step 1:
The PET flakes are first saturated with water by leaving it in a bowl of water covered with foil for an extended period of time, simulating the maximum saturation level that can be expected from washing the PET. The PET flakes are then dried in the ambient air to dry off the adherent water, this leaves the PET flakes saturated with absorbed water at ambient conditions.
Step 2:
45 g of this PET is used in each experiment. Four separate experiments are conducted in an oven at temperatures of 80, 100, 120 and 150 C. After 5, 10, 20, 30, 45, 60, 90 and 120 mins a sample of 5 g is taken during each experiment. This gives a sample at 8 points in time for 4 different temperatures, for a total of 32 samples. These samples are to be tested in a dryer that measures weight, so the absorbed moisture inside the PET flakes can be determined.
Each experiment will take approximately 2.5 hours, including preparation time.

Materials:
- 180g PET
- Bowl of water covered with foil
- 4 Oven-proof tray(s) that can hold 45g PET
- Oven that can be set to 80, 100, 120 and 150 C
- Moisture analyser
- Jar or other container to transport the PET flakes from the oven to the moisture analyser (containing silica gel pebbles or other desiccant)

Availability:
Every Monday and Tuesday, from 8 AM until 6 PM

Experiment:
Sometimes the moisture analyzer seems to give unreliable results. The results could be one order of magnitude higher than comparable experiments. These results are ignored.

The moisture analyzer works by evaporating water from the specimen at 120 degrees. This means that for specimens that are held in the oven at 120 degrees for an extended amount of time, the analyzer should measure a moisture level of 0%. The moisture analyzer should not be able to evaporate any more moisture.
The moisture that is measured, could be the moisture that is absorbed by the PET during transport from the oven to the moisture analyzer.

Figure 21: Research plan for tests in the oven

3.2.2 PP container

To transport the dried PET, a container has to meet the following requirements.

- Made from a material impenetrable by water
- Cheap
- Easy to transport
- Lightweight
- Closes airtight

The container that meets these requirements is a PP container with a screw lid. Even though it says in the specifications that this container is not airtight, experiments submerging these containers in water showed that no water entered the containers. [18]

3.2.3 IR thermometer

To measure the temperature in the microwave, an IR-thermometer is used. The IR thermometer has a resolution of $0,1°C$ and an accuracy of $\pm 1,5°C$, so the measurements are rounded off to the nearest degree. [10]

3.3 Determining material properties

3.3.1 Viscometry using an Ubbelohde

Relative viscosity An Ubbelohde, shown in figure 22, is a device that measures the relative viscosity of a fluid by measuring the time it takes for a fixed amount of fluid to pass through a narrow capillary. To use it, first the reservoir (6) is filled through the filling tube (9) until the fluid level is in between the filling marks (7). The venting tube (2) and the filling tube are closed, allowing the fluid to be sucked up through the capillary tube (1), into the feeding bulb (3). When all the tubes are opened, the fluid flows out of the feeding bulb, through the timing bulb (4), through the capillary (8), back into the suspended level bulb (5). The time it takes for the fluid to pass through the timing bulb is measured, by either manually of automatically starting a timer when the fluid passes by the first timing mark (M1) and stopping the timer when it passes by the second timing mark (M2). This measurement is conducted for different concentrations and for the pure solvent, allowing the analysis described in paragraph 2.2.2 to be conducted.

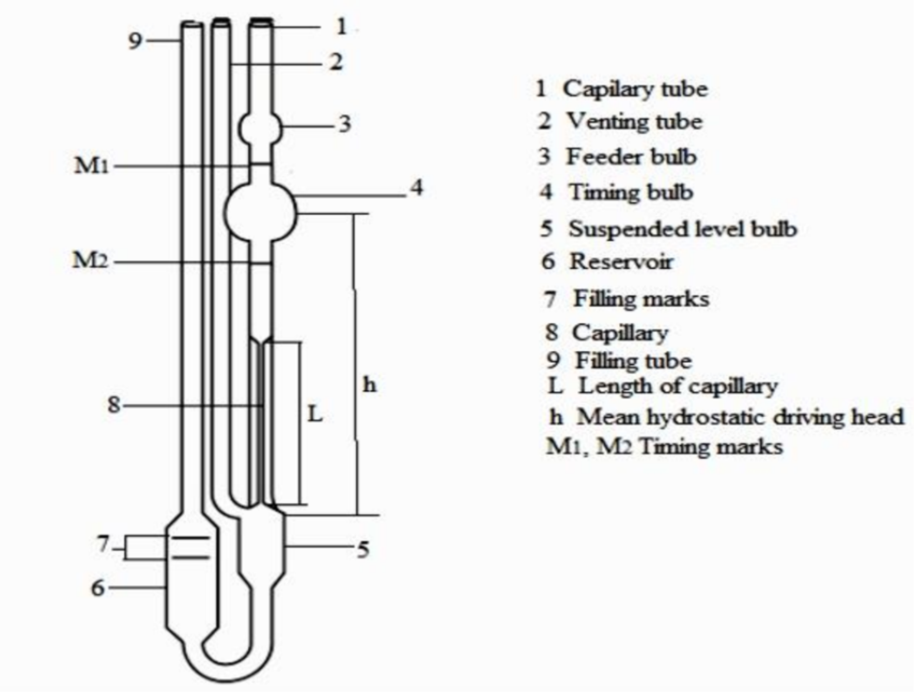


Figure 22: Schematic depiction of an Ubbelohde [51]

The composition of the specimen must be known to correctly determine the intrinsic viscosity. Preferably, the specimen does not contain any fillers at all. To ensure a good IV measurement, virgin PET pellets are used for this part of the research.

Hagenbach correction

When the solution enters the capillary, indicated by the number (8) in figure 22, the fluid accelerates due to the reduction in cross section. This leads to a pressure drop that has to be corrected for. To correct for this effect, a Hagenbach correction is applied, which is calculated at the GETEC laboratory by the Viskey software with formula 27.

$$E = \frac{1,661V^{3/2}}{L(2KR)^{1/2}} \tag{27}$$

Here $E[mm^2s]$ is the constant of Hagenbach with funnel-shaped capillary ends, $V[ml]$ is the flow-through volume, $L[mm]$ is the length of the capillary, $K[mm^2/s^2]$ is the viscometer constant and $R[mm]$ is the radius of the capillary. For the viscometry performed at GETEC, this gives $E = 50,031$ when using DCA as a solvent.

With this correction factor, the correction time can be calculated with formula 28.

$$t_H = \frac{E}{K_C T_g^2} \tag{28}$$

Here t_H is the Hagenbach correction time, t_g is tot flow-through time of the solution and K_C is a viscometer constant found on the certificate.

The corrected flow through time can then be found simply by formula 29. It should be noted that whenever the flow-through time is mentioned, this flow-through time has already been corrected with the Hagenbach correction.[33]

$$t_{Corrected} = t_g - t_H \tag{29}$$

3.3.2 Determining moisture content using a Radwag Moisture Analyser

To obtain insight in the moisture content after each experiment, a small tabletop moisture analyzer, the Radwag MA 50/1.X2.IC.A Moisture Analyzer, was available. It fulfills the definition of a thermogravimetric analyser (TGA), measuring the change in weight as the specimen is heated. From the weight reduction the moisture content can be derived.

In using this moisture analyser, the guide was carefully followed. Due to a lack of a specific drying program for PET, the drying program for HDPE was used, because it was found that these plastics required similar settings.[62]

3.3.3 Determining moisture content using a vaporimeter

To determine the moisture content of the PET more accurately and to verify the measurements done with the Radwag moisture analyser, the moisture content of a few samples are measured with a vaporimeter at GETEC. To measure the moisture content, the sample was put inside a tube. A vacuum was created inside the tube and heating elements were placed around the tube. When the tube and the containing specimen was heated at 190 °C, as is the standard for PET. The moisture inside the PET is evaporated and disturbs the vacuum. This disturbance of the vacuum can be measured accurately, which can be used to determine the moisture content in the specimen. Here it should be noted that other substances than gas could be evaporated as well. Using virgin PET minimizes the risk of the presence of contaminants.

3.4 Safety

Several elements of the research required special attention to safety. This mainly concerned building the experimental setup, as described in section 3.1 and the experiments to determine the IV, described in the next section. To inventorize and evaluate the risks, a Risk Inventarization and Evaluation (RI&E) is made with the assistance of Solutivity Advisering, a health and safety advising company. To evaluate the risk, the factor exposure is eliminated from this RI&E in consultation with the safety expert. The control measures taken led to a different approach to some parts of the thesis, most notably the application of a different desiccant and the employment of external experts as described in the next section.

3.5 Analysis at GETEC

As part of obtaining results, experiments to determine the IV and to determine the moisture in the specimen were conducted at an external lab. The decision to measure the IV in an external lab was made to mitigate risk number 24 in the risk inventarisation and evaluation added in appendix I. Measuring the moisture at a third party could give some more insight in the reliability of the measurements done with the Radwag Moisture Analyser.

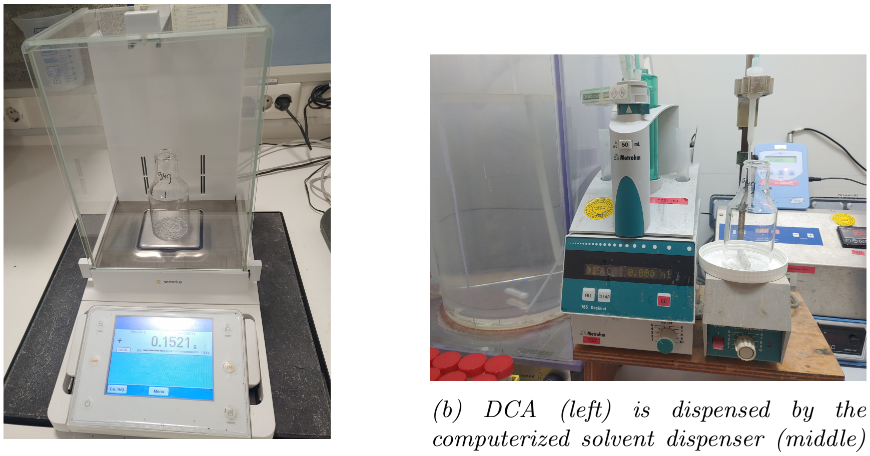
3.5.1 IV measurements

Grinding
The specimen was prepared for dissolution by grinding 30 g in a uniaxial grinder with sieve size 1.5 mm X 1.5 mm, shown in figure 23. One droplet of the antistatic agent Nopcostat HS was added to prevent the specimen from becoming statically charged. The grinding was performed for a maximum of 90 seconds, in accordance with sections 1 and 5 of ISO 1628. If the grinding time would be exceeded, the temperature of the specimen could increase due to friction in the grinder. The elevated temperature could induce post condensation, which alters the IV of the specimen, making the measurements unreliable.

Figure 23: Image of the uniaxial grinder used

Dissolution procedure
On a scale with an accuracy of 10^{-4} g between 1 and 2.5 grams of PET granulate is weighed in a glass flask. The standardized solution, maintaining a 1:200 PET to DCA ratio, was prepared. This process involved a computerized solvent dispenser accurately calculating and dispensing the necessary solvent amount into the flask to achieve the desired mixture. A magnetic stirrer was added and the flask was then placed on a heater. The temperature maintained at a constant 90 degrees Celsius for a duration of 15 minutes, during which the

solution was continuously stirred. Occasionally, some granulate adheres to the walls of the flask and is not dissolved properly. This solution is not applicable for viscometry, since the undissolved granulate may clog the Ubbelohde. Moreover, the solution did not dissolve all the PET, so the solution does not have the required ratio of 1:200, making any measurements unreliable. After complete dissolution, the solution is cooled down to approximately 25 degrees for use in the viscometer. These steps are shown in figure 27, where 24a shows weighing the sample, 24b shows the dispensing of the DCA, 24c shows the mixture in the bath of 90 °C and 24d shows some undissolved PET in the flask.



(a) Weighing the specimen

(b) DCA (left) is dispensed by the computerized solvent dispenser (middle) into the flask with specimen (right)

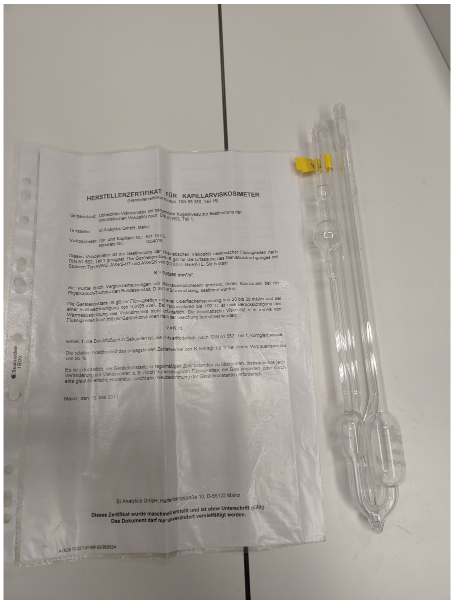
(c) Mixture continuously stirred and maintained at a constant temperature of 90 °C

(d) Undissolved PET stuck to the side of the flask, visible in the middle of the upper part of the meniscus

Figure 24: Dissolving PET in DCA

Viscometry

The Ubbelohdes used are placed in a bath which remains constant at 25 degrees (+/-0.1 degrees). In this bath, four Ubbelohdes intended for DCA are placed. These Ubbelohdes are calibrated in accordance with ISO 1628 for application of DCA. A daily first-line inspection, supplemented with a periodic external calibration, ensures that the Ubbelohdes produce reliable results. The viscometer performs the measurements as described in paragraph 3.3.1. The measurements are repeated at least three times, until three measurements are done that fall within a predetermined precision.



HERSTELLERZERTIFIKAT FÜR KAPILLARVISKOSIMETER
(Herstellerzertifikat M nach DIN 55 350, Teil 18)

Gegenstand: Ubbelohde-Viskosimeter mit hängendem Kugelniveau zur Bestimmung der kinematischen Viskosität nach DIN 51 562, Teil 1.

Hersteller: SI Analytics GmbH, Mainz

Viskosimeter: Typ- und Kapillare-Nr.: 541 13 / Ic
Apparate-Nr.: 1054016

Dieses Viskosimeter ist zur Bestimmung der kinematischen Viskosität newtonscher Flüssigkeiten nach DIN 51 562, Teil 1 geeignet. Die Gerätekonstante K gilt für die Erfassung des Meniskusdurchganges mit Stativen Typ AVS/S, AVS/S-HT und AVS/SK von SCHOTT-GERÄTE. Sie beträgt

$K = 0{,}02896\ mm^2/s^2$.

Sie wurde durch Vergleichsmessungen mit Normalviskosimetern ermittelt, deren Konstanten bei der Physikalisch-Technischen Bundesanstalt, D-38116 Braunschweig, bestimmt wurden.

Die Gerätekonstante K gilt für Flüssigkeiten mit einer Oberflächenspannung von 20 bis 30 mN/m und bei einer Fallbeschleunigung von 9,8105 m/s^2. Bei Temperaturen bis 100 °C ist eine Berücksichtigung der Wärmeausdehnung des Viskosimeters nicht erforderlich. Die kinematische Viskosität ν in mm^2/s von Flüssigkeiten kann mit der Gerätekonstanten nach der Gleichung berechnet werden:

$$\nu = K \cdot t,$$

wobei **t** die Durchflußzeit in Sekunden ist, die -falls erforderlich- nach DIN 51 562, Teil 1, korrigiert wurde.

Die relative Unsicherheit des angegebenen Zahlenwertes von K beträgt 1,2 % bei einem Vertrauensniveau von 95 %.

Es ist erforderlich, die Gerätekonstante in regelmäßigen Zeitabständen zu überprüfen. Insbesondere jede Veränderung am Viskosimeter, z. B. durch Verwendung von Flüssigkeiten, die Glas angreifen, oder durch eine glasbläserische Reparatur, macht eine Neubestimmung der Gerätekonstanten erforderlich.

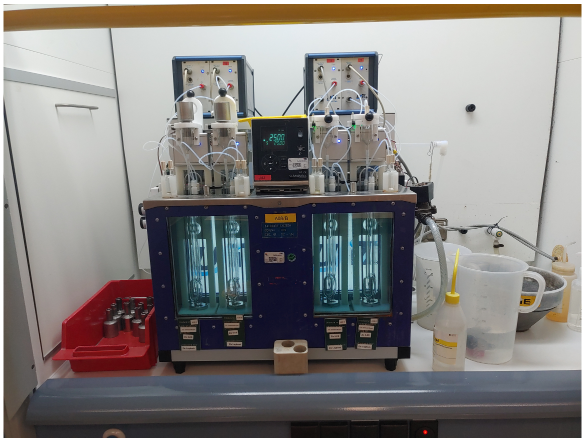
Mainz, den 12. Mai 2011

SI Analytics GmbH, Hattenbergstraße 10, D-55122 Mainz

Dieses Zertifikat wurde maschinell erstellt und ist ohne Unterschrift gültig.
Das Dokument darf nur unverändert vervielfältigt werden.

AGQSF0027-814M-00/950524

Figure 25: Ubbelohde used in viscometry, shown with the accompanying certificate



(a) Setup for four simultaneous IV measurements with four Ubbelohdes in a bath at a constant temperature of 25 °C

(b) View through the glass panel, showing two Ubbelohdes in more detail

Figure 26: The setup with the Ubbelohdes used to measure the IV

3.5.2 Moisture analysis

The moisture in the specimen was measured using a vaporimeter as described in paragraph 3.3.3 and in accordance with ISO 15512:2019-05. The measurement can be performed at 130 °C, 160 °C or 190 °C. It was conducted at 190 °C, as is prescribed for PET. In this experiment, the assumption is made that the only gas evaporating from the PET specimen is water vapour. The vaporimeter is shown from the front in figure 27a and a view through the glass side panel is given in figure 27b.

(a) *Three vaporimeters shown from the front. The left and right vaporimeter have the heating element over the specimen, whereas the middle vaporimeter has the heating element positioned down*

(b) *View through the glass side panel of the vaporimeter, showing the tube connected to the sample tube on the left. The tubes on the top are connected to measuring devices, measuring the disturbance in the vacuum in the glass ball*

Figure 27: The vaporimeter used to determine the moisture content

3.6 Relevant parameters

In this paragraph, the relevant parameters are summarized. These parameters are used throughout this paper and this paragraph will be referred to extensively.

3.6.1 Universal parameters

Universal parameters

Parameter	Symbol	Value	Dimensions	Notes
Ambient temperature	T_s	20	$[°C]$	
Temperature of specimen at t=0	T_0	20	$[°C]$	
Kinematic viscosity of the ambient air	v	$1,506e^{-5}$	$[m^2/s]$	From Engineering toolbox [5]
Thermal diffusivity of the ambient air	α	$21,7$	$[m^2/s]$	From Engineering toolbox [7]
Density of the ambient air	ρ	$1,204$	$[kg/m^3]$	From Engineering toolbox [4]
Density of PET	ρ	1360	$[kg/m^3]$	From From MatWeb [13]
Thermal conductivity of the ambient air	k	$25,87e^{-3}$	$[W/(mK)]$	From Engineering toolbox [6]
Thermal conductivity of the PET	k	0,190 - 0,290	$[W/(mK)]$	From MatWeb [13]
Radius of pellets PET	l	$0,004$	m	See figure 28

Figure 28: Size measurement of 20 pellets show an average radius of 4mm

3.6.2 Experiments in the oven

Parameters for experiments in the oven

Parameter	Symbol	Value	Dimensions	Notes
Temperature in oven	T_∞	80, 100, 120, 150	$[{}^\circ C]$	Different temperatures for different experiments
Kinematic viscosity of the air at T_∞	v	$2,088e^{-5}$, $2,297e^{-5}$, $2,513e^{-5}$, $2,967e^{-5}$	$[m^2/s]$	From Engineering toolbox [5]
Thermal diffusivity of the air at T_∞	α	$30,4$, $33,5$, $36,7$, $41,7$	$[m^2/s]$	From Engineering toolbox [7]
Density of the air at T_∞	ρ	1, $0,947$, $0,898$, $0,834$	$[kg/m^3]$	From Engineering toolbox [4]
Thermal conductivity of the air at T_∞	k	$30,32e-3$, $31,62e^{-3}$, $32,99e^{-3}$, $35,00e^{-3}$,	$[W/(mK)]$	From Engineering toolbox [6]
Airspeed	u	0,05	m/s	See paragraph 3.6.4
Characteristic length	L_c	0,03	m	$= A/p$, with A the area and p the perimeter of the surface. The length and width are 0,1 and 0,15 m [28]

3.6.3 Experiments in the microwave

Experiments in the microwave

Parameters for experiments in the microwave

Parameter	Symbol	Value	Dimensions	Notes
Temperature in microwave at t_{max}	T_∞	78	$[°C]$	see paragraph 4.1
Kinematic viscosity of the air at t_{max}	υ_{tmax}	$2,068e^{-5}$	$[m^2/s]$	From Engineering toolbox [5]
Thermal diffusivity of the air at T_{max}	α_{tmax}	$30,1$	$[m^2/s]$	From Engineering toolbox [7]
Density of the air at t_{max}	ρ	$1,006$	$[kg/m^3]$	From Engineering toolbox [4]
Thermal conductivity of the air at t_{max}	k	$30,09e{-}3$	$[W/(mK)]$	From Engineering toolbox [6]
Airspeed	u	0,05	m/s	See paragraph 3.6.4
Characteristic length	L_c	0,04	m	$= A/p$, with A the area and p the perimeter of the surface. The diameter is 0,16 m [28]

3.6.4 Airspeed in microwave and oven

To estimate the airspeed in the microwave and in the oven, the airflow of a
commonly used fan for household electronic devices (found to be 127 $\mathrm{m}^3/h)isdividedbytheverticalback-$
$to-frontcross-sectionalareainsidethemicrowaveoven(height38cmanddepth50cm)$[2][11]$.Thisyieldsanavera$
$loopconfigurationwithobstructionslikebendsandthedesiccantmaterial,thefancannotcreatethespecifiedairsp$
$0,05$ m/s. The airflow in the oven is less obstructed, but the oven has a larger cross sectional area. These effects are assumed to cancel out, the airflow in the oven is therefore estimated to have the same magnitude as in the experimental setup.

4 Results

4.1 Temperature in microwave

During the drying process, the temperature inside the microwave was measured at regular time intervals of 5 minutes. To measure the temperature, the microwave had to be opened for the IR-thermometer to measure the temperature. Opening the microwave this frequently would probably impact the drying process, so the PET pellets of these experiments were not used to determine material properties. The measurements of the temperature in the microwave are plotted and a curve was fitted to show the temperature progression, as shown in figure 29. Here the temperature reaches 80 °C. If the door of the microwave was not opened frequently and if the microwave power output was not interrupted, the temperature of the flakes would probably increase faster than shown in figure 29. Therefore, a temperature of 78 °is used as a representative estimation for the temperature during the whole experiment, as seen in paragraph 3.6.3.

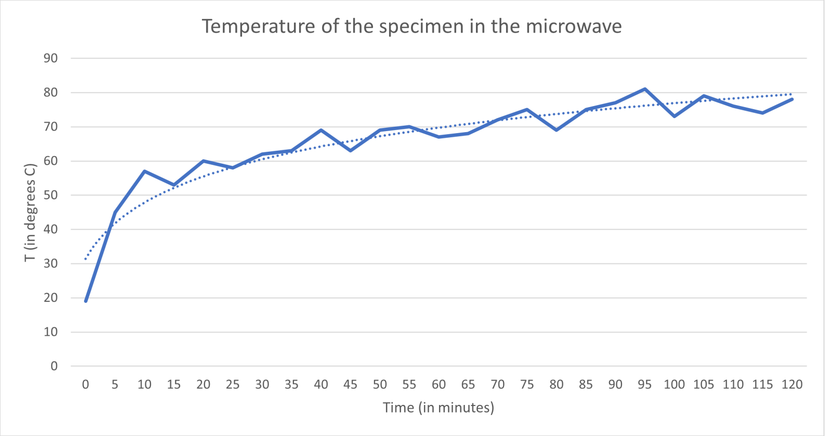


Figure 29: Temperature of the specimen inside the microwave

4.2 Moisture content

To determine the moisture content of the specimen, two different methods are used, which are described in paragraphs 3.3.2 and 3.3.3. The results of these methods are given here.

4.2.1 Radwag Moisture Analyser

The Radwag Moisture Analyser was used according to the procedure prescribed in paragraph 3.3.2 to determine the drying process in the oven and in the experimental setup. Here the results are first given for the oven, then for the microwave and finally the combined results are shown.

Moisture content at t_0
The moisture content before the pellets were dried was measured before each experiment. In total 6 experiments were conducted, measuring an average moisture content before the experiment of 0,906 weight % with a standard deviation of 0,037 weight %. For better visualization of the drying process, this first measurement is omitted in the figures.

Oven
Virgin PET pellets were placed in the oven for 0, 5, 10, 20, 30, 45, 60, 90 and 120 minutes, at 80, 100 and 120 °C. Each measurement is conducted twice and the average of these two measurements is plotted. In figure 30 the results are plotted to show the different drying characteristics. In figure 31 the results are plotted on a logarithmic scale. In paragraph 2.5.1 it is shown that the diffusion of water over time obeys an exponential relationship, therefore the three datasets are fitted along an exponential curve.

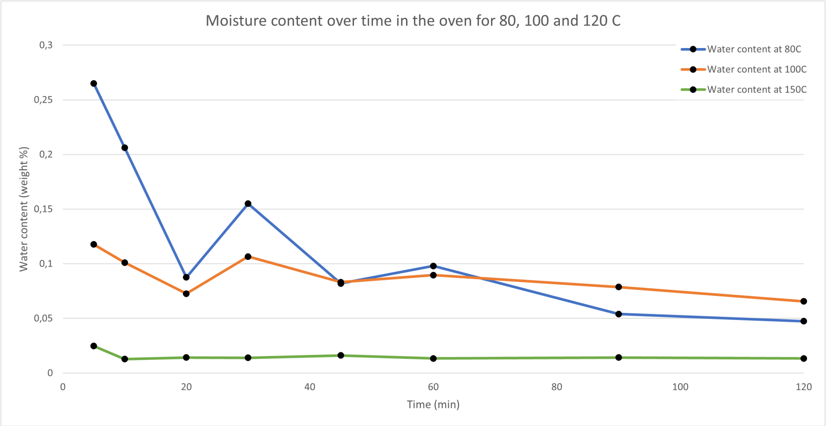


Figure 30: Moisture content of the sample in the oven at 80, 100 and 150 °C, at 8 time intervals

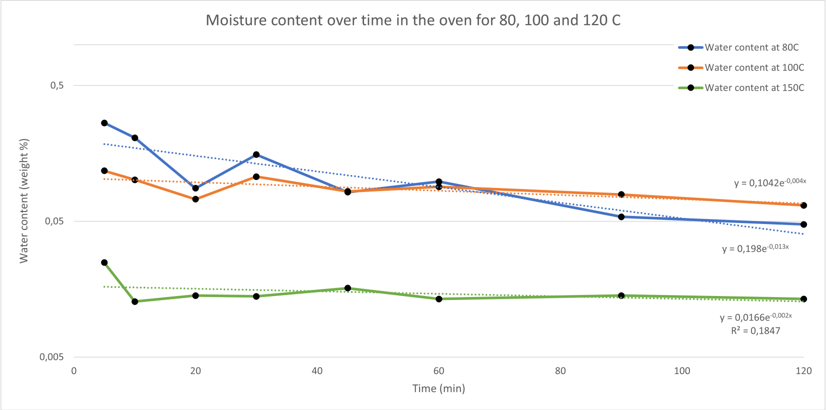


Figure 31: Moisture content of the sample in the oven at 80, 100 and 150 °C, displayed in a logarithmic scale. Each of the three datasets is fitted with an exponential curve

The Radwag Moisture Analyser gives some spread at each measurement. The average standard deviation of the measurements is ≈ 0,06 weight %. This means that the measurements for a low moisture content have a low relative precision.

After the specimen has resided in the oven at 150 °C for an extended amount of time, the Radwag Moisture Analyser still measures a relatively constant moisture content averaging of approximately 0,015 weight %. The Radwag Moisture Analyser has operating temperature of 130 °C and heats the specimen for a few minutes. For these experiments, the moisture analyser probably measures the moisture that was absorbed by the PET in the time between the experiment and the measurement. It can therefore be concluded that the PET absorbs anywhere between 0,01 and 0,02 weight % moisture in between the experiment and the measurement.

Experimental setup

The virgin PET pellets were microwaved in batches of 50 g at 100 W. At the same time intervals as the experiments in the oven, the moisture content in the pellets was measured. This experiment was repeated three times, shown in figures 32 and 33. Interestingly, there seems to be a spread in the measured moisture content. This could be due to irregularities in the drying process or due to inaccuracies from the moisture analyser.

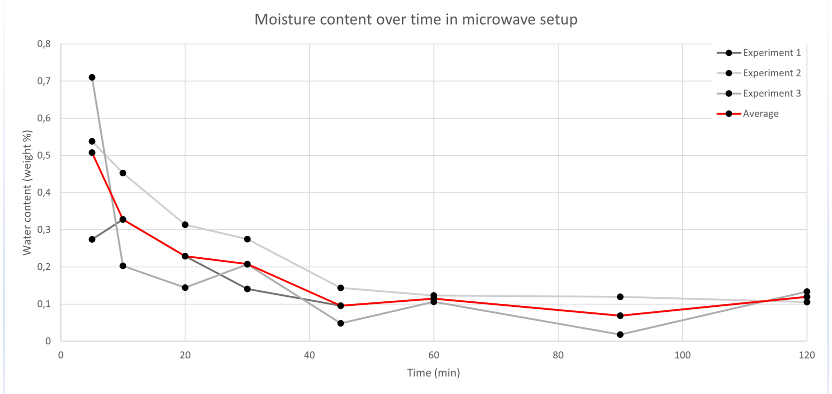


Figure 32: Moisture content of the sample in the microwave setup, at 8 time intervals

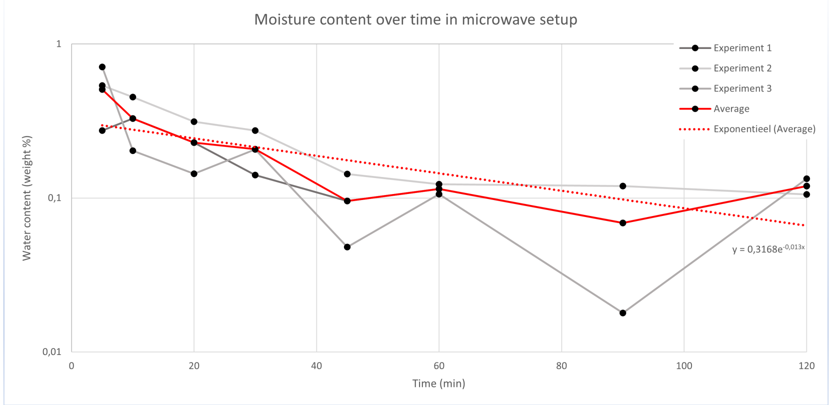


Figure 33: Moisture content in the microwave setup, displayed in a logarithmic scale. The dataset is fit with an exponential curve

Combined

To compare the traditional drying method, in the oven, to the proposed drying method in the microwave, the drying processes are plotted in the same graphs, in figures 34 and 35. In these graphs it can be clearly seen that the microwave setup dries the sample more slowly than the oven. Moreover, the moisture content after the drying procedure is much higher than the moisture content for experiments in the oven.

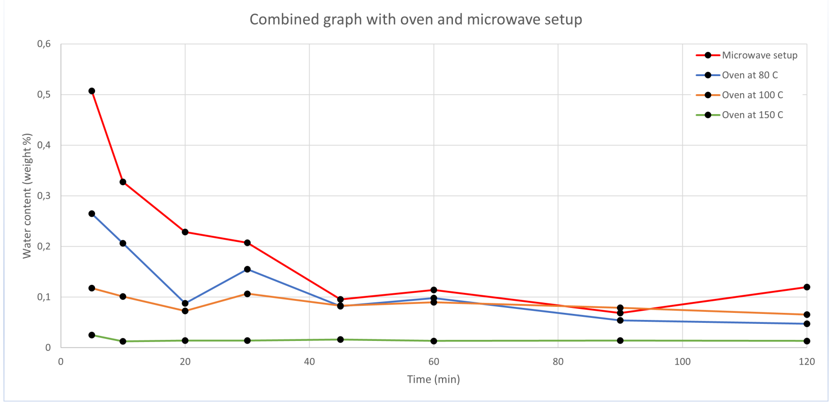


Figure 34: Moisture content of the sample in both the oven and the microwave setup, at 8 time intervals

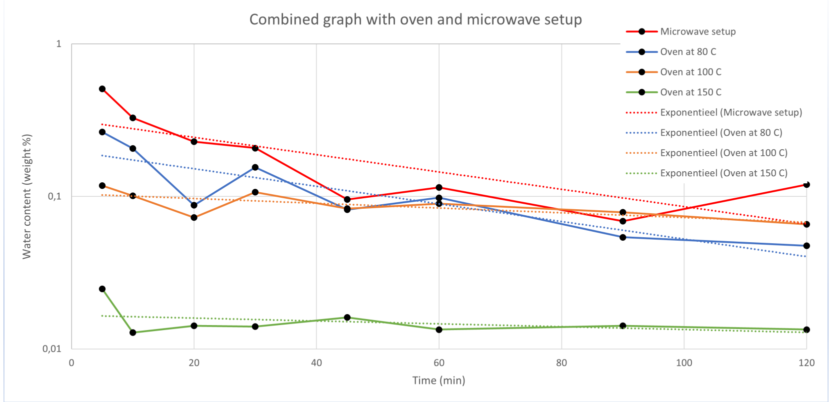


Figure 35: Moisture content in both the oven and the microwave setup, displayed in a logarithmic scale. The trendline for all the datasets are shown as well

4.2.2 Vaporimeter

According to the procedure described in 3.3.3, three moisture measurements were conducted. Two measurements were of specimens that have dried for 120 minutes either in the microwave setup or in the oven at 150 °C. The third measurement was of untreated virgin PET pellets. The results are given in table 6.

Sample	Measured water content (in weight %)
Control virgin PET	0,210
Dried in oven at 150 °C	0,013
Dried in microwave setup	0,126

Table 6: Table of moisture content measurements using the vaporimeter

In this table, the measured moisture content of the virgin PET is 0,210 weight %. In the moisture content measurements conducted by the Radwag Moisture Analyser, the moisture content was consistently measured at around 0,906 weight %. In prior research, the absorbed water by PET in equilibrium with similar surroundings was found to be 2200 ppm [55].
The moisture content of the PET dried in the oven at 150 °C is similar to the moisture content found by the Radwag Moisture Analyser. This moisture content is therefore likely the moisture absorbed from the air in between the experiment and the measurement.

4.3 IV measurements

The IV measurements were conducted according to the procedure described in 3.5.1. Three IV measurements were done for each of the three specimens. From these measurements, the average and standard deviation are calculated. The results are shown in table 7.

Sample	$1^{st}(dl/g)$	$2^{nd}(dl/g)$	$3^{rd}(dl/g)$	Avg (dl/g)	Std. (dl/g)
Control virgin PET	0,810	0,798	0,794	0,801	0,0047
Sample dried in oven at 150 °C	0,794	0,796	0,790	0,793	0,0031
Sample dried in microwave setup	0,793	0,8	0,802	0,798	0,0083

Table 7: Results of IV measurement

The virgin PET measures an intrinsic viscosity of 0,801 dl/g, which is in the range of intrinsic viscosities used for bottled water and soft drinks. This is in accordance with the claim that the provided virgin PET is intended for use in the manufacturing of these bottles.
Especially the specimen dried in the oven exhibits a decrease in intrinsic viscosity. Still, the intrinsic viscosity remains well in the range of 0,78 – 0,85 dl/g, making both dried samples suitable for manufacturing water and soft drink bottles.

5 Discussion

5.1 Calculations

5.1.1 Heat balance

The calculated microwave energy absorption is much smaller than the calculated heat loss due to convection and radiation. In calculating the microwave energy absorption, the effect of reflecting microwaves against the inner wall of the microwave oven is neglected. A microwave oven is designed in a way to maximize said reflection, so probably this effect explains the large difference between the calculated energy absorption and dissipation. The heat balance for an industrial microwave dryer would be different, as will be explained in paragraph 5.2.2.

The calculated microwave absorption is for 100 MHz or for 1 GHz. Figure 10 shows a higher dielectric loss of tap water at higher frequencies, signifying that at the used microwave frequency of 2,45 GHz more microwave irradiation will be absorbed by the water.

5.1.2 Differential equation for mass transport

The boundary condition 2 in equation 24 in paragraph 2.5.1 states that the concentration at the surface of the flake is always 0 for any $t \neq 0$. This boundary condition does not describe the system well, because even though a desiccant is used to dry the air, the humidity of the air is never zero. The solution of this boundary equation merely provides inside in the development of the concentration profile.

5.2 Experimental setup

5.2.1 Use of experimental setup

In between experiments, the microwave setup is not always fully cooled down, due to limited time and a voluminous amount of experiments. The consecutive experiments probably showed a more rapid drying rate. This difference could be significant, because the temperature of the microwave could reach 80 °C after an experiment. The drying process of an oven at 80 °C can be seen in figures 30 and 31. This shows that the effect of preexisting heat in the microwave cannot be ignored. This residual heat might also explain the spread in the drying process observed in figures 32 and 33. When microwave heating would be applied to dry plastics in an industrial setup, this would most likely be done in a continuous operation, meaning that the PET would also enter a preheated environment. These higher drying rates might therefore be a better predicting of the behaviour in an industrial setup.

5.2.2 Energy losses

Even though the experimental setup has 60x more energy output per kg PET than the industrial machines, the PET does not dry sufficiently for injection moulding processes. This is most likely due to energy losses in the experimental setup that can and should be avoided in an industrial setup. Four improvements identified that help prevent losses. In an industrial setup, the thickness of the layer of water containing PET should be much larger than in the experimental setup. In paragraph 2.4.2 it was found that this thicker layer would absorb a bigger portion of the microwave irradiation, resulting in less losses from unabsorbed microwave irradiation. Another prevention of energy loss can be obtained by preheating the dry air before entering the industrial setup, decreasing or even eliminating the heat loss found in paragraph 2.4.3. Also, continuous operation of the microwave dryer prevents cooling down, eliminating this energy loss as well. Finally, a more effective thermal insulation should be employed, decreasing the heat loss to the surrounding walls found in paragraph 2.4.4. These four improvements are likely present on industrial dryers and explain why the much higher specific energy of the microwave results in poor drying results.

5.2.3 Pellets or flakes

The experiments were done with pellets, whereas in industry applications it is expected that PET flakes are dried. Since flakes have a larger surface area for the water to evaporate from and the diffusion length for water to reach the surface is much smaller, the drying rate for flakes is likely much higher than for PET pellets. On the other hand, the larger surface area improves heat transfer between the flakes and the dry air, increasing the effectiveness of preheating the air to avoid heat losses.

5.3 Measurements

5.3.1 Radwag Moisture Analyser

The moisture content of virgin PET is expected to be 0,22 %, as explained in paragraph 4.2.2. This amount of moisture was measured by the vaporimeter at GETEC, but the Radwag Moisture Analyser could not reproduce similar results. Instead, the Radwag Moisture Analyser consistently measured a moisture content of 0,906 weight % with a standard deviation of 0,037 weight %, as shown in paragraph 3.3.2. This indicates a systematic error, making the Radwag Moisture Analyser precise, but not accurate for measurement with a higher moisture content.

For measurements for lower moisture contents, the results have a much lower relative precision. This lower relative precision is due to the small amount of mass evaporated. For most measurements after 30 minutes, moisture content measured is below 0,15 weight %. In this measurement range, the standard deviation of the measurements is approximately 0,06 weight % on average. This low relative precision has the benefit of a higher absolute accuracy, because for a lower moisture content, the measurements are within the range of one standard deviation, as can be seen in figure 36.

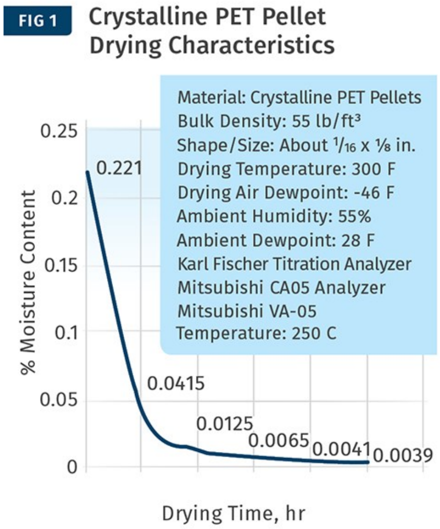


Figure 36: Process diagram of moisture content over time for drying PET at 300 F (≈ 150 °C), showing measurements that agree within one standard deviation [9]

The Radwag Moisture Analyser incidentally produced a measurement that differed orders of magnitude from other measurements of the same sample. These results were ignored and the measurements were repeated. Occasionally, the scale of the Radwag Moisture Analyser showed drift. To eliminate possible causes of this drift, the analyser was placed on a vibration insulating surface, decreasing the effect of vibrations from heavy traffic and the nearby port. Another measure was an extra hood over the whole analyser to protect it from the influence of outside airflow. The drift occurred less frequently with these measures, but it was not entirely eliminated. The cause of the drift was not found.

In between experiments, the Radwag Moisture Analyser was not always cooled. Even though this was not advise in the guide, cooling the Radwag Moisture Analyser seemed advisable to prevent natural convection inside the measurement chamber. Whenever feasible, the Radwag Moisture Analyser was cooled as much as possible in between experiments. Whether or not drift occurred did not show correlation with the temperature of the Radwag Moisture Analyser.

5.3.2 Analysis at GETEC

Usually, the specimens are delivered to GETEC sealed. Even though the tubes that carried the specimen were closed airtight and are made of material impenetrable by water, it is possible water from the atmosphere was absorbed by the specimens, making the results inaccurate. Also, in the process of taking the sample from the experiment and in the process of dosing the sample in the measurement device, the sample could have absorbed moisture from the ambient air.

The sample that was dried in the oven at 150 °C was taken one day before the tests at GETEC. The sample dried in the experimental setup was taken two days before the tests at GETEC. This would not have influenced the intrinsic viscosity, but it could have influenced the moisture content. The sample taken from the oven shows a moisture content of 0,013 weight %, which is in line with the expected moisture content found in paragraph 4.2.1. Therefore, the moisture absorbed from the ambient air through the PP container during two days is probably minimal.

5.4 Results

5.4.1 Moisture content

As expected, the higher the temperature in the oven, the sooner a low moisture content is achieved, as can be observed from figure 35. At 60 minutes and 90 minutes, this figure shows that drying at 80 °C produces a lower moisture content than drying at 100 °C. The most logical explanation for these results is the low precision at low moisture contents described in paragraph 5.3.1.

A moisture content of 50-100 ppm is allowed for injection moulding, as explained in paragraph 2.2.3. The moisture content of the virgin PET (2160 ppm) and of the microwave dried PET (1260 ppm) are far above this threshold. The oven dried specimen (130 ppm) likely absorbed some moisture from the air between the experiment and the measurement and is therefore likely suitable for injection moulding immediately after drying. As explained in section 5.2, an industrial microwave dryer is likely to achieve a significantly low moisture content after some improvements on the experimental setup are made.

5.4.2 Intrinsic viscosity

The intrinsic viscosity decreases slightly for both drying in the oven and drying in the microwave. Interestingly, the specimen dried in the oven exhibits a bigger decrease in intrinsic viscosity. One hypothesis is that this decrease is due to pyrolysis. The onset of pyrolysis for virgin PET is approximately 300 °C, but the onset of pyrolysis for contaminated PET is much lower, approaching 150 °C [30]. Oven drying contaminated PET, as is conventional in the industry, may therefore induce some pyrolysis, but this cannot explain the loss in intrinsic viscosity in the conducted experiments.

The measurements on intrinsic viscosity were conducted on a single sample for each drying method. The standard deviation might be misleading, because it merely gives the standard deviation of the various measurements conducted on the single sample. It does not give insight in the spread of the intrinsic viscosity of multiple samples. The observed decrease in intrinsic viscosity might be due to random deviations in the limited sample size.

6 Conclusion

An experimental setup using microwave irradiation and dry air to dry PET (Polyethylene terephthalate) is built. This setup is used to gain insight in the drying mechanisms and to compare this novel drying mechanism to the traditional oven drying method. The scale of this setup is much smaller than the scale of an industrial dryer. This difference in scale resulted in higher heat losses, a lower temperature and less diffusion, resulting in poorer drying capabilities than would be achieved on an industrial scale. Measurements on specimens dried with the experimental setup provided quantitative information on the PET in the drying process.

Measurements are conducted to determine the moisture content in the PET during the drying process. The drying processes of a conventional oven at 80, 100 and 150 °C and the experimental setup are compared. These measurements are conducted with a Radwag Moisture Analyser, which appeared to have an error. At high moisture contents the measurements seemed to be precise, but inaccurate and at low moisture contents the measurements seemed imprecise, but accurate. Taking this into account, a quantitative description of the drying processes in a conventional oven and in the experimental setup are given. As expected, the moisture content decreases most rapidly for higher oven temperatures. The experimental setup decreases the moisture content more slowly than the oven at 80 °C.

Measurements are also done to determine the intrinsic viscosity after the drying process. The intrinsic viscosity is determined for the virgin PET, for the PET dried at 150 °C and for the PET dried with the experimental setup. The measurements show a slight decrease in intrinsic viscosity, which is especially significant for the drying process in the oven, as shown in table 8. The standard deviation is of multiple measurements on a single sample. The measured decrease might result from a limited sample size. The intrinsic viscosity of the final product is well within the region of 0,78-0,85 dl/g usable for water and soft drink bottles. No indications of unfeasibility of microwave assisted drying are found.

Specimen	Average IV (dl/g)	Standard deviation IV (dl/g)
Virgin PET	0,801	0,0047
Oven dried PET	0,793	0,0031
Microwave dried PET	0,798	0,0083

Table 8: Results of intrinsic viscosity (IV) measurements

7 Recommendations

Further research should be conducted to investigate the efficiency and performance of an industrial setup. Here a large scale prototype could be developed, or an existing industrial microwave dryer can be used. The effect on the quality indicators, intrinsic viscosity and moisture content, should be quantified. In this setup, the airflow should go through the recyclate, instead of passing over the bulk. This setup can definitively prove the feasibility of applying microwaves in drying PET.

The process of drying the PET flakes should be modeled, to predict the drying behaviour in the setup. The optimal values for the drying parameters, like temperature, airspeed and relative humidity should be calculated. This optimization should be done for several cases, for example minimized energy consumption, maximum drying rate and minimized cost.

A potential problem with a microwave dryer is that a region with a higher water content absorbs more microwave irradiation, increasing the temperature locally to unacceptable levels. Future research should evaluate if this potential problem is a problem for the functioning of the dryer. If this problem occurs, potential solutions include devising a control mechanism for the microwave dryer, mixing the recyclate whilst drying and centrifuging the recyclate before it enters the microwave dryer.

In future research, the moisture measurements should be more accurate and more precise. An exponential curve should be fitted along the data points to accurately describe and predict the moisture content during the drying process. This additional insight in the drying process might reveal possible optimisations to the drying process, for instance increasing or decreasing the microwave output during the process.

During the recycling process, fines are produced. Some research to the origin of these fines is done and presented in appendix II. Further research should find the origin, cause and prevention method of these fines. For further recommendations on this topic, see the recommendations section of appendix II.

Despite high expectations, considerable investments, patents and products in catalogues, microwave dryers appear to have gotten out of favor in the industry. To acquire insight in the reasons for this loss of interest, market research should be performed. This market research should contain a cost analysis of oven dryers and microwave dryers. A survey taken at different relevant companies should provide insight in recent developments that might influence the development of a microwave dryer.

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Appendices

I Safety

The Risk Inventarization and Evaluation (RI&E) is made with the assistance of Solutivity Advisering, a health and safety advising company, and is presented on the last page of this appendix.

When working on my project, I noticed that the closest fire hose was often blocked by pallets of produced recycled material, see figure 37 for some examples in June and July. After notifying the head of the factory multiple times without much changes, I also took matters in my own hands as can be seen under risk numbers 10 and 16, acquiring a fire extinguisher suitable for electrical installations.

The certificate of the fire extinguisher provided expired 3 years ago, as can be seen in figure 38. After consulting Solutivity Advisering, I learned that a fire extinguisher is most likely still good for use when the pressure gauge shows that the device is still under pressure. The gauge is still in the green, so this fire extinguisher is probably good for use. Fortunately, I did not encounter any fire-related issues during this thesis.

Figure 37: Unreliable access to the fire hose

Figure 38: Certificate of fire extinguisher has expired, but the pressure gauge shows enough pressure

Risk inventarisation and evaluation

Author	Ernst Tobias Meijer
Date	26-07-2023

Building experimental setup

General info			Without measures						With measures						
Number	Activity	Risk	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment	Measure	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment
1	Disassemble microwave	Cutting hands	3	Unlikely, possible	3	Severe, trip to hospital	Improvement necessary		Wear protective gloves	0,5	Plausible, extremely unlikely	1	Meaningful, first aid	Acceptable risk	
2	Using adhesives and silicone	Skin irritation	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk		-	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	Protective gloves are available
3	Putting quicklime into stocking	Dehydration of exposed bodyparts	6	Likely	3	Severe, trip to hospital	Improvement necessary		Wear gloves and glasses	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	
4	Drilling holes	Drill slips/shoots out	3	Unlikely, possible	3	Severe, trip to hospital	Improvement necessary		Apply less pressure, fixate the object	0,5	Plausible, extremely unlikely	3	Severe, trip to hospital	Acceptable risk	
5	Cutting aluminium tube	Cutting hands	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk		Wear protective gloves	1	Possible long-term	1	Meaningful, first aid	Acceptable risk	Risk has occured
6	Cutting and shaping chicken wire	Cutting hands/arms	6	Likely	1	Meaningful, first aid	Real risk		Wear protective gloves	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	

Testing experimental setup

General info			Without measures						With measures						
Number	Activity	Risk	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment	Measure	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment
8	Turning on microwave	Short circuit	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk		-	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	
9	Running experiment	Exposure to radiation	1	Possible long-term	3	Severe, trip to hospital	Acceptable risk		-	1	Possible long-term	3	Severe, trip to hospital	Acceptable risk	
10	Running experiment	Microwave ignites	1	Possible long-term	7	Long lasting/chronic damage	Real risk		Place fire extuingisher (type B or C) nearby Do not leave running equipment unattended	0,5	Plausible, extremely unlikely	7	Long lasting/chronic damage	Acceptable risk	Fire fighting equipment at facility was not in order. I used a portable fire extuingisher
11	Running experiment	Specimen ignites	1	Possible long-term	1	Meaningful, first aid	Acceptable risk		Only water during testing of setup	0,1	Virtually impossible	1	Meaningful, first aid	Acceptable risk	
12	Running open loop experiment	Quicklime in air	1	Possible long-term	1	Meaningful, first aid	Acceptable risk		-	1	Possible long-term	1	Meaningful, first aid	Acceptable risk	
13	Replacing quicklime	Drying of exposed areas	10	Almost certain	3	Severe, trip to hospital	Improvement necessary		Wear gloves and glasses	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	
14	Handeling hot water	Burning	1	Possible long-term	1	Meaningful, first aid	Acceptable risk		Wait until it cools down	0,2	Practically impossible	1	Meaningful, first aid	Acceptable risk	

Using experimental setup

General info			Without measures						With measures						
Number	Activity	Risk	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment	Measure	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment
15	Running experiment	Exposure to radiation	1	Possible long-term	3	Severe, trip to hospital	Acceptable risk		-	1	Possible long-term	3	Severe, trip to hospital	Acceptable risk	
16	Running experiment	Microwave ignites	1	Possible long-term	7	Long lasting/chronic damage	Real risk		Place fire extuingisher (type B or C) nearby Do not leave running equipment unattended	0,5	Plausible, extremely unlikely	7	Long lasting/chronic damage	Acceptable risk	Fire fighting equipment at facility was not in order. I used a portable fire extuingisher
17	Regenerating silica gel in oven	Burning on oven	3	Unlikely, possible	3	Severe, trip to hospital	Improvement necessary		Use gloves	1	Possible long-term	1	Meaningful, first aid	Acceptable risk	
18	Preparing specimens	Cutting	1	Possible long-term	1	Meaningful, first aid	Acceptable risk		-	1	Possible long-term	1	Meaningful, first aid	Acceptable risk	
19	Preparing specimens	Poisoning (biological)	3	Unlikely, possible	3	Severe, trip to hospital	Improvement necessary		Use gloves and wash hands	0,5	Plausible, extremely unlikely	3	Severe, trip to hospital	Acceptable risk	
20	Preparing specimens	Poisoning (chemical)	0,2	Practically impossible	3	Severe, trip to hospital	Acceptable risk		-	0,2	Practically impossible	3	Severe, trip to hospital	Acceptable risk	

Data collection

General info			Without measures						With measures						
Number	Activity	Risk	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment	Measure	Chance of occurance	Meaning chance	Effect	Meaning effect	Risk score	Comment
21	Handling flakes	Stings from needle shaped flakes	6	Likely	1	Meaningful, first aid	Real risk		Wear gloves	0,5	Plausible, extremely unlikely	1	Meaningful, first aid	Acceptable risk	Risk has occured
22	Loading moisture analyser	Skin burn from hot tray	6	Likely	1	Meaningful, first aid	Real risk		Wear gloves	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	
23	Emptying moisture analyser	Skin burn from hot tray with speci	6	Likely	1	Meaningful, first aid	Real risk		Wear gloves	3	Unlikely, possible	1	Meaningful, first aid	Acceptable risk	
24	Determining IV using Ubbelohc	Injury from aggressive chemicals	3	Unlikely, possible	7	Long lasting/chronic damage	Improvement necessary		IV measurements at external lab	0,2	Practically impossible	7	Long lasting/chronic damage	Acceptable risk	
25			3	Unlikely, possible	3	Severe, trip to hospital	Improvement necessary			0,5	Plausible, extremely unlikely	3	Severe, trip to hospital	Acceptable risk	
26			0,2	Practically impossible	3	Severe, trip to hospital	Acceptable risk			0,2	Practically impossible	3	Severe, trip to hospital	Acceptable risk	

II Fines

In the recycling process fines are created. Fines are small plastic flakes that are too small for further processing. Currently there is no standard further process for these fines, so they are stored separately. These fines can be considered a loss in the recycling process. It is therefore important that the amount of fines created is as small as possible.
It is observed that more fines are created if the hot washing is performed at a higher temperature of around 80°C as compared to hot wash at a lower temperature of around 60°C. One would expect that materials become tougher at higher temperatures, which mitigates the formation of fines. Toughening at higher temperature is not the dominant mechanism, there is another mechanism at play that promotes the formation of fines.
As a part of this thesis, some research to the formation of fines is conducted, in order to understand the recycling processes better.

II.1 Analysis

A sample is taken from the collected fines and is observed in more detail. Here it is seen that the shape of the fines seems to be random, with an overrepresentation of an elongated shape. Explanations of this overrepresentation include abrasion along the edges of the flakes and errors resulting from using a sieve.
Abrasion along the edges of the flakes can occur during transportation of the flakes, either using mechanical screws or using pressurized air. This abrasion induces stress mostly at the edges of the flakes, resulting in fines with an elongated shape. It is observed that more fines are created when the transportation mechanism is pressurized air, supporting this explanation.
If elongated shapes approach the mesh end-on, they might pass, whereas if they approach the mesh side-on, they do not pass. This results in more elongated shapes passing through the mesh, falsely implying that elongated particles are overrepresented. This is a well known phenomenon, studied mostly in the field of geology [66].
Most likely, both mechanisms are at play simultaneously.

To verify that the fines separated from the recycled plastic are indeed mostly PET, a sink-float analysis is performed in a liquid medium of water with detergent, shown in figure 39. Here the vast majority of the fines are sunk to the bottom, only a small minority of the fines floats in this liquid medium. This indicates that the material is indeed mostly PET.

Figure 39: Sink-float analysis of the fines in a large container, showing most fines sink

II.2 Hypotheses

Multiple explanations are possible. Two explanations that are discovered in more detail are given here.

II.2.1 Fines released by washing of glue

This hypothesis assumes that the fines are created in the grinding and shredding process. Grinding and shredding of the plastic is done in multiple steps in several machines, so the plastics are cut many different times in many different angles. This creates a wide size distribution of the flakes, also creating very small flakes, the fines. These fines remain stuck to the flakes by glue residue. The hot wash removes more glue at higher temperature, which in turn frees more fines. The fines are thus already present in the shredded PET. The reason why most of the fines are PET and no other type of plastic, is because most glue on consumer plastic is used on PET bottles.

II.2.2 Crystallization

In the grinding and shredding process, the PET deforms plastically. This deformation induces crystallinity, which creates a harder and more brittle phase than the surrounding plastic. The hot wash at higher temperature promotes the formation of crystalline regions, increasing the crystallinity of the mechanically induced crystalline regions. During transportation of the plastic by screws and/or pressurized air, the flakes are abraded. This stress induces brittle fractures, creating the fines. The reason why this effect occurs on PET and not on other types of plastic, is because

II.3 Experiments

To test the hypotheses, multiple experiments can be done, which are itemized below.

- Roast PET flakes which are washed at different temperatures to see how much glue residue is present
- Perform near infrared spectroscopy (NIR) to identify the material of the fines.
- Perform X-ray diffraction on the fines to evaluate the crystallinity of the fines.
- Observe a flake from before the hot washing under an optical microscope, to find if any crystalline regions or fines are present at the edges.

The first two experiments are performed and described in the subsequent paragraphs.

II.3.1 Roast PET washed at different temperatures

Materials:

- Sample (see below)
- NaOH (1.5M)
- Tubiwash
- Water
- Tin foil
- Detergent

Tools:

- Glass beaker (3L)
- Water cooker capable of heating to 65°C, 70°C, 75°C, 80°C and 85°C
- Heating/mixing platform
- Magnetic stirrer
- Measuring cylinder (50mL)
- PH meter
- Scale
- Timer/stopwatch
- Oven capable of heating to 100°C and 220°C
- PPE (chemical gloves, thermal protection gloves, safety glasses)

Sample collection and preparation:

Material is gathered just before it would enter the hot-washing process in the industrial setup. The 3L glass beaker is used to perform a sink-float of the sample, in a liquid medium of water with detergent. During the sink-float all the plastic that floats (mostly comprised of PE and PP) is discarded. After a few repetitions, this leaves a sample that sinks in water (mostly comprised of PET).

Experiment:
Washing:

1. Add 1L of water, heated to 65°C in the water cooker, 30 mL of NaOH, 10 mL of tubiwash and the magnetic stirrer to the 3L glass beaker
   PPE: Use chemical gloves and safety glasses for all these steps
2. With the heating turned on and the mixing set at speed 3 on the heating/mixing platform, add 100g of wet PET to the beaker.
   Note: Add small amounts of the PET at a time to prevent the magnetic stirrer from stalling
3. Measure the temperature of the mixture and cover the beaker with tin foil to insulate the mixture. Set a timer/stopwatch for 12 minutes.
4. After 12 minutes of washing, remove the tin foil and measure the temperature again. Then rinse the PET flakes 3 to 4 times under fresh to remove any chemical residues.
5. Repeat the above steps for the water cooker set at 70°C, 75°C, 80°C and 85°C to obtain a total of 5 washed specimens.

Experiment:
Roasting:

1. Dry the 5 samples in a preheated oven at 100°C for 3 hours to rid the specimens from any residual water.
   PPE: Use thermal protection gloves for this and the next step
2. Roast the 5 samples in a preheated oven at 220°C for 3 hours
3. Evaluate the colour of the specimens, more brown means more contamination, indicating the presence of more glue.
   PPE: Use any gloves for this step

Results:
The temperatures during the experiments are given in table 9. It is easily observed that the actual temperature is much lower than the temperature set by the water cooker. The hottest temperatures of the hot wash (around 80°C) are thus not mimicked by this experiment.

Sample #	$T_{Watercooker}$	Measured T_0	Measured T_{End}	T_{Avg}
1	65	57,9	54,9	56,4
2	70	59,9	56,5	58,2
3	75	66,6	62,3	64,5
4	80	68,3	64,0	66,2
5	85	75,7	70,5	73,1

Table 9: Table of process temperatures of the different samples

The five samples after roasting have been photographed with and without flash. The pictures of these samples is shown in figure 40. Some of the flakes are extremely dark brown. These flakes are most likely made out of PET laminated with another type of plastic. This lamination contains contaminants which turn into this dark brown colour during roasting.
Even though it is not very obvious from the pictures, a slight difference in colour is observed. The samples washed at a higher temperature seem less brown than the samples washed at a lower temperature. This is shown in more detail in figure 41.

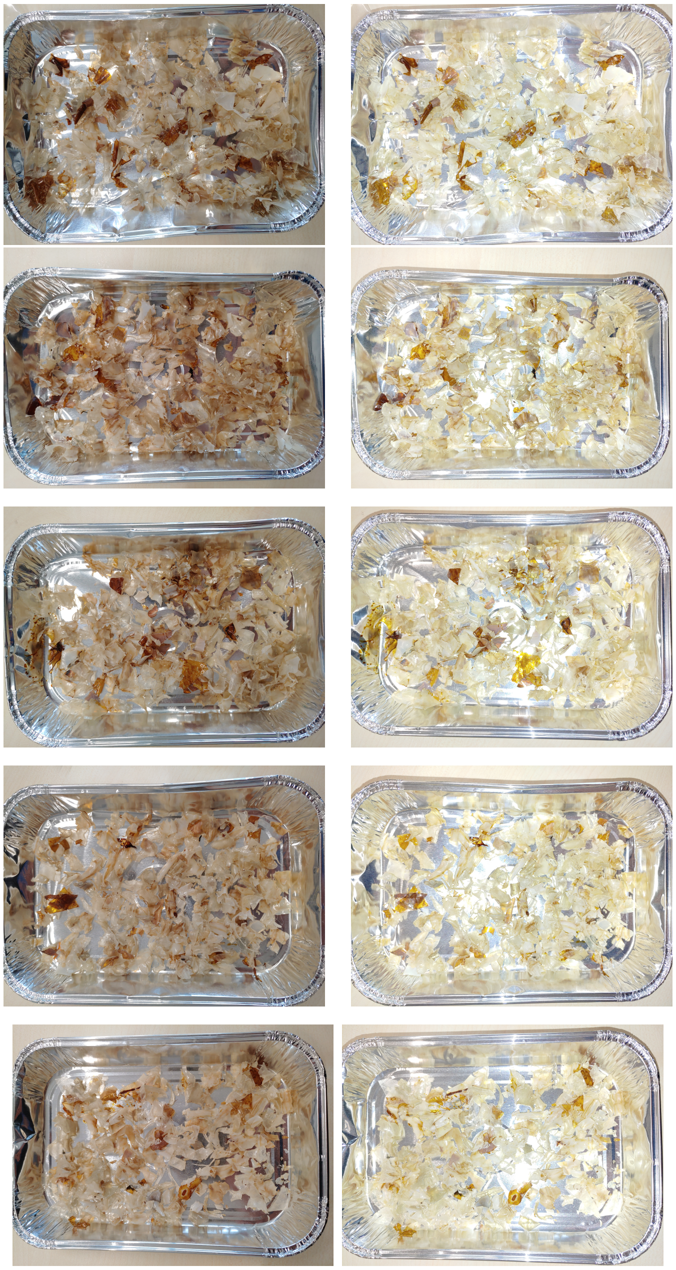


Figure 40: Pictures of the roasted flakes, pictures on the left are taken without flash and pictures on the right are taken with flash. top to bottom the temperature setting on the water cooker was 65° C, 70° C, 75° C, 80° C and 85° C

Figure 41: Combined pictures, top without flash, bottom with flash, with on the left side the roasted flakes washed with the water cooker set to 65° C and on the right side the roasted flakes with the water cooker set to 85° C

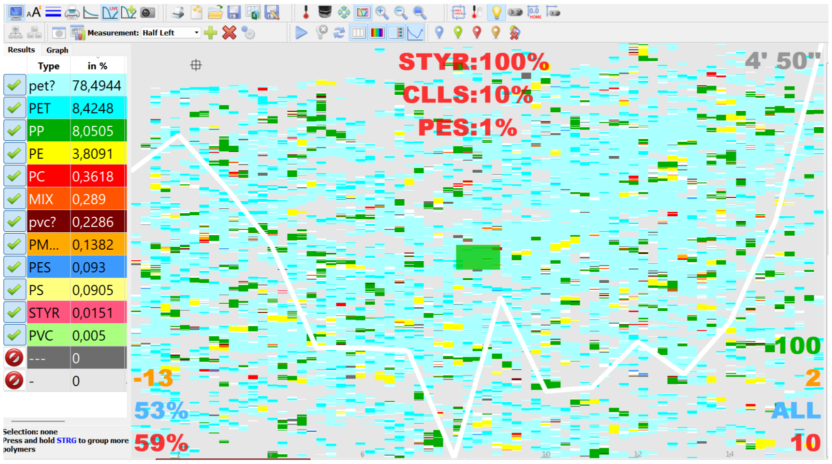
II.3.2 Near Infrared Spectroscopy (NIR) on the fines

Methods:

The fines that are produced at PRA (Plastics Recycling Amsterdam) are scanned by an NIR spectrometer. The used spectrometer is sIRoPAD manufactured by GUT [37]. The fines are spread out over the platform of the sIRoPAD as shown in figure 42.

Figure 42: sIRoPAD ready to start its measurement

Results:
The spectrometer was not calibrated to optimally detect PET. Where the measurements say *pet?* it is assumed to be PET, which has proven to be a correct assumption in prior usage of this spectrometer. As shown in figure 43, 86.9192 % of the fines are identified as PET, 8.0503 % is identified as PP and 3.8091 % is identified as PE.



Measurement: Half Left

	Type	in %
✓	pet?	78,4944
✓	PET	8,4248
✓	PP	8,0505
✓	PE	3,8091
✓	PC	0,3618
✓	MIX	0,289
✓	pvc?	0,2286
✓	PM...	0,1382
✓	PES	0,093
✓	PS	0,0905
✓	STYR	0,0151
✓	PVC	0,005
⊘	---	0
⊘	-	0

Selection: none
Press and hold STRG to group more polymers

Figure 43: Result of sIRoPAD measurement, showing a majority of PET present

II.4 Discussion

It is not very clear in the pictures of the roast PET that the samples are less brown after hot washing at a higher temperature. It is possible that the reflection of the tin foil basket or the presence of the very dark brown flakes obscures judgement. Still, when observing the flakes with the naked eye, the difference between the two samples is clear. A hotter wash produces flakes that are less brown after roasting. These results show that a hotter hot wash indeed removes more impurities, which are most likely glue. It can however not be stated that this is also the reason why more fines are present when the hot wash is set to a higher temperature.

The temperature of the wash was significantly lower than the setting on the water cooker. The water temperature in the experiments reached an maximum average temperature of 73,1 °C, which is lower than 80-85 °C which is used in the industry.

The sIRoPAD is not designed to analyse material of dimensions smaller than 1 mm. Because of the small size of the fines, the sensitivity of the spectrometer was increased, which could lead to errors in measurement. The increase sensitivity setting uses less data to identify the material, which leads to a bias towards identifying material as PP. Even with this bias towards PP, PET is the most detected material. It is safe to say that the fines are mainly comprised of PET. The sIRoPAD gives too many significant figures, the composition of the bulk is not known as precisely as implied by the results. It does however give good insight in the approximate composition.

II.5 Conclusion

The fines have been identified to be mostly PET. The material is contaminated, most likely with glue residues.

II.6 Recommendations

There are two possible experiments to evaluate the hypothesis that fines adhere to the flakes by glue. Firstly, the glued fines could be observed directly on the flakes with optical microscopy. Secondly, using a sieve, the particle size distribution of the flakes could be determined before and after the hot wash.
Even though indicators of glue residue were found, the hypothesis of crystallization should also be tested. This hypothesis can be tested by measuring the crystallinity before and after the hot wash with X-ray diffraction spectroscopy. If an increase in crystallinity is found, it is then useful to device a mechanical test that evaluates if the crystalline parts of the flakes increase the brittleness locally and may explain the formation of flakes.
These experiments are recommended as future research.

III Building, verifying and using setup

In this appendix some iterations in building, verifying and using the setup are shown. Lessons were learned from these iterations and may prove useful if similar experiments are repeated

III.1 Building the setup

III.1.1 Adhesives

The first lesson learned in building the setup is that product information can not always be trusted. “Suitable for all surfaces” did not mean suitable for my surfaces. More specifically, two types of glue and kit were not suitable to fixate the silicone putty, as shown in figure 44. Eventually silicone putty is avoided in attaching the pipes to the microwave and the dryer. The structures inside the microwave were fixated with Bison Poly Max [8]. This kit did not bond strongly with the silicone putty, but strong enough to secure the bond in the enclosed environment, as shown in figure 45.

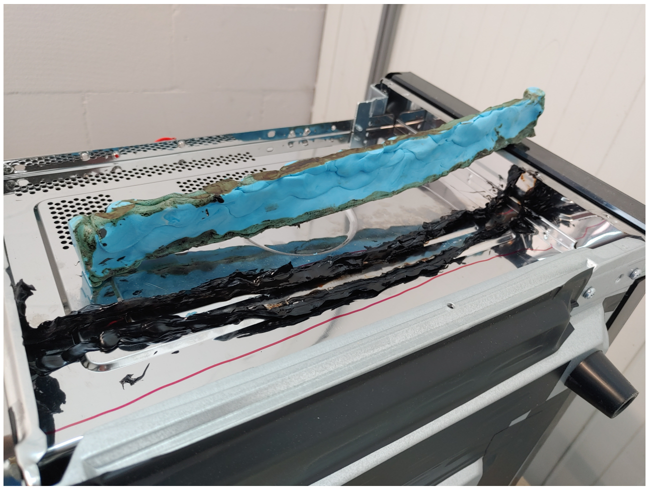


Figure 44: Loose silicone putty structure with the black kit it was supposed to adhere to

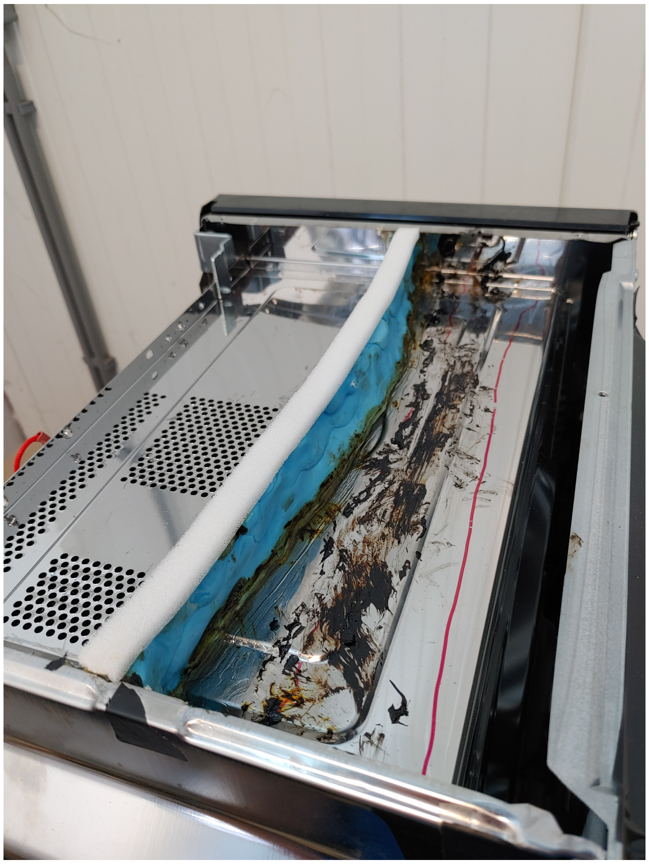


Figure 45: Silicone putty structure adhered with the Bison Poly Max glue

III.1.2 Desiccant

Instead of the ordered calcium oxide (quicklime), calcium hydroxide arrived. An alternative desiccant was found in Portland cement. Portland cement contains approximately 65% quicklime and was therefore expected to have similar desiccant properties as pure quicklime. The design was to put the quicklime in stockings inside a chicken wire cage in the desiccant dryer, see figure 46. This chicken wire cage allowed for easy replacement of the quicklime. The Portland cement consists of smaller particles than the holes in the stockings, so it leaked through. This problem would not have occurred with the larger size grains of the ordered quicklime. It was also found that the Portland cement was saturated relatively quickly, as shown in paragraph III.2.1 of this appendix. Moreover, this desiccant posed some health and safety issues as described in Appendix I.

Figure 46: Portland cement in stockings inside the chicken wire cage in the dryer

After some research, it was found that CaO does achieve an extremely low relative humidity, but does so extremely slowly compared to other desiccants. This is shown in figure 47. All these reasons combined led to the use of silica gel.

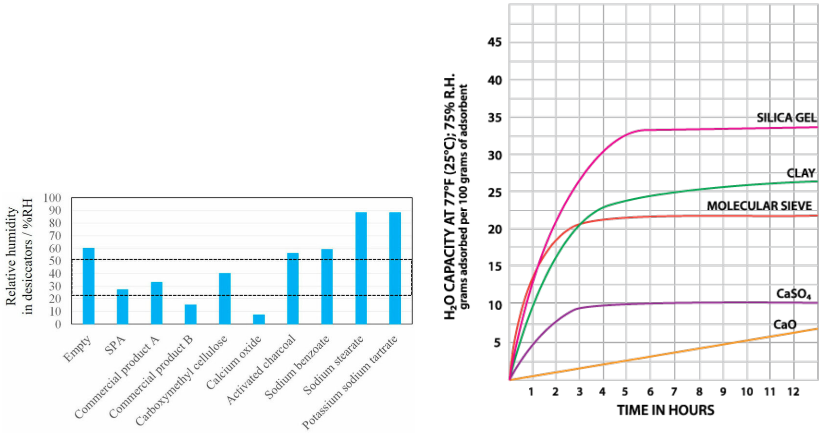


Figure 47: Figures showing the equilibrium relative humidity of CaO and the absorption rate compared to common desiccants [65] [3]

III.1.3 Silicone putty

It is recommended to avoid the use of silicone putty, because it does not adhere well to common off-the-shelf products. This problem was also encountered when the pipes were attached to the microwave and the dryer. Initially, an attachment was designed to attach the pipes to the microwave in a way that was detachable, which is shown in figure 48. This attachment used the silicone putty and could therefore not be attached securely. The pipes were attached in a way that was not detachable, as shown in figure 49. Here several types of tape are used to ensure both an airtight and heat resistant seal.

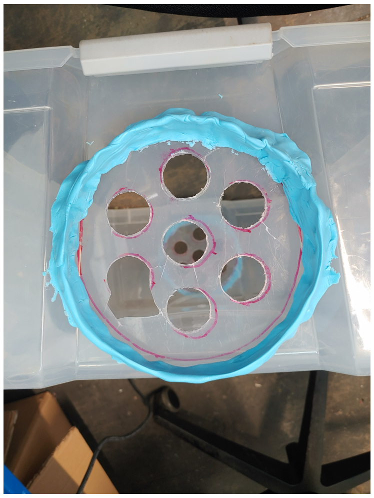


Figure 48: Silicone putty attachment

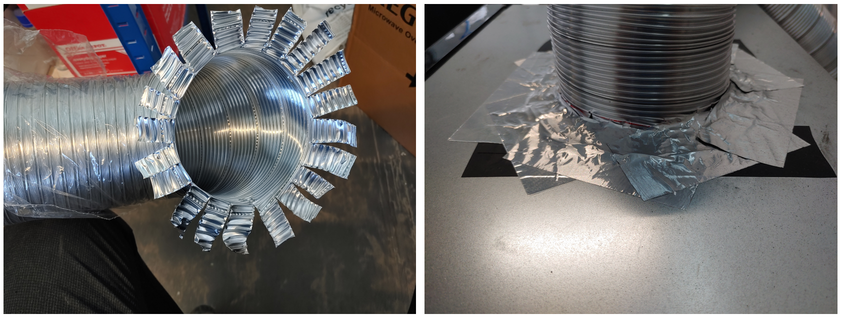


Figure 49: Pipes not detachable fixed to the microwave

III.2 Testing the setup

To verify that the setup works as intended, several tests were conducted. The first test was in an early stage of developing the setup, to heat a bowl of water in the microwave without the pipes connected, see figure 50. This test was to see if the microwave would not overheat. The thermo-hygrometer was put on the air outlet of the microwave, and measured an increase in temperature and humidity. The graph of one of these first experiments, heating a bowl of water for 30 minutes at $700W$ is shown in figure 51. The spikes in the relative humidity show the pulse width modulation method used to achieve an average power of $700W$. Interestingly, the relative humidity reduces as the temperature increases, but the dew point temperature remains approximately constant. This graph shows that the microwave setup and the thermo-hygrometer work well. During these tests it was observed that the microwave itself became hot when high microwave powers were tested. As explained in paragraph 3.1.3 of the main text, the microwave experiments are conducted at $100W$. At this operating power the microwave setup did not heat up significantly, reducing the risk of the microwave setup catching fire. Moreover, it was observed that in an experiment at $900W$, the microwave automatically turned off when it reached an elevated temperature.

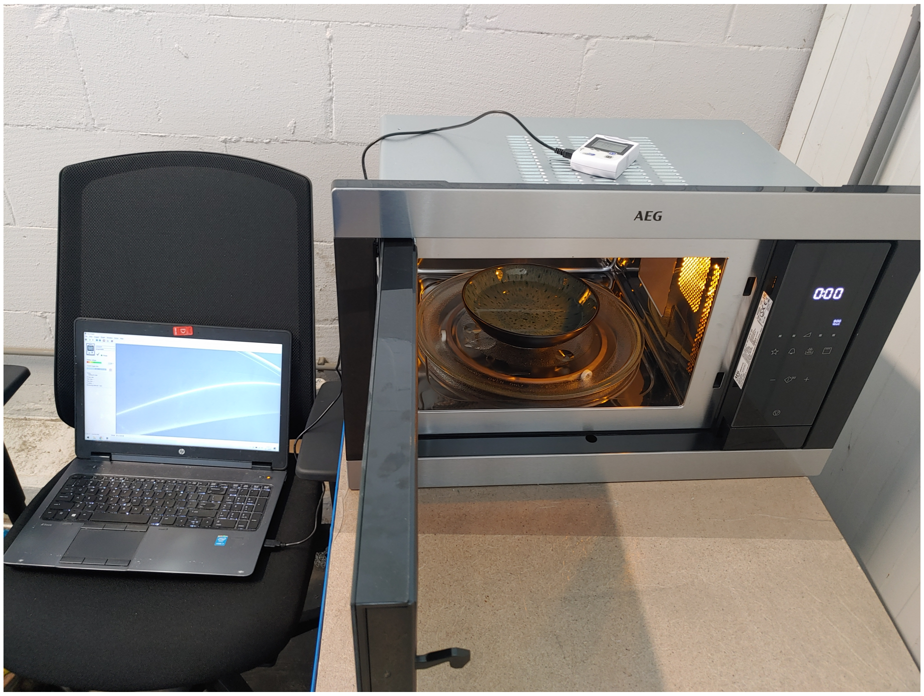


Figure 50: Setup for the test without the pipes connected

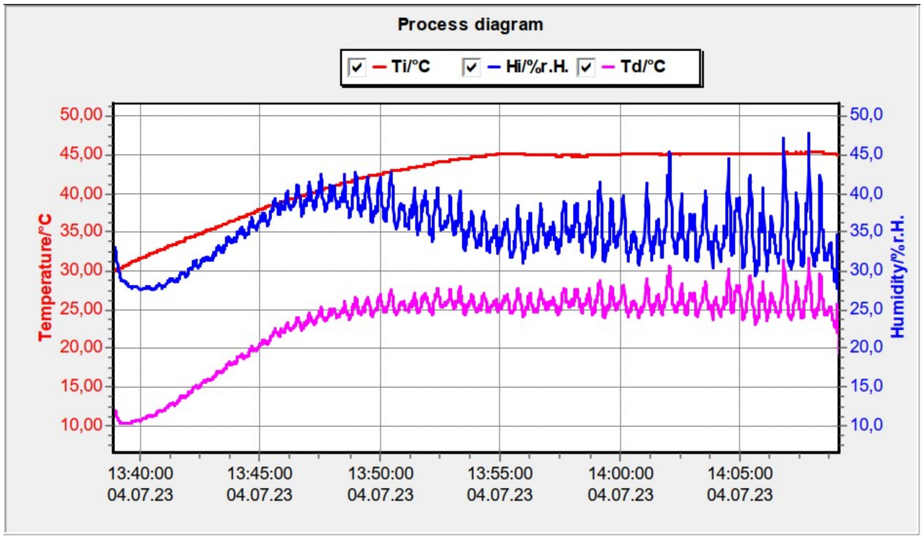


Figure 51: Test with no pipes attached. Bowl of water for 30 minutes at 700 W

III.2.1 Desiccant material and hygrometer

The air in the dryer was only dry for a few tests. Afterwards the dryer seemed to have lost its drying capabilities. Replacing the desiccant, which was Portland cement at this stage of building the setup, did not seem to change the drying capabilities of the dryer. This led to the hypothesis that the cement lost its drying capabilities. To verify this hypothesis, the hygrometer was put inside the bag of cement overnight. The process diagram is shown in figure 52 and analysis and follow-up of this experiment are shown in figure 53.

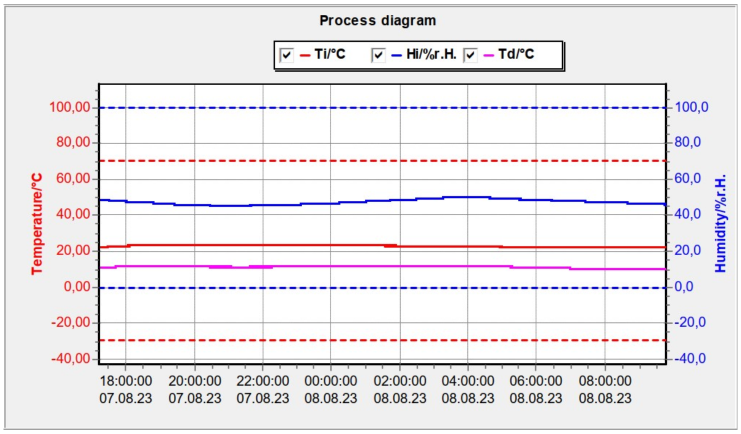
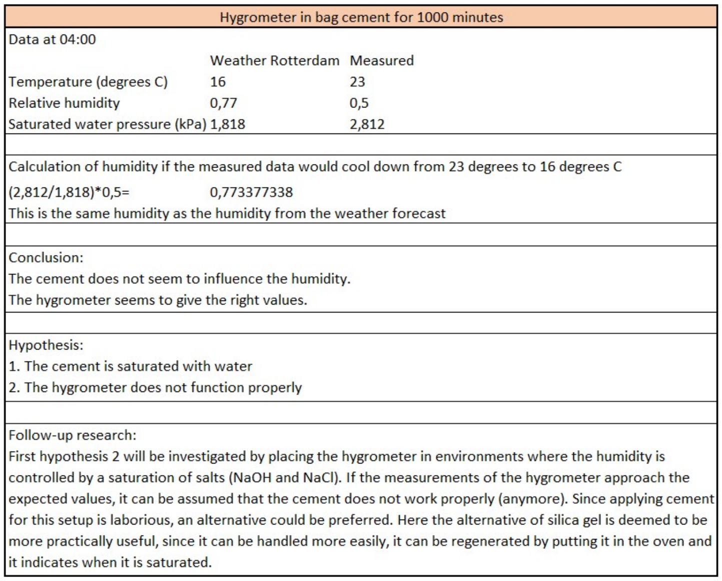


Figure 52: Process diagram of the experiment to put the hygrometer in the bag of cement overnight



Hygrometer in bag cement for 1000 minutes

Data at 04:00

	Weather Rotterdam	Measured
Temperature (degrees C)	16	23
Relative humidity	0,77	0,5
Saturated water pressure (kPa)	1,818	2,812

Calculation of humidity if the measured data would cool down from 23 degrees to 16 degrees C

(2,812/1,818)*0,5= 0,773377338

This is the same humidity as the humidity from the weather forecast

Conclusion:

The cement does not seem to influence the humidity.

The hygrometer seems to give the right values.

Hypothesis:

1. The cement is saturated with water
2. The hygrometer does not function properly

Follow-up research:

First hypothesis 2 will be investigated by placing the hygrometer in environments where the humidity is controlled by a saturation of salts (NaOH and NaCl). If the measurements of the hygrometer approach the expected values, it can be assumed that the cement does not work properly (anymore). Since applying cement for this setup is laborious, an alternative could be preferred. Here the alternative of silica gel is deemed to be more practically useful, since it can be handled more easily, it can be regenerated by putting it in the oven and it indicates when it is saturated.

Figure 53: Results and analysis of the experiment to put the hygrometer in the bag of cement overnight

As can be seen in figure 53, one hypothesis is that the hygrometer does not function properly. To evaluate this hypothesis, the hygrometer was put in bowls with chemical that control the relative humidity. Of one experiment, with NaOH, this experiment is shown in figure 54. The resulting process diagram is shown in figure 55, which shows that the relative humidity approaches 9% at a temperature of $20°C$, as was expected from literature [40]. This experiment was repeated with NaCl, saturated with water, which should give a relative humidity of 75 %. The process diagram is shown in figure 56 and shows that the relative humidity steadily increases. This experiment was cut short, because these results proved that the hygrometer worked as intended. This led to the conclusion that the bag of cement was fully saturated with water and lost its hygroscopic properties. This in turn supported the decision to use silica gel as a desiccant material.

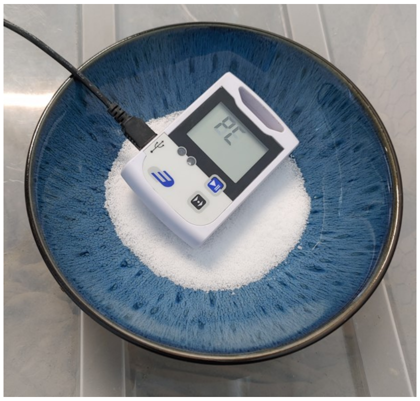


Figure 54: Bowl filled with NaOH. During the experiment, this bowl was covered with tin foil

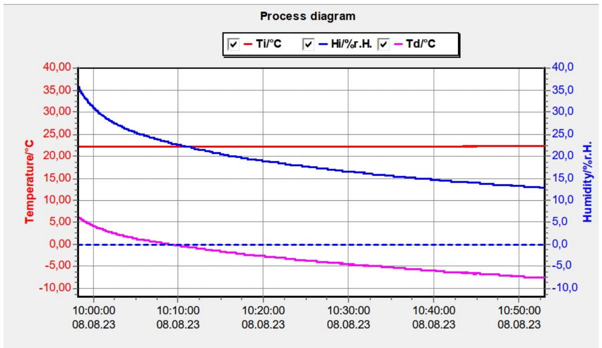


Figure 55: Process diagram of the hygrometer in the bowl of NaOH, showing it approaches the expected relative humidity of 9%

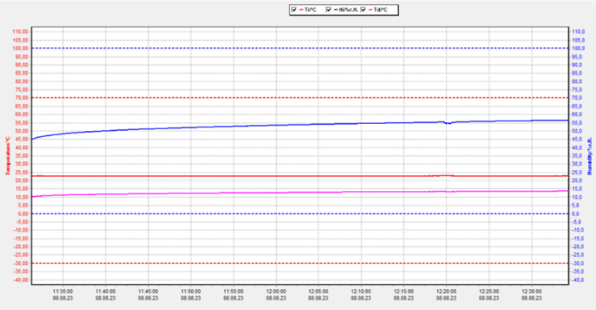


Figure 56: Process diagram of the hygrometer in the bowl of NaCl, showing it approaches the expected relative humidity of 75%

III.2.2 Open or closed system

The microwave setup could work in a closed loop configuration, meaning that the air outlet of the microwave is the air inlet of the dryer. The closed loop configuration is only more effective if the humidity of the microwave outlet is lower than the ambient humidity. To verify if this is the case, a bowl with $0,1L$ water was placed in the microwave setup in closed loop configuration and was microwaved for $100min$ at $100W$. The measured humidity at the microwave air outlet is shown in the process diagram in figure 57. It can be seen that the relative humidity at the air inlet is approximately 38 %. At the time of this experiment, the ambient humidity was approximately 44 %. Therefore, the closed loop configuration is more efficient than the open loop configuration. Moreover, in the experiments with PET only a very small amount of water evaporated, leading to a much lower relative humidity than observed in this process diagram.

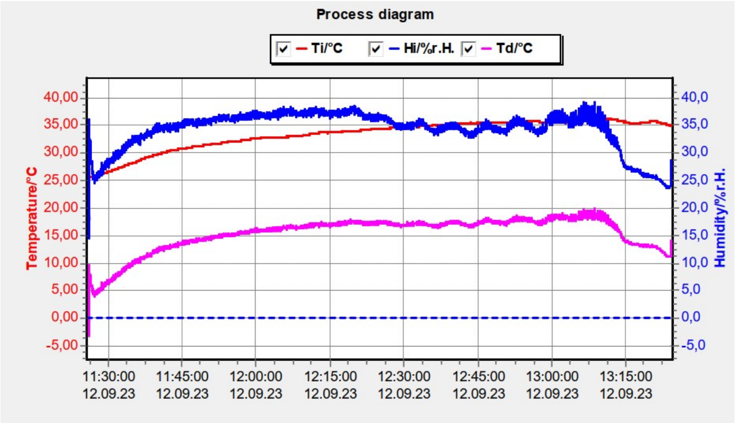
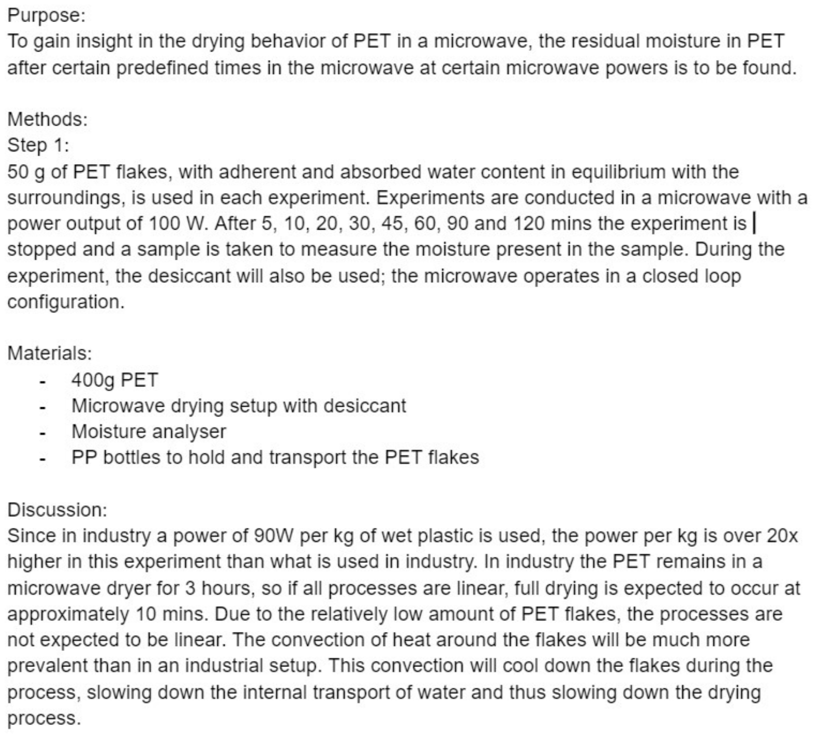


Figure 57: Measuring the temperature and the relative humidity at the microwave outlet in the closed loop configuration

III.2.3 Pellets or flakes

Initially, flakes taken from the recycling process were used. These flakes have just been washed and contained soap, water and the composition of these plastics was not consistent and unknown. The research plan is shown in figure 58. During these experiments, it became evident that the composition of the specimens was very inconsistent. One experiment was conducted after manual sorting, but this proved to be too time consuming to be a feasible solution.



Purpose:
To gain insight in the drying behavior of PET in a microwave, the residual moisture in PET after certain predefined times in the microwave at certain microwave powers is to be found.

Methods:
Step 1:
50 g of PET flakes, with adherent and absorbed water content in equilibrium with the surroundings, is used in each experiment. Experiments are conducted in a microwave with a power output of 100 W. After 5, 10, 20, 30, 45, 60, 90 and 120 mins the experiment is stopped and a sample is taken to measure the moisture present in the sample. During the experiment, the desiccant will also be used; the microwave operates in a closed loop configuration.

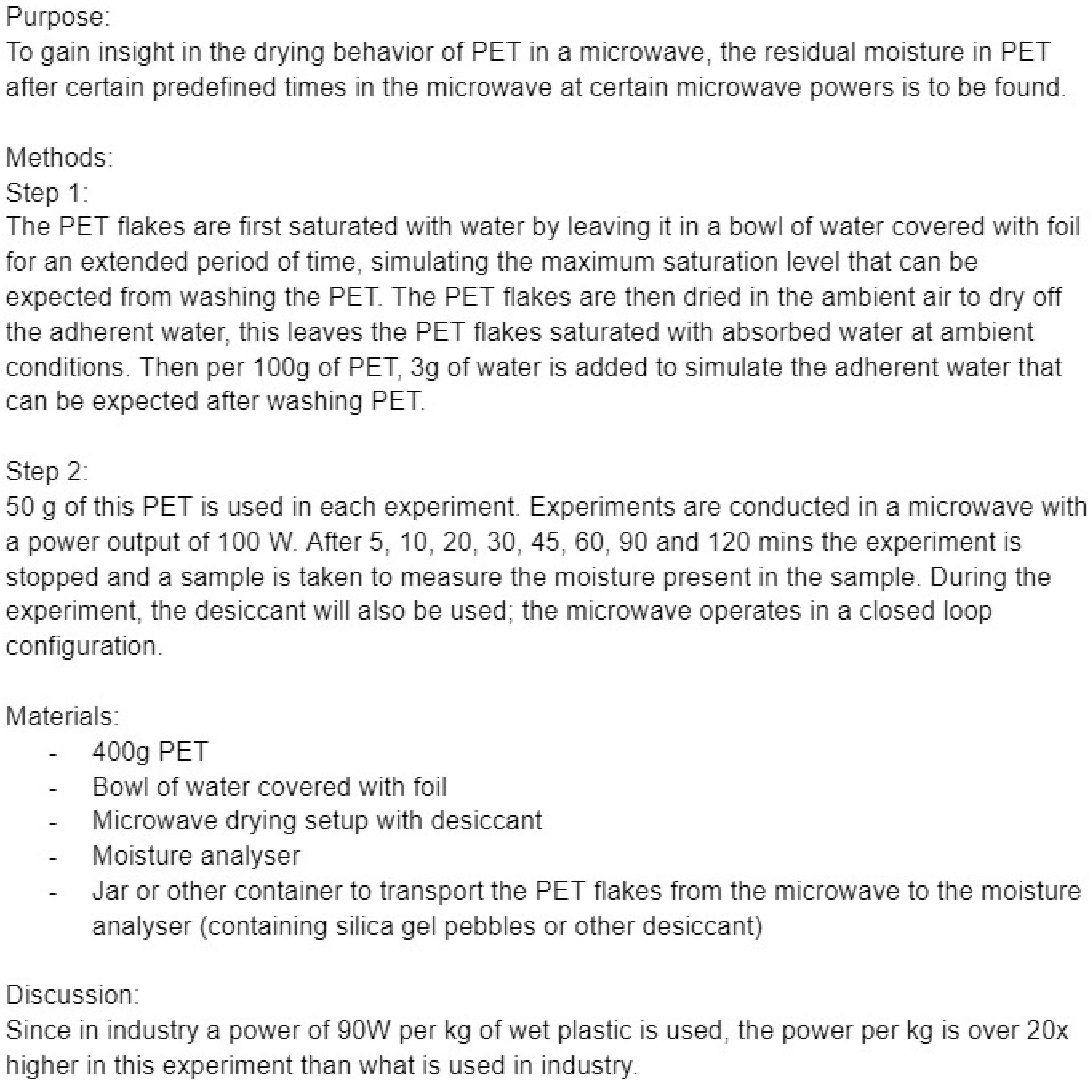
Materials:

- 400g PET
- Microwave drying setup with desiccant
- Moisture analyser
- PP bottles to hold and transport the PET flakes

Discussion:
Since in industry a power of 90W per kg of wet plastic is used, the power per kg is over 20x higher in this experiment than what is used in industry. In industry the PET remains in a microwave dryer for 3 hours, so if all processes are linear, full drying is expected to occur at approximately 10 mins. Due to the relatively low amount of PET flakes, the processes are not expected to be linear. The convection of heat around the flakes will be much more prevalent than in an industrial setup. This convection will cool down the flakes during the process, slowing down the internal transport of water and thus slowing down the drying process.

Figure 58: Research plan drying recycled materials in the microwave

To acquire a source of PET flakes with less inconsistency in the composition, PET bottles were collected. A bulk of pure PET bottles was not readily available, so the collection of these bottles proved laborious. When 500 g of bottles were collected, the experiment shown in figure 59 was performed. The collected PET bottles had to be ground in smaller flakes. The available grinder needed even larger quantities of material to function, so the fines were made by cutting the material with scissors, again labor intensive.



Purpose:
To gain insight in the drying behavior of PET in a microwave, the residual moisture in PET after certain predefined times in the microwave at certain microwave powers is to be found.

Methods:
Step 1:
The PET flakes are first saturated with water by leaving it in a bowl of water covered with foil for an extended period of time, simulating the maximum saturation level that can be expected from washing the PET. The PET flakes are then dried in the ambient air to dry off the adherent water, this leaves the PET flakes saturated with absorbed water at ambient conditions. Then per 100g of PET, 3g of water is added to simulate the adherent water that can be expected after washing PET.

Step 2:
50 g of this PET is used in each experiment. Experiments are conducted in a microwave with a power output of 100 W. After 5, 10, 20, 30, 45, 60, 90 and 120 mins the experiment is stopped and a sample is taken to measure the moisture present in the sample. During the experiment, the desiccant will also be used; the microwave operates in a closed loop configuration.

Materials:
- 400g PET
- Bowl of water covered with foil
- Microwave drying setup with desiccant
- Moisture analyser
- Jar or other container to transport the PET flakes from the microwave to the moisture analyser (containing silica gel pebbles or other desiccant)

Discussion:
Since in industry a power of 90W per kg of wet plastic is used, the power per kg is over 20x higher in this experiment than what is used in industry.

Figure 59: Research plan drying collected PET bottle flakes in the microwave

These two experiments led to the conclusion that virgin PET pellets are the best option to use in the experiments, because they provide a consistent composition and are readily available in large quantities.