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Remediated thermal-treated soil and tar-containing asphalt as secondary filler and sand in self-compacting concrete

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ABSTRACT

The shortage of high-quality fine aggregate as an essential component of concrete has become an emerging world-wide concern for the construction industry . Concrete typically comprises up to 30% fine aggregate, which largely influence the strength and durability of the final product. Therefore, finding suitable substitutes for natural fine aggregate has become an important aspect of current concrete research.

In this study, we investigated the suitability of using remediated thermal-treated soil and tar-containing asphalt as secondary raw materials in self-compacting concrete (SCC) mixture. The remediated materials were used as both (1) fine aggregate replacement to replace all the river sand, and (2) partial filler/supplementary cementitious material (SCM) replacement. The modified Andreasen and Andersen (A&A) particle packing model was used to determine the optimal replacement level. Based on the optimal mixture design, the impact of the replacement on the fresh and mechanical properties of SCC was evaluated. Additionally, the pozzolanic reactivity of the fine fraction (<125 μm) within the secondary sand was assessed and compared to that of limestone powder. Our findings confirm that using remediated thermal-treated soil and tar-containing asphalt can produce a more circular, sustainable SCC by replacing high-quality natural sand and limestone filler and reducing the environmental impact of conventional SCC. This study contributes to finding viable alternatives to natural fine aggregate and promotes the use of recycled materials in construction.

Key Words: Thermal-treated soil, tar-containing asphalt, recycling, secondary raw materials, self-compacting, concrete

INTRODUCTION

The shortage of high-quality fine aggregate as one of the essential components of concrete has become an emerging world-wide concern for our construction industry [1]. Concrete typically contains up to 30% fine aggregate and their quality thereby can significantly influence the strength and durability of the final product. Thus, finding suitable replacements for silicate-rich river sand and other high-quality fine aggregate has become an important aspect of current concrete research. Meanwhile, the Dutch government aims to be fully circular by 2050 [2]. To achieve this goal, the economy is to operate entirely on recycled raw materials and all waste is to be converted into usable materials, thus providing an alternative to ever scarcer primary raw materials.

Besides the conventional fine recycled aggregate sourced from construction demolished waste (CDW), abundant secondary raw material can be harvested from remediated fractions of contaminated soil and tar-containing asphalt granulate (TAG), both of which show abundant availability within the Netherlands. One on hand, soil can be contaminated by a variety of organic compounds, e.g., oil residues, which severely limits its uncontained use in the environment. In year 2019, roughly 3.2 million ton (Mton) contaminated soil was treated [3]. On the other hand, reclaimed TAG are also considered contaminated since it contains carcinogenic polycyclic aromatic hydrocarbons (PAHs). As a result, they cannot be reused freely within new asphalt pavements in the Netherlands since 2017, when legislations have been passed to limit the tolerable concentration of PAHs. It is estimated that 60 Mtons of tar-containing asphalt has been laid in the Netherlands along. More than 20 Mton of TAG has already been reclaimed and the remaining 40 Mton will be removed over the coming decades, as part of infrastructure maintenance and renewal. It is estimated that the annual TAG stream in the Netherlands is roughly 1.4 Mton per year [4]. This quantity may increase due to TAG imports from other countries within European Union, e.g., Germany and Belgium.

For both waste fractions, thermal cleaning of soil and TAG is an innovative and effective treatment method. In 2019, 238 kt of contaminated soil was thermally remediated to allow other applications of the soil [3].As for the TAG, the Environmentally Responsible Road Management Code was introduced in 2008 [5]. By signing the Code, road management authorities declare that any TAG removed must always be cleaned thermally until 2035. In general, thermally cleaned, high-quality material (Soil and TAG) can be used in new products without risk with a declaration from the covenant partners. In fact, the use of thermal-treated soil and TAG, comprising filler, fine and coarse aggregate, has been gaining in popularity and research interest, in part due to the enormous scale of concrete production and the resulting potential for carbon emissions reduction [6].

Within Renewi Mineralz & Water, up to 2 Mton of waste is processed annually and 95% of them have been transformed into reusable new materials. The contaminated soil and TAG is thermally-treated together to obtain secondary raw materials such as filler, sand and gravel. Among them, the secondary sand fraction with a particle size range of 0/4 mm (0/0.157 in.) meets the generic requirements for class industrial soil. Furthermore, it has the potential to be used as an alternative to primary sand in the concrete industry and major infrastructure projects . The fine fractions <0.125 mm (<0.0049 in.) also has potential to replace partial primary filler. As a result, using secondary sand from thermal-treated soil and TAG could help to preserve natural resources and meanwhile reduce the embodied CO_2 emissions of the concrete. However, knowledge gaps still exist in research and application on using these fractions within concrete. Their influence on the fresh properties, mechanical properties, and environmental impact of concrete have not yet been investigated. Their potential use in high-value construction materials such as self-compacting concrete (SCC) is also largely unknown. This has already created tangible obstacles for sustainability transition and circularity within the concrete sector.

In this study, we investigated the feasibility of using remediated thermal-treated soil and TAG as secondary raw materials (hereafter secondary sand) in self-compacting concrete (SCC) mixture. These secondary raw materials were used for both (1) fine aggregate replacement to replace the river sand, and (2) partial filler/supplementary cementitious material (SCM) replacement. The modified Andreasen and Andersen (A&A) particle packing model was used to determine the optimal replacement level. Based on the optimal mixture design, the impact of the replacement on the fresh and mechanical properties of SCC was evaluated. Additionally, the pozzolanic reactivity of the filler fractions within the was assessed and compared to that of limestone powder. The environmental impact of using secondary sand in SCC was also assessed.

MATERIALS

The primary raw materials used to prepare SCC in this study were CEM III/A 52.5 N, limestone filler, natural sand 0/4, and gravel 4/16. The secondary fine aggregate used was the FORZ®Sand 0/4 secondary sand (SS) produced by Renewi Mineralz & Water locally in the Netherlands. The physical properties of raw materials including density, medium particle size and the water adsorption of fine and coarse aggregate are given in Table 1. The chemical composition measured by X-ray fluorescence (XRF) analysis and the physical properties of the cement, limestone filler and two kinds of sand are shown in Table 2.

Table 1 Physical properties of raw materials

Materials	Density	Water adsorption	D50
Unit	kg/m^3 (lb/ft^3)	%	µm (in.)
CEM III/A 52.5 N	3100 (193.5)	-	7.9 (0.0003)
Limestone filler	2650 (165.4)	-	15.6 (0.0006)
Natural Sand 0/4	2630 (164.2)	0.5	522 (0.0205)
FORZ®Sand 0/4	2580 (161.0)	1.0	336 (0.1323)
Gravel 4/16	2630 (164.2)	0.7	10064 (0.3962)

Table 2 Chemical compositions of cement, filler, and sand

Oxide (wt.%)	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	$CaCO_3$
CEM III/A 52.5 N	22.74	5.78	2.73	55.82	7.35	3.76	0.11	0.43	-
Limestone Filler	0.34	0.2	0.07	-	4.2	0.05	0.02	0.01	97.46
River Sand 0/4	95.45	0.84	0.48	0.82	1.71	0.10	-	-	-
FORZ®Sand 0/4	85.55	3.62	1.57	3.98	0.47	0.20	0.41	1.10	-

METHODS

Mixture design of SCC with secondary sand using particle packing model

In this study, the SCC mixtures with secondary sand (SS) were designed following optimum particle packing determined by the modified Andreasen and Andersen (A&A) model [7]. This continuous particle packing model have been proven to be effective for mixture design for SCCs [8] as well as ultra-high performance concrete (UHPC)[9]. The modified A&A model for mixture PSD is shown in Equation (1):

$$P(D) = \frac{D^q - D_{min}^q}{D_{max}^q - D_{min}^q} \tag{1}$$

where P is the cumulative volume fraction of particles that are finer than a certain diameter D, D_{min} and D_{max} are the minimum and maximum diameter of solid particles, respectively, and q is the distribution modulus. The value of q here determines the proportion between the fine and coarse particles in the mixture. Brouwers et al. [10] reported that a q value range of 0 to 0.28 would theoretically lead to an optimal packing of concrete. In this study, the q is fixed as 0.23 following the studies by Hunger et al. [11], which recommended using q in the range of 0.22 to 0.25 for the mixture design of SCC.

The volume proportions V_i (%) of each individual material in Eq. (2) are adjusted until an optimum fit between the particle size distribution (PSD) of the composed mixture P_{mix} and the target $P_{optimum}$ is reached. This was achieved using a MATLAB optimization algorithm based on the Least Squares Method (LSM) as shown in Eq. (3). When the deviation between the target curve and the composed mix, expressed by the sum of the squares of the residuals (RSS) at defined particle sizes, is minimized, the packing of the concrete composition of the concrete can be considered as optimum.

$$P_{mix} = \sum_{i=1}^{n} V_i \, PSD_i \tag{2}$$

$$RSS = \sum_{i=1}^{n} \left[P_{mix}(D_i) - P_{optimum}(D_i)\right]^2 \tag{3}$$

Following the optimization in the particle packing, the optimized grading curves of the SCC mixtures REF, SS50, and SS100, together with the target curve and PSDs of all raw materials are illustrated in Fig. **1**. In general, the reference concrete mixture (SCC-REF) has high limestone filler content. In mixture SCC-SS50 and SCC-SS100, 50% and 100% of natural sand was replaced by FORZ®Sand, respectively.

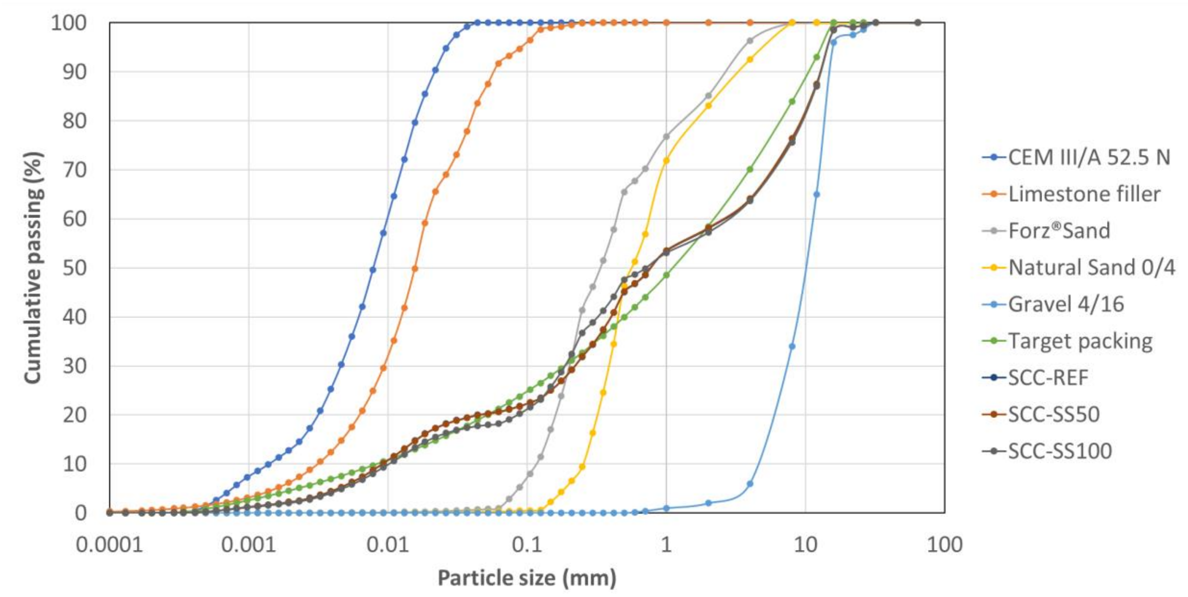

Fig. 1 Optimized grading curves of the SCC mixtures REF, SS50, and SS100 together with the target curve and PSDs of all raw materials

The developed SCC mixtures with secondary raw materials are listed in Table 3. Owning to the higher content of filler fractions (<0.125 mm) in FORZ®Sand, the use of limestone filler is also reduced by 33.6% and 61.8% in SCC-SS50 and SCC-100, respectively.

Table 3 Mixture design of self-compacting concrete

Materials Unit	SCC-Ref kg/m^3 (lb/ft^3)	SCC-SS50 kg/m^3 (lb/ft^3)	SCC-SS100 kg/m^3 (lb/ft^3)
CEM III/A 52.5 N	350 (21.8)	350 (21.8)	350 (21.8)
Limestone filler	200 (12.5)	146 (9.1)	84 (5.2)
Natural sand 0/4 mm	813 (50.7)	457 (28.5)	-
FORZ®Sand 0/4 mm	-	444 (27.7)	933 (58.2)
Gravel 4/16 mm	792 (49.4)	750 (46.8)	772 (48.2)
Superplasticizer	3.25 (0.2)	3.25 (0.2)	3.25 (0.2)
Water	175 (10.9)	175 (10.9)	175 (10.9)
Total powder content*	550 (34.2)	546 (34.1)	542 (33.8)
Water/cement ratio	0.5	0.5	0.5
Water/powder ratio	0.318	0.321	0.324

*Note: The total powder content considers the content of cement, limestone filler as well as fine fractions (<0.125 mm) in the sand fraction.

Reactivity test of the filler fraction within secondary sand

Since the fraction under 0.125 mm (0.0049 in.) within FORZ®Sand can serve also as limestone filler replacement in SCC, it should have at least similar reactivity in comparison to limestone filler so that it will not negatively influence the strength development. To evaluate the reactivity of this filler fraction (<0.125 mm) within FORZ®Sand, the FORZ®Sand 0/4 was first sieved using the 0.125 mm (0.0049 in.) sieve and was then milled using a lab-scale ball mill until a similar PSD as the limestone filler was reached. The PSDs of the original FORZ®Sand 0/4, FORZ®Sand 0/0.125 sieved/milled, and limestone filler are shown in Fig. 1. As illustrated, the FORZ®Sand 0/0.125 mm after milling has almost identical PSDs as limestone filler.

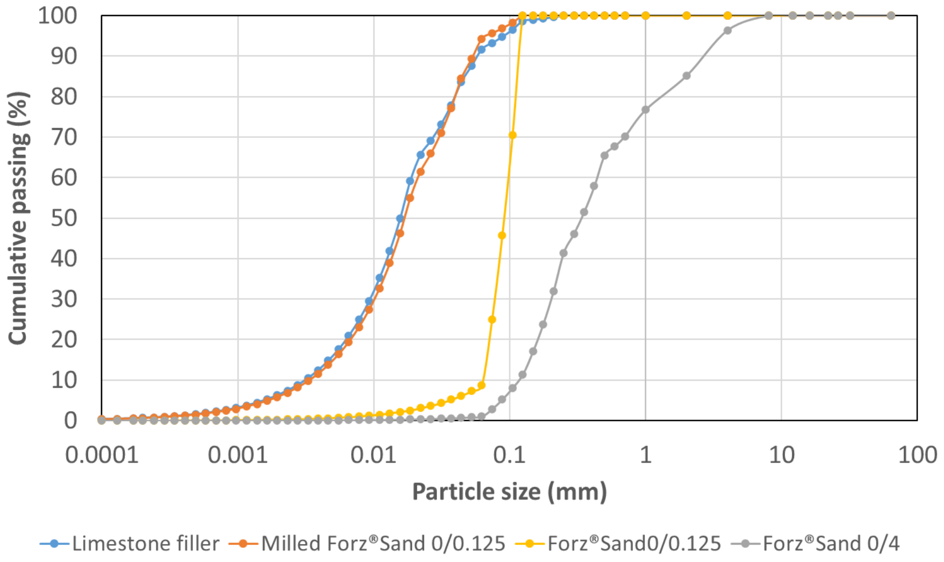

Fig. 2 Particle size distributions of the original FORZ®Sand 0/4, FORZ®Sand 0/0.125 after sieving, milled FORZ®Sand 0/0.125, and limestone filler

The reactivity of the milled FORZ®Sand 0/0.125 fraction was then compared with that of limestone filler by checking the compressive strength development of the cementitious pastes with same fillers content. The binder of the paste consists of 50 wt.% of CEM I 42.5 N and 50 wt.% filler fraction (milled FORZ®Sand 0/0.125, or limestone filler). The water to binder (w/b) ratio was kept constant as 0.5. The two paste samples were cast into 40 mm x 40 mm x 160 mm (1.57 in. x 1.57 in. x 6.30 in.) polystyrene molds. After demolding, the compressive strength of the two paste samples were tested at 3, 7, and 28 days following EN196-1. The similar PSDs of the two filler fractions guarantees the main influence on the compressive strength development is owing to the differences in the cement hydration process rather than the differences in particle packing.

Fresh properties test

The fresh properties of SCC mixtures were tested 15 min after mixing in accordance with EN 12350-12. The J-ring slump flow was used to examine the flowability of the SCC and also to determine the slump flow class. Additionally, the viscosity was determined using the time for the J-ring flow to reach a 500 mm (19.68 in.) spread ($T500_J$) as well as the flow time in V-funnel tests. Furthermore, the passing-ability of the SCC was also assessed during the J-ring flow tests, which was calculated as the differences between the height difference or so-called blocking step (B_J) due to the blockage of the flow by the J-ring. Smaller B_J thereby indicates higher passing-ability of the mixture. Lastly, the consistency/segregation resistance of the SCC was also characterized using the sieve segregation test according to EN 12350-11. The result is the percentage of fine materials passing through the sieves.

Mechanical properties test

After the testing of the fresh properties, the SCC were cast into cubic molds with a size of 150 mm x 150 mm x 150 mm (5.91 in. x 5.91 in. x 5.91 in.). After demolding, the samples were cured in a climate box at 20 °C with relative humidity (RH) >95% until testing age. The mechanical properties including the compressive strength, splitting tensile strength at 1, 7, and 28 days were determined according to the EN 12390-3 and EN 12390-6.

Environmental impact assessment

The environmental impact calculation follows the EN 15804 standard, one of the most significant publications to evaluate the sustainability of constructions materials [12]. The standard proposed 4 life-cycle stages for the calculation of LCA, including product stage, construction process stage, use stage and end of life stage. The scope of the calculation limits to the products stage (“cradle to gate”) due to limited data available for other stages. It is important to note that only the information related to raw materials supply is considered (A1). The information for transportation and manufacturing are considered similar and are thereby not included in this study.

The environmental impact of the developed SCCs were determined in order to quantitatively assess their sustainability in comparison to conventional SCC. The life cycle inventory data of different ingredients used for SCC with FORZ®Sand and conventional SCC preparation are listed in Table 4. The data for primary raw materials were collected either from existed EPD database [13]. It is worth noting that the determination of the environmental impact of FORZ®Sand following the underlying principles of the cut-off approach in EN15804 [12] . The cut-off point between the primary and secondary system complies with the end-of-waste criteria of the standard EN15804. In other words, it is required that the environmental impact of waste processing is fully attributed to the waste producing system rather than the output material after processing. As a result, the Forz®Sand is environmental burden-free.

Table 4 Life cycle inventory data of different ingredients in SCC

Ingredients	Global warming potential kg CO_2 Equiv./kg (lb CO_2 Equiv./lb)	Environmental cost indicator (€/ton)
CEM III/A 52.5 N	0.461	31.04
Limestone powder	0.0310	2.78
Natural sand 0/4	0.0026	0.30
FORZ®Sand 0/4	0	0
Gravel 4/16	0.0039	0.51
Superplasticizer	0.724	91.60
Water [a]	0	0

[a] Assumed to be negligible.

In total three SCCs mixtures were included in this study. In this study, the environmental impact assessment is only limited to global warming potential (GWP) and environmental cost indicator (ECI) to reflect the total environmental impact of the concrete. Both GWP and ECI are associated with functional unit to be 1 m^3 of material. The GWP and ECI were calculated following Eqn. (4) and (5), respectively.

$$GWP_{total} = \sum M_i \cdot GWP_i \tag{4}$$

$$ECI_{total} = \sum M_i \cdot ECI_i \tag{5}$$

Where GWP_{total} and ECI_{total} indicate the GWP (kg CO_2 Equiv./kg) and ECI (€/m^3) of one specific SCC, respectively. M_i is the of mass fraction (kg/m^3 and lb/ft^3) of component in the SCC mixture design presented in Table 3. GWP_i and ECI_i are the GWP and ECI of specific component, which are listed in Table 4.

RESULTS AND DISCUSSIONS

Reactivity of filler fraction within secondary sand

The compressive strength of cement paste blended with 50 wt.% milled FORZ®Sand and limestone filler at 1, 7 , and 28 days are shown in Fig. 3. It is clear that using the milled FORZ®Sand fraction lead to a superior compressive strength development. At 28 days, the compressive strength is increased by 9.1%. Since both the milled FORZ®Sand fraction 0/0.125 and the limestone filler has almost identical PSDs, with the same mixture design of the paste, the superior strength could not be associated with better particle packing. Instead, it is most probably the filler fractions within the FORZ®Sand has a higher degree of reactivity compared to limestone filler. This hypothesis is highly feasible, considering that the fine fraction within FORZ®Sand went through thermal treatment with high average temperature up to 500 °C (932 °F) in the rotary kiln. Some of the clay minerals such as kaolinite (dehydroxylation happens at temperature 400 to 600 °C (752 to 1112 °F) [14]) could be transformed into calcined clay during the treatment process, which can later contribute positively to the hydration process of cement and thereby the strength development.

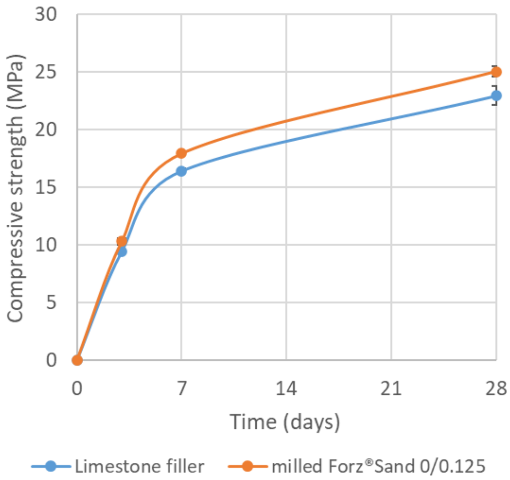

Fig. 3 Compressive strength development of cement paste blended with 50 wt.% milled FORZ®Sand 0/0.125 and limestone filler

Further tests using R3 tests to determine the reactivity of the filler fraction within the FORZ®Sand is recommended, so that direct evidence on the higher reactivity could be obtained. This will not only confirm and clarify its higher reactivity than limestone filler but also give important insights for further optimization of the production process, e.g., thermal treatment process, for further reactivity improvement.

Fresh properties

The J-ring slump flow of the SCC mixtures are shown in Fig. 4. From the visual inspection, all three SCC mixtures exhibit good cohesion without bleeding and segregation. The SCC-REF mixture has a J-ring slump flow of 690 mm (27.2 in.). Increasing the replacement of natural sand by FORZ®Sand increased the J-ring slump flow to 720 mm (28.3 in.) at 50% replacement and 730 mm (28.7 in.) at 100% replacement. According to EN 206-1, all these SCCs meet the SF2 slump flow class.

Fig. 4 J-ring slump-flow of SCC mixtures

Other fresh properties, including J-ring slump flow time T500$_J$ (s), V-funnel flow time (s), blocking step (mm), and segregated mass (%) are summarized and compared in Fig. 5. The T500$_J$ for all SCCs is lower than 2s. Compared to the slump-flow time T500$_J$ of one specified mixture, the T500$_J$ is always higher due to the hindrance of the flow by the J-ring. Therefore, it is conceivable that T500$_J$ for the designed SCC should all be lower than 2s. Additionally, the V-funnel flow time was also measured, which lies within 4.8s to 6.1s. According to EN 206-1 and EN 13369, all SCCs thereby meet the requirements for the viscosity class of VS1 (T500$_J$ <=2s).

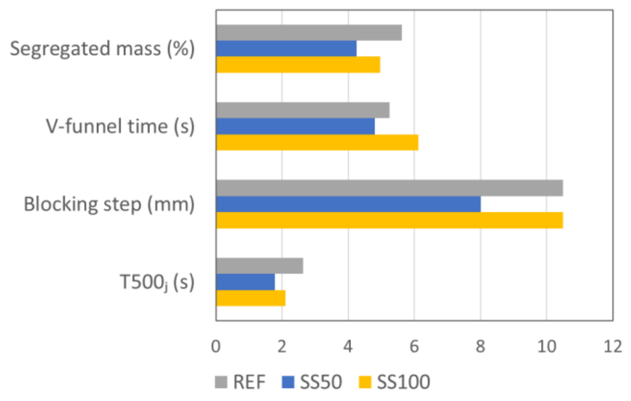

Fig. 5 Fresh properties of SCC: Segregated mass (%), V-funnel flow time (s), blocking step (mm), and J-ring slump flow time T500$_J$ (s)

For the passingability, the SCC-REF has B$_J$ value of 10.5 mm (0.41 in.) while the B$_J$ for SCC-SS50 and SCC-SS100 is 8 mm (0.31 in.) and 10.5 mm (0.41 in.), respectively. The B$_J$ values for conventional SCC normally lie in the range of 0 to 15 mm (0 to 0.59 in.) when a 16 bars J-ring is used [15]. Therefore, the passingability is considered satisfactory for all SCCs. Furthermore, it is found that replacing natural sand by FORZ®Sand does not lead to reduction in passingability. In fact, the passingability of SCC with FORZ®Sand replacement is considered comparable or even better compared to that of SCC-REF. Lastly, the consistency/segregation resistance of SCCs was evaluated by the segregated mass during the sieve tests. For all SCCs, the segregated mass portion is between 4.3% to 5.6%, which meets the SR1 class (SR<23%).

It could be summarized that replacing natural sand using FORZ®Sand in SCC lead to comparable fresh properties of SCC. Considering the flowability, viscosity and passingability of the SCCs, all of them meet the fresh properties requirements for most of the structural applications such as floors and slab, as well as walls and piles [16].

Compressive strength and splitting tensile strength

The compressive strength (fc) of SCCs are given in Fig. 6 (a). The fc for three SCCs at 1d is similar, which is about 26.5 MPa (3843 psi), meeting the production/demoulding requirement for precast industry. At 7 and 28 days, the fc of SCC-SS50 are similar with SCC-REF. However, compared to SCC-REF, SCC-SS100 has a 13.1% higher fc at 7 day and 6.1% higher fc at 28 days. The 28-day compressive strength is 59.3 MPa (8600 psi), which also meets the strength class of C50/60.

Following the discussion in section, it is most probably the filler fraction within the FORZ®Sand has a higher degree of reactivity compared to limestone filler, which contribute positively to the hydration process of cement and thereby the strength development.

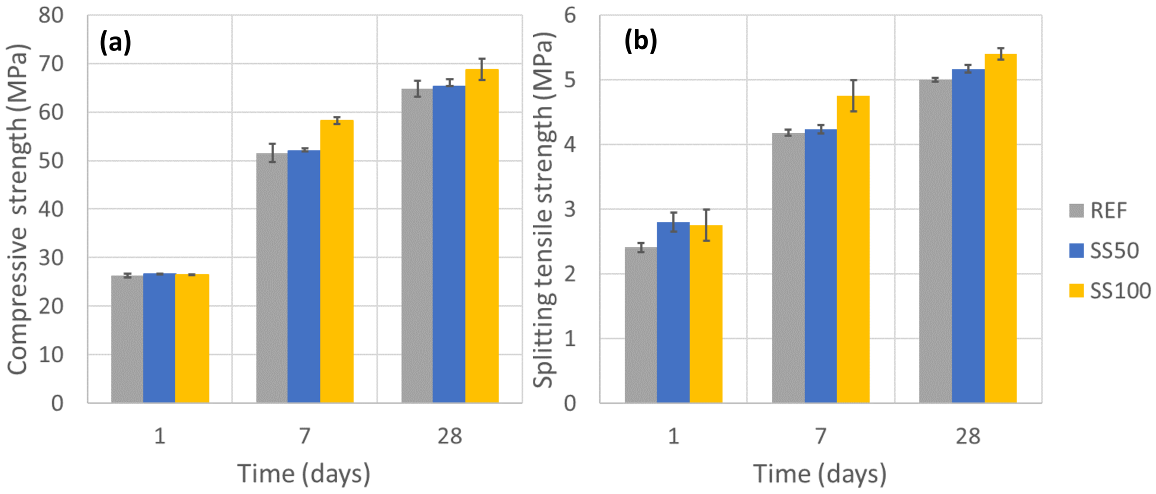

Fig. 6 (a) Compressive strength and (b) splitting tensile strength of SCC mixtures: REF, SS50, and SS100

The splitting tensile strength (ft) of SCCs are given in Fig. **6** (b). The ft development shows a similar trend as the fc whereas some minor difference were observed in 1 day results. At 1 day, SS50 and SS100 seems to have a bit higher ft than SCC-REF.

Environmental impact reduction using FORZ®Sand

The use of FORZ®Sand as replacement for natural sand and filler lead to significant reduction in GWP as well as the total environmental impact of the filler+aggregate fractions in SCC. The total environmental impact can be reflected by the environmental cost indicator (ECI), which is a very simple monetized indicator. It reflects the total environmental impact of the composite materials. An ECI thus suggests a better sustainability performance. In the study here, only the filler+aggregate fraction is considered because the cement content is identical among the three SCC mixtures (Table 3).

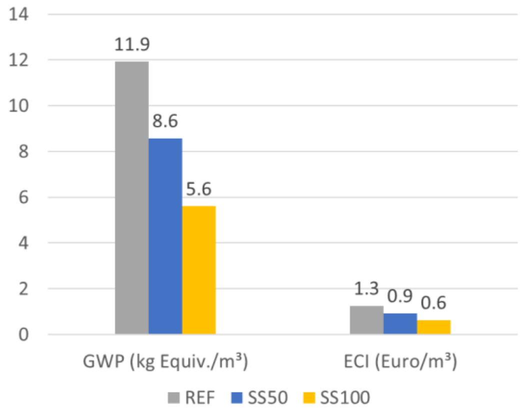

Fig. 7 GWP and ECI of filler +aggregate fractions in SCC mixtures: REF, SS50, and SS100

With the use of FORZ®Sand, the filler+aggregate fraction in SCC-50 has a 28.1% lower GWP and 53.0% lower ECI than the SCC-REF. Compared to that in SCC-REF, the GWP and ECI of filler+aggregate fraction in SCC-SS100 is 53.0% and 49.9% lower, respectively. These results suggests that using FORZ®Sand from remediated thermal-treated soil and tar-containing asphalt can produce a more circular, sustainable SCC compared to SCC produced using only primary raw materials.

CONCLUSIONS

This study explores the suitability of using remediated thermal-treated soil and tar asphalt as secondary raw materials in SCC. Following the modified Andreasen and Andersen (A&A) particle packing model, SCC with 50 wt.% and 100 wt.% replacement of river sand with also partial replacement of limestone filler has been successfully developed. The following conclusions could be draw:

- Using modified A&A particle packing model proves efficient in develop SCC mixtures with secondary raw materials.
- The FORZ®Sand can effectively replace 100% natural sand and up to 61.8% limestone filler in conventional reference SCC.
- The fresh properties and mechanical properties of SCC with FORZ®Sand replacing natural sand and limestone filler is comparable to those of conventional reference SCC.
- It is most probable that the filler fraction within FORZ®Sand has higher pozzolanic reactivity than limestone filler.
- Using remediated thermal-treated soil and tar-containing asphalt can produce a more circular, sustainable SCC by replacing high-quality natural raw materials and reducing the environmental impact of the filler and aggregate fractions within conventional SCC. This study contributes to finding viable alternatives to natural fine aggregate and promotes the use of recycled materials in construction.

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