



Mechanical and physical characteristics of alkali- activated mortars incorporated with recycled polyvinyl chloride and rubber aggregates

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ABSTRACT

One of the ways to achieving net-zero concept in the construction industry is to use alternatives to Portland cement (OPC) and virgin aggregates for concrete manufacturing. Recycled rubber and polyvinyl chloride (PVC) aggregates in conjunction with low-carbon binders can be potentially utilised to substitute natural sand and reduce the negative environmental impacts of OPC. A replacement of natural sand (up to 70% by volume) in alkali-activated materials (AAMs) with recycled rubber and PVC particles derived from tyre waste and insulation coating of electric wires, respectively, was investigated in this study. The performance of developed AAMs was evaluated using a comprehensive testing program including mechanical, physical and micro-structure assessments. AAM composites with PVC and rubber particles outperformed natural aggregate composites in terms of thermal resistivity, water absorption, volume permeability voids (VPV), and high-frequency sound insulation. Results showed that 70% PVC mixture achieved the lowest water absorption rate and thermal conductivity with a reduction of 73% and 20%, respectively, compared to the control mixture. A maximum reduction of 34% in VPV was observed in the 70% rubber mixture when compared to the control mixture. In terms of mechanical properties of waste stream aggregates, PVC outperformed rubber. The results showed that 30% replacement of PVC and rubber would produce composites with 7-day compressive strengths of 35 MPa and 25 MPa, respectively, which can be used to produce high-load bearing structures. The Energy-dispersive X-ray Spectroscopy (EDX) was performed to detect chloride leaching from PVC aggregates, where results indicated that no leaching had occurred after more than 90 days of casting. Regarding the carbon emission, the carbon footprint of AAM composites is decreased by using the polymeric fractions in place of sand. The developed composites of this study can be used safely in non-load bearing structural elements with promising physical and mechanical performance.

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1. Introduction

The disposal of synthetic plastic and rubber wastes in landfills is one of the causes of global warming and climate change. Lowering CO₂ emission is a fundamental step to nature's recovery, rebalancing the ecosystem and mitigating global warming's negative impact on the planet. By 2050, Eleven billion metric tonnes of plastic are expected to be accumulated, which will emit about 6.5 Gt CO₂ -eq into the environment [1,2]. On the other hand, approximately one billion tyres are produced globally each year, and almost 50% of production ends up discarded in landfills after completing their service life. Moreover, it is expected that 5 billion tyres will be manufactured by 2030, with 1.2 billion tyres discarded in landfills, which will pose a severe threat to the ecosystem balance [3]. Additionally, marine species easily ingest plastic wastes dumped in the marine ecosystem, which might lead to higher fatality rates between these species and the pollution of the marine ecosystem [4].

Polyvinyl chloride (PVC) waste, explored as a replacement for natural aggregates in this study, is one of the most harmful plastic wastes to the environment due to the high constituent of Chloride in its chemical structure (about 57%) [5]. The valorisation of such harmful waste into concrete and employing it as a construction material might reduce its hazardous effect on the environment.

The sustainability credential of the proposed work in this study is related to the traditional concrete, which is considered a non-environmentally friendly product as it consumes not only natural resources of sand and gravel at critical levels but also its main ingredient, Portland Cement (PC), is responsible for about 8% of the global CO₂ emissions [6]. Traditional concrete consists of 50%–80% natural aggregates [7], and it is the most consumed material on earth, at about 25 gigatonnes every year and around 3.5 tonnes per capita [8]. Furthermore, every year, 26 to 30 billion tons of aggregate are consumed globally in the concrete industry, where sand and gravel share about 65%–85% of the consumed aggregates [9]. Several types of natural aggregates alternatives such as construction demolition waste materials (CDW) [10]. Similarly, plastic and/or rubber aggregates have been employed as a unique way to reduce landfill waste [11]. Moreover, various waste materials were employed as aggregates to produce pavement porous concrete [12] and pervious concrete [13]. On the other hand, natural aggregates such as sand or gravel have rough surfaces and more wettability than plastics, which play a critical role in increasing the binding with the cementitious matter and enhancing cementitious composites' performance [14]. Due to the hydrophobic nature, smooth surface, and low fire resistance of plastic particles, it became essential to pre-treat plastics before their integration into concrete to improve their function as aggregates. Similarly, physical and chemical pre-treatment of rubber particles is also needed to enhance their performance when used as aggregates [15]. Physical pre-treatment is simply washing rubber to remove dirt and impurities and improve rubber surface wettability. At the same time, chemical pre-treatment exposes the rubber surface to various chemical solutions or pre-coating it with cementitious materials to make its surface more hydrophilic [15]. The challenge is to reduce the gap in performance between conventional and recycled materials [16]. One more serious problem this study deals with is the PC presence in concrete and its related issues with CO₂ emission. Alkali-activated materials (AAMs), including geopolymers, are a suitable alternative generated by using by-product materials of other industries such as fly ash, silica fume, metakaolin and ground granulated blast slag (GGBS), which are activated by alkali solutions or solid powders, it worth nothing that binder type and activators ratios affecting the bulk characteristics of final composites [17]. These materials could have 5 to 6 times less CO₂ emissions than PC [18]. Additionally, AAMs have excellent thermal stability, chemical resistance, and minimal shrinkage. Using AAM concrete instead of PC would save 0.25 million tonnes of coal and 80 million power units and prevent 1.5 million tonnes of CO₂ emissions [19].

Zhang et al. (2021) [20] stated that 10% replacement of river sand with crumbs rubber in geopolymer mixtures improved both mechanical strengths and freeze-thaw resistance. Mechanical strengths improvement may be attributed to rubber pre-treatment with NaOH, which enhanced surface roughness and adhesion. It is worth noting that only up to 10% rubber replacement improved mechanical and physical properties, while replacement beyond 10% resulted in declining matrix bonding structure. Freeze-thaw resistance development is credited to rubber's good energy absorption and hydrophobicity; however, the matrix structure becomes more porous and develops cracks with the increase in rubber crumb replacement ratio. Valente et al. (2021) [21] evaluated the effect of waste tyre rubber aggregates on the mechanical and microstructural properties of OPC and AAM matrixes. The rubber altered the chemistry and the proper strength development in AAM samples, implying more significant mechanical losses under compression. However, improved rubber-matrix compatibility was observed in the geopolymer mixes due to the influence of mix design components (alkali activator and silica fume) on the chemical-physical functionalization of rubber, inducing an exemplary behaviour under bending.

Hu et al. (2021) [22] used PVC as coarse aggregates (14.75–14 mm) in OPC cementitious composites at different replacement ratios (0–30%). They concluded that 20% PVC coarse aggregates replacement was ideally for gunite machines used in tunnels and roadways support that produced lightweight, elastic, crack resistance and effective energy absorption concrete. Ceran et al. (2019) [23] investigated PVC as fine aggregates in different replacement ratios with river sand (0–45%) in an OPC composites-based concrete. The addition of PVC reduced both compressive and tensile strength between (28–80%) and (5–50%), respectively, depending on the PVC content in the concrete mixes. However, introducing PVC fine aggregates to OPC improved the composite's thermal and electrical resistivity between (35–46%) and (25–125%), respectively, depending on the PVC content.

This study aims to develop a low carbon cementitious composite with incorporated waste to replace natural aggregates. Alkali-activated materials are used with three replacement ratios of PVC and rubber with natural sand, namely, 30%, 50% and 70%. A comprehensive analysis is then carried out on the resultant specimens and benchmarked against control samples made with 100% sand aggregates. Most of the studies focusing on replacing natural sand with waste stream aggregates are carried out on OPC-based cementitious composites. To the best of the authors' knowledge, there is a limited number of studies on AAMs with waste aggregate replacements. Moreover, there are no studies, to the best of author's knowledge, on recycled PVC incorporation in AAM-based cementitious composites, however, the group has carried out investigations on rubber aggregates in AAM [24].

2. Experimental framework

Fig. 1 details the experimental framework and comprehensive characterisation techniques adopted in this study to thoroughly investigate the performance of low-carbon cementitious composites with waste aggregate replacements.

2.1. Materials

The alkali-activated cementitious composite in this study is constituted of (1) fly ash (Cemex, UK) which meets the normal fineness (N) Category of British standard BS EN 450–1:2012; (2) ground-granulated blast furnace slag (Hanson Heidelberg Cement, UK), which complies with EN15167-1; (3) micro-silica fume (J. Stoddard & Sons Ltd); (4) graded sand including two different sand sizes of 0–0.5 mm and 0.5–1.0 mm following BS EN 410–1:2000; (5) sodium silicate (Na_2SiO_3) solution with the $\text{SiO}_2/\text{Na}_2\text{O}$ mass ratio of 3.23 (Solvay SA, Portugal); (6) 10 mol/l sodium hydroxide (NaOH) solution (Fisher Scientific, Germany); and (7) attapulgite nano-clay (supplied by Attagel 350, Lawrence Industries Ltd., UK).

Ground tyre rubber aggregates supplied by the European Tyre Recycling Association (ETRA, Brussels, Belgium) and PVC aggregates provided by AlterEco (Padova, Italy), were incorporated as natural aggregate replacements. Rubber and PVC aggregates were produced by mechanical grinding of scrap tyres and insulation coating of electric wires, respectively, at ambient temperature. Thereupon, both aggregates were sieved to achieve a 0–2 mm nominal size gradation. Microstructure analysis of recycled aggregates demonstrated rough rounded surfaces with jagged edges for PVC aggregates (Fig. 2-a), while smooth flat surfaces with straight sharp edges for rubber aggregates (Fig. 2-b).

2.2. Mix formulation and material preparations

A total of seven (see Table 1) AAM mixtures were produced. The control AAM mix formulation was selected based on the authors' previous research, where the mechanical performance, workability and durability of the control mix were optimised [25]. The AAM mixtures were prepared in three separate stages. First, the precursor materials, including FA, GGBS, SF, attapulgite (AT) nano-clay additive, and aggregates, including graded sand, rubber, and PVA, were mixed for 5 min at 250 rpm in dry state employing a planetary mixer (Kenwood, Germany). In this study, natural sand aggregates were replaced by recycled aggregates (i.e., rubber and PVC) with fractions of 30 vol.-%, 50 vol.-%, and 70 vol.-%. The dosage of attapulgite nano-clay additive was constant and set to 1% by weight of binder based on the authors' previous study. Secondly, the alkali activator solutions (i.e., a combination of NaOH and Na_2SiO_3) with a fixed mass ratio of 1:2 were mixed for 5 min using a magnetic stirrer. Finally, premixed alkali solutions were gradually added to the dry mixture and mixed for 10 min at 450 rpm until the homogeneous AAM mixtures were achieved. After the mixing process, AAM fresh mixtures were cast using prismatic moulds with dimensions of $160 \times 40 \times 40 \text{ mm}^3$ (three samples of each mix) and placed in the oven for 24 h at 60°C (heat curing stage), followed by 6 d and 89 d (air curing stage) at ambient temperature.

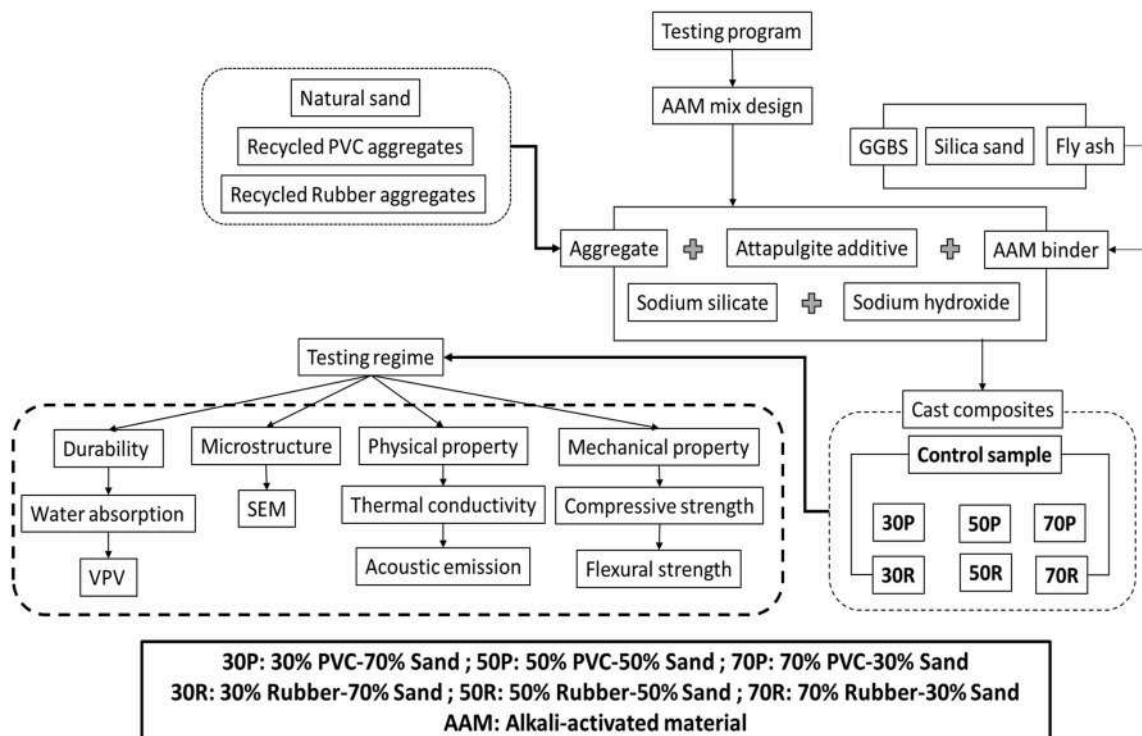


Fig. 1. Experimental design and analysis framework.

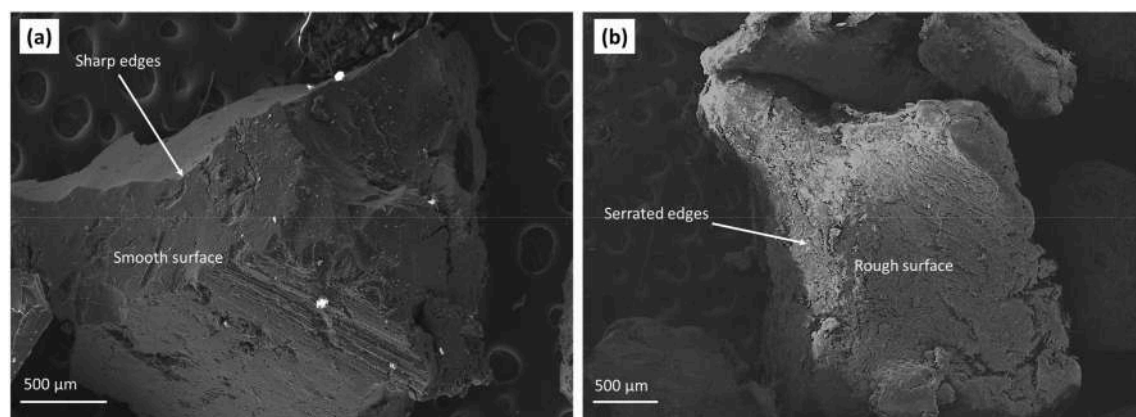


Fig. 2. SEM images of (a) rubber and (b) PVC recycled aggregates.

Table 1
Mix formulation details for AAMs with PVC and rubber at different replacement ratios.

Sample ID	Binder (wt.%)			Aggregate (vol%)				Nano Clay 1 (wt.%)	Sodium Silicate 66.6 (wt.%)	Sodium Hydroxide 33.3 (wt.%)
	FA	GGBS	Silica sand	Natural sand		Recycled aggregate				
				0–0.5 mm	0.5–1 mm	PVC	Rubber			
Control sample	60 (482.3)	25 (200.9)	15 (1206)	60 (602.1)	40 (401.4)	0	0	1 (7.8)	66.6 (214.3)	33.3 (107.2)
30P	60 (487.4)	25 (203.1)	15 (121.9)	42 (396.1)	28 (264.1)	30 (165.9)	0	1 (7.9)	66.6 (216.6)	33.3 (108.3)
50P	60 (486.4)	25 (202.7)	15 (121.6)	30 (282.3)	20 (188.2)	50 (275.8)	0	1 (7.9)	66.6 (216.2)	33.3 (108.1)
70P	60 (481.2)	25 (200.7)	15 (120.4)	18 (167.8)	12 (111.8)	70 (382.6)	0	1 (7.8)	66.6 (214.1)	33.3 (107.1)
30R	60 (487.8)	25 (203.2)	15 (121.9)	42 (396.4)	28 (264.3)	0	30 (156.6)	1 (7.9)	66.6 (216.8)	33.3 (108.4)
50R	60 (468.9)	25 (195.3)	15 (117.2)	30 (272.1)	20 (181.4)	0	50 (250.8)	1 (7.6)	66.6 (208.4)	33.3 (104.2)
70R	60 (474.1)	25 (197.5)	15 (118.5)	18 (165.1)	12 (110.0)	0	70 (354.5)	1 (7.7)	66.6 (210.7)	33.3 (105.3)

*The amounts between brackets are kg/m³.

2.3. Experimental tests

2.3.1. Microstructure analysis

Scanning electron microscopy (SEM) was used to analyse the microstructure of AAMs (Supra 35VP, Carl Zeiss, Germany). At least ten samples of size 8 mm³ were analysed for each composition. Before the SEM analysis, all the samples were gold-coated (Edwards S150B sputter coater) to promote the electrical conductivity of AAM samples. The chloride leaching within the AAMs containing PVC aggregates was assessed using SEM coupled with energy dispersive spectroscopy (EDS) (EDAX, USA) [26]. Assessing chloride leaching was aimed at detecting the trace of chloride in the mixtures and confirming the safe use of recycled PVC aggregates. To the best of authors knowledge, no research has employed EDS testing for such a reason.

2.3.2. Mechanical properties

According to BS EN 196–1:2016, the mechanical performance of AAM samples (i.e., flexural, and compressive strengths) was evaluated after 7 and 90 days of curing using an Instron 5960 Series Universal Testing System. The aforementioned test method have been used in the authors previous study [27]. The load rate for both flexural and compressive tests was set to 1 mm/min. The mechanical performance of the AAMs was examined after 90 days of curing to evaluate the potential degradation of recycled PVC and rubber in a high alkaline environment [28]. For each composition, three and six specimens were used for the flexural and compressive strength tests, respectively, as per the standard.

2.3.3. The volume of permeable void

Based on the authors previous studies, the boil-water saturation (BWS) method was used on the AAM samples after 7 d of curing to evaluate the volume of permeable voids percentage (VPV) [28]. As per ASTM C 642, a batch of three prismatic samples with

dimensions of $160 \times 40 \times 40 \text{ mm}^3$ was cast for each composition. Then, the weight of samples was measured after oven-drying for 48 h at 110°C (A) and immersing in water for 48 h at ambient temperature followed by submerging in boiling water for 5 h (B). The VPV is an essential property of concrete because it affects the transport mechanism of abrasive liquids and gasses through the concrete [29]. Based on the standard, Eq. 1 was used to calculate the VPV percentage of each sample.

$$\text{Volume of permeable voids (\%)} = \frac{B - A}{B - C} \times 100 \quad (\text{Eq.1})$$

Where A represents the oven-dried mass(g), B represents the saturated surface-dry mass (g) after 48 h of immersion in tap water and 5 h of boiling, and C represents the apparent mass(g) of concrete after immersion in water.

2.3.4. Water absorption

As reported in several research studies, the transport properties of cementitious composites are directly related to their durability [30–32]. In order to assess the durability of developed composites in this study, the water absorption test was carried out in line with ASTM C1585-13 with some modifications. A batch of three cubic samples with dimensions of $50 \times 50 \times 50 \text{ mm}^3$ was examined for each composition. One side of each cube was permitted to absorb water, while the other five sides were sealed with epoxy sealer. The same testing strategy has been employed in a study conducted by Jang et al. (2022) [33]. 0 min, 5 min, 10 min, 15 min, 30 min, 1 h, 2 h, 3 h, 4 h, 6 h, 12 h, 24 h, 48 h, and 72 h are the time intervals for weighing samples. The following equation (i.e., Eq.2) was used to calculate the water absorption.

$$\text{Water absorption (\%)} = \frac{M_t - M_0}{M_0} \times 100 \quad (\text{Eq.2})$$

Where, M_0 (g) represents the oven-dried mass, and M_t (g) represents the saturated surface-dry mass.

2.3.5. Thermal conductivity analysis

The thermal conductivity test of the AAM samples was carried out following the ASTM D7984 employing a C-Therm TCi thermal analyser (C-Therm Technologies, New Brunswick, Canada). The test method has been previously employed in a study conducted by Luo et al. (2016) [34]. A current was applied to an alumina sensor's heating element providing a small amount of heat. The heat provided results in a rise in temperature at the interface between the sensor and the composites, typically $1\text{--}3^\circ \text{C}$. This temperature increment at the interface induced a change in the voltage drop of the sensor element. The rate of increase in the sensor voltage was used to determine the thermal conductivity of the composite materials [34]. Measurements were made at room temperature on a batch of three prismatic samples of $150 \text{ mm} \times 40 \text{ mm} \times 40 \text{ mm}$, considering three measurements for each sample. A contact agent (bidi stiller water) was applied between the sensor and the sample to reduce the thermal resistance to a negligible level. Each testing point involved four thermal conductivity measurements, implying a total of twelve experimental values per formulation. Thermal conductivity value was obtained from the averaged measurement results in each sample.

2.3.6. Acoustic insulation analysis

Sound insertion loss (SIL) is an acoustic quantity used to identify the noise barrier properties of wood, glass, masonry, metal, or concrete materials implemented for noise abatement interventions in the civil and architectural fields. Rigorously, SIL is defined as the reduction of noise level at a given location due to the placement of a noise control device (noise barrier) in the sound path between the sound source and that location [35]. In this research, SIL was considered an indicator to evaluate the influence of polymer aggregates on the noise attenuation performance of the AAM mixes. The experimental characterisation was conducted by impedance tube system, following the ISO 7235 standard method, on a batch of three prismatic samples of $140 \times 40 \times 40 \text{ mm}^3$. Experimental evaluation of the SIL-index measured the sound pressure level reduction when the test material was installed in the tube between two microphonic transducers. More technical details on the measuring system and test method can be found in previous research work [36]. SIL was investigated in two frequency bands: low-medium frequency range (63 Hz, 125 Hz, 250 Hz, 500 Hz) and high-frequency range (1000 Hz, 2000 Hz, 3000 Hz, 4000 Hz). By averaging the SIL values in each frequency range, it is possible to evaluate the Sound reduction index (SRI), a performance indicator that provides a comprehensive and more precise evaluation of the acoustic attenuation behaviour. SRI is a single value (in dB) that defines the reduction in sound intensity when it passes through a partition, determining the effectiveness of the element/material as an acoustic insulator. SRI is assessed in the two frequency bands in agreement with the following Equations (3) and (4):

$$SRI_{\text{low-middle frequency range}} = \frac{SIL_{(63 \text{ Hz})} + SIL_{(125 \text{ Hz})} + SIL_{(250 \text{ Hz})} + SIL_{(500 \text{ Hz})}}{4} \quad (\text{Eq. 3})$$

$$SRI_{\text{high frequency range}} = \frac{SIL_{(1000 \text{ Hz})} + SIL_{(2000 \text{ Hz})} + SIL_{(3000 \text{ Hz})} + SIL_{(4000 \text{ Hz})}}{4} \quad (\text{Eq. 4})$$

2.3.7. Carbon footprint estimation for sand replacement with polymer aggregates

Carbon footprint assessment was implemented in this work with the purpose of evaluating the impact related to the replacement of sand with recycled PVC and tyre rubber aggregates. Following the carbon footprint calculation model and the carbon-emission coefficients (EC) of the raw materials already adopted by the authors in a previous research work [24], the embodied CO₂ (ECO₂) of the AAM composites incorporating the two polymeric fractions was estimated. In assessing the EC-index related to the processing of the

recycled aggregates, Life Cycle Assessment (LCA) SIMAPRO software was used, in compliance with ISO 14040 standard [37]. Both recycling models consist of an input energy stream from the European medium-voltage electricity supply needed for grinding process and an avoided product that corresponds to the amount of recycled material that is recovered. The latter flow has the same dimensions as the functional unit, as it is assumed that all the waste entering the system is processed and made recyclable. Furthermore, the environmental contribution of an avoided product corresponds to a negative value in terms of eco-impact and therefore can be interpreted as a net environmental benefit in terms of carbon footprint. The environmental impacts of the processes were evaluated by means of the IPCC 2021 impact assessment method which provides results in CO₂-equivalent (kg CO₂ emitted/kg recycled material produced). In the recovery processes, secondary data from the Ecoinvent 9.3 database were used, which is recognised as the largest and most consistent life cycle inventory (LCI) database.

3. Results and discussions

3.1. Mechanical properties

The mechanical properties of AAMs are influenced by the type, characteristics, and volume fraction of aggregates. The flexural and compressive strengths of AAMs incorporating rubber and PVC aggregates are shown in Fig. 3. The results revealed that increasing the volume fractions of PVC and rubber aggregates led to a progressive descending trend in both compressive and flexural strengths. This could be attributed to the smooth surface and low specific gravity of plastic/rubber aggregates compared to the natural sand aggregates, resulting in a reduced matrix density and consequently diminished mechanical performance [38]. It has also been reported that the different nature of rubber and PVC from natural aggregates in terms of shape, surface texture, and hydrophobicity inhibits suitable

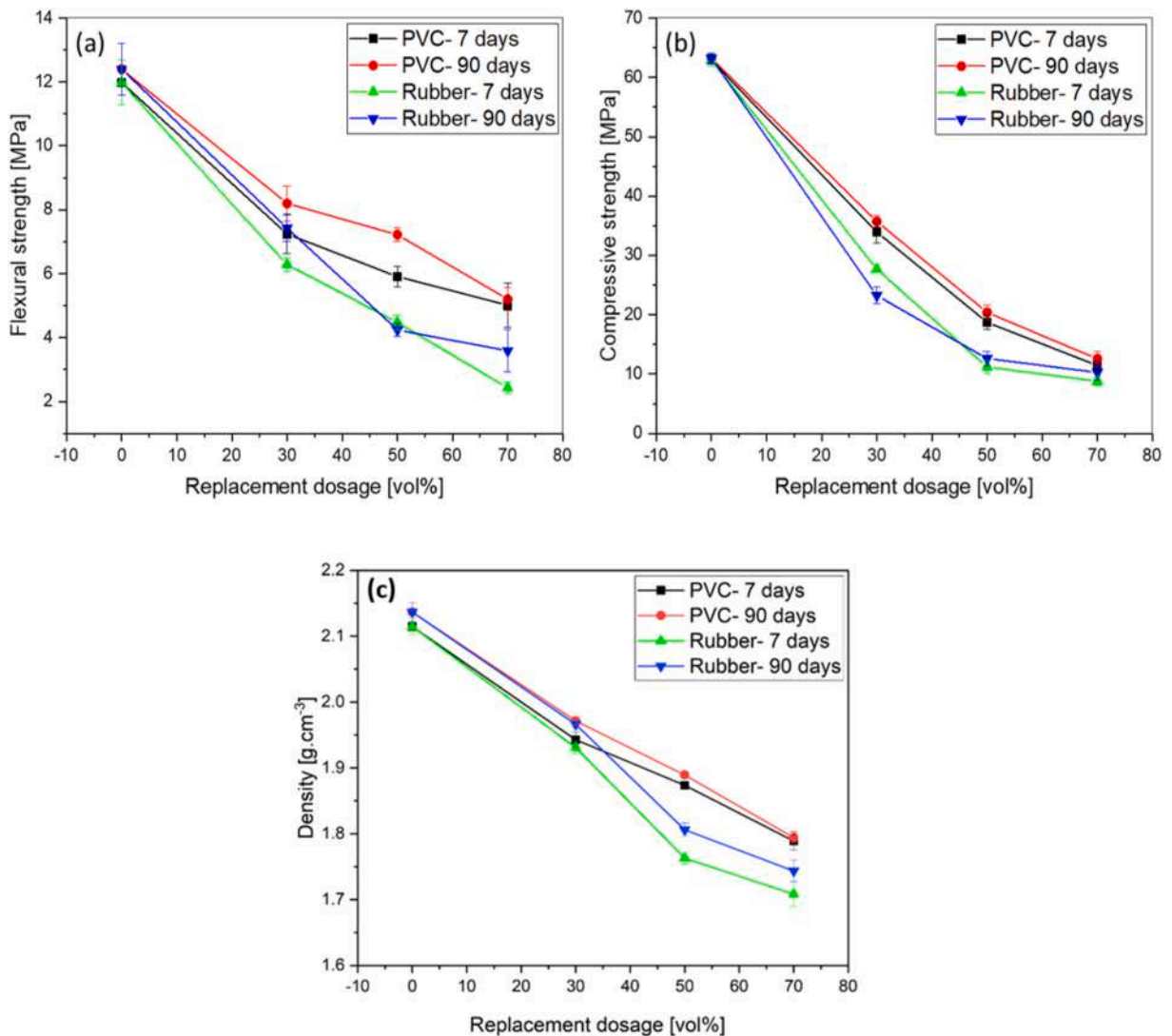


Fig. 3. a) Flexural and b) compressive strength and c) density at 7 and 90 days for AAMs with PVC and rubber aggregate replacements at different ratios.

aggregate matrix interfacial bonding [14,15].

The results show that replacing natural sand aggregates with PVC aggregates exhibited higher flexural and compressive strength values than their rubber counterparts in 7 d and 90 d of curing. It is worth mentioning that the curing age has a minor impact on the AAMs' mechanical properties. Fig. 3-a shows that at 7 and 90 days, the flexural strength of AAMs containing the maximum PVC replacement fraction (i.e., P70) was 5.6 MPa and 6 MPa, respectively, which is 20% and 30% higher than the values registered for R70 samples. Compressive strength results followed a similar trend (see Fig. 3-b), where the compressive strength of P70 was 40% and 30% higher than the values registered for R70 at 7d and 90d, respectively.

The difference in mechanical performance verifies the better compatibility of PVC aggregates within the AAM matrix due to the noticeable difference between the surface texture of PVC and rubber aggregates (see Fig. 2). The higher mechanical performance of AAMs containing PVC aggregates can be explained, as shown in Fig. 4, where PVC aggregates possess a rough rounded surface with grooves and notches that generate an intimate aggregate and matrix interface. As a result, the delamination phenomenon has not appeared in AAMs containing PVC aggregates (see Fig. 4a and b) compared to the AAMs containing rubber aggregates of smooth surface and tapered borders, as observed in Fig. 4 c-d. It is evidenced in the author's previous study and another research conducted by Li et al. (2019) [21,39], that the instinct alkaline surface treatment of rubber particles as a result of the existence of NaOH in the AAM matrix can enhance the compatibility of rubber-matrix, which is due to the surface impurity removal and increased surface roughness. However, more research is needed to determine the impact of potential alkali surface modifications on the PVC aggregates. It should be noted that the SEM was used solely for comparison purposes of the aggregate-matrix interfacial interaction. Due to the large dimensions of both rubber and PVC aggregate, no differences in the matrix morphology can be observed.

Several studies also reported that introducing rubber aggregates in AAM mixes reduced compressive and flexural strength due to rubber's hydrophobic character and its distinct nature from natural sand aggregates [40,41]. On the other hand, the addition of PVC aggregates to OPC mortar reduces its compressive and flexural strength [42]. As a result, sand replacement with PVC should not exceed 30% to reserve mortar's strength [43], which supports the findings of this study. The incorporation of PVC and rubber in AAMs

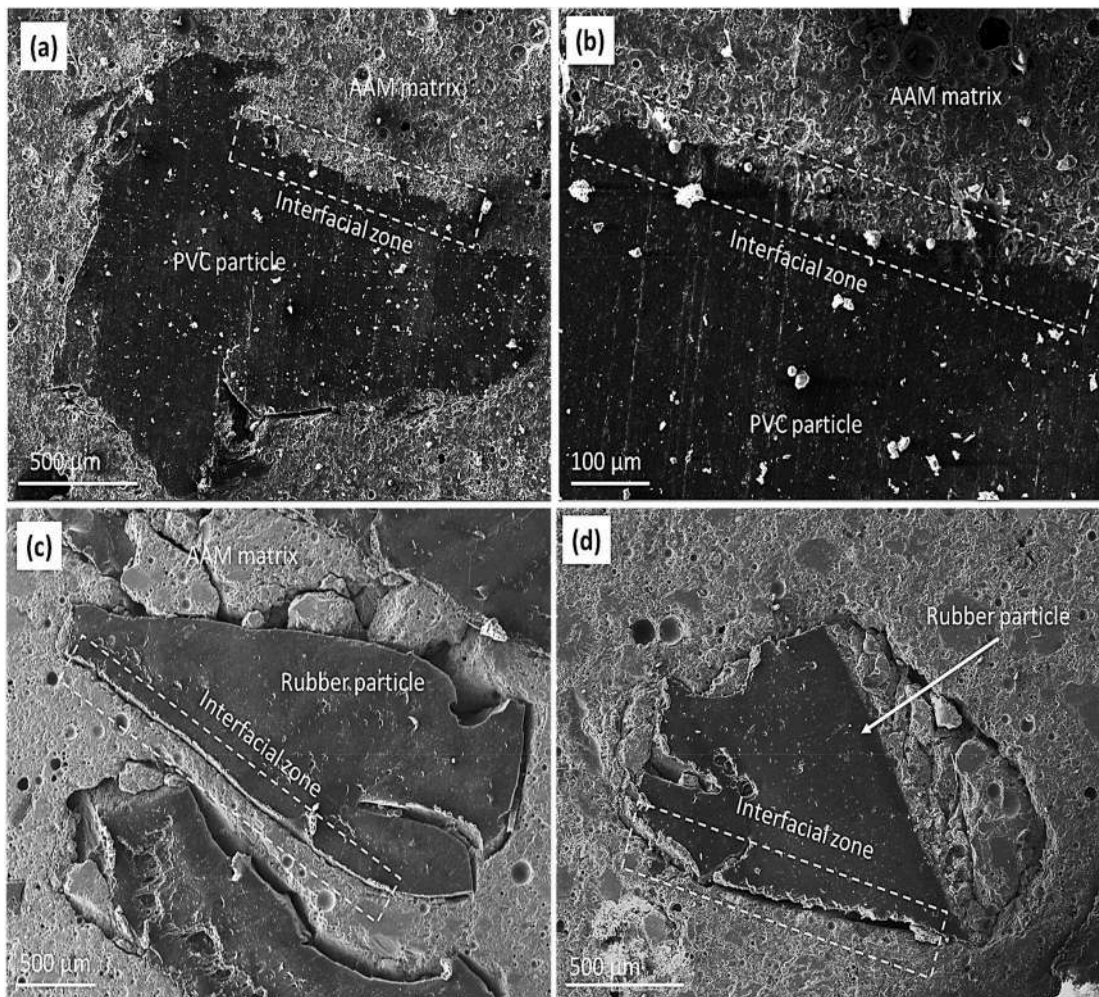


Fig. 4. SEM Images of AAM composites with PVC (a and b) and rubber aggregates (c and d).

composites has generally had a negative impact on their mechanical performance due to their hydrophobicity as well as different surface texture and shape compared to the natural sand. However, it is worth noting that composites with PVC aggregates achieved better mechanical properties than those with rubber aggregates at the same replacement ratios. Eventually, in this work, the main two factors that affected the mechanical performance of AMM composites are: (1) the type of aggregates (PVC or rubber) and (2) the amount of incorporated aggregates. These observations are in line with previous works [44,45]. Moreover, the addition of recycled rubber and PVC resulted in reduction in composites' density (see Fig. 3 c). This can be due to two reasons: (1) rubber and PVC have lower density than natural aggregates and (2) the weak interfacial bonding between the binder and the rubber and PVC aggregates creates air voids inside the matrix which reduces total mass of the composites.

3.2. Physical properties

3.2.1. Thermal conductivity

As seen in Fig. 5, it is apparent that incorporating rubber or PVC aggregates in the AAMs improved the thermal resistivity of the resulted composites. The thermal conductivity of AAMs progressively reduced from 1.485 w/mk for the control sample to 1.35 w/mk, 1.31 w/mk, and 1.23 w/mk for R30, R50, and R70, respectively. The thermal conductivity of samples containing PVC aggregates was lower or comparable to that of rubber counterparts. Among all compositions, 70P exhibited the lowest thermal conductivity (i.e., 1.192 W/m.k), which was 20% lower than the control sample. This reduction in thermal conductivity could be due to the lower thermal conductivity of polymer materials than natural sand aggregates, which contain a high percentage of conductive silicates [46]. The low thermal resistivity of the examined composites is likely due to the AAM composites being consolidated and intrinsically reduced porosity, resistant to chemical attack and heat stress [19]. Due to the PVC and rubber aggregates' lightweight, an inhomogeneous distribution of particles could occur inside the composite matrix [47]. This non-uniform distribution of aggregates affects the overall thermal resistivity of the composites. The composites' section with fewer polymers concentration has higher thermal conductivity than the rest of the composite. Belmokaddem et al. (2020) [48] reported that OPC composites incorporated with PVC aggregates at four different ratios, 0%, 25%, 50% and 75%, resulted in thermal conductivity values of 1.86, 1.26, 0.89 and 0.61 W/m.k, respectively, with about 70% thermal conductivity reduction at 75% PVC replacement. It is worth mentioning that the thermal conductivity of the AAMs control samples, i.e., AAM with 100% sand aggregate, in this study is 1.45 W/m.k which is 0.385 W/m.k lower than that of Belmokaddem et al. (2020) [48] OPC control samples, implying that AAMs composites have greater thermal resistance than OPC composites. Záleská et al. (2018) [49] reported a thermal conductivity reduction of about 80% when OPC-based composites were incorporated with different recycled plastic at 50% replacement with natural sand. The lowest composites' density was reported to be 895 kg/m³ at 50% recycled plastic replacement, while the density of the cementitious composite with 50% PVC replacement in this study was about 1900 kg/m³. This is an important indicator that the AAM-based composites have better matrix structure and interlocked particles with fewer voids, while OPC-based composites with less density could mean weaker interfacial bonding of the matrix structure and presence of more voids. Voids inside concrete play a major role in reducing heat transport inside the matrix [50]. In another study conducted by Záleská et al. (2019) [51], scraped rubber aggregates incorporated in OPC-based composites had similar behaviour to recycled plastics aggregates in reducing thermal conductivity by about 60% at 30% replacement of scraped rubber aggregates with natural sand. This value is much higher than the reduction observed in the composites in this study, i.e., about 7% reduction in thermal conductivity at the same replacement ratio. The higher reduction of the rubberised OPC-based composites could be due to the low density of about 1543 kg/m³, when compared to rubberised AAM-based composites in this study, which is about 2000 kg/m³. Low density reflects porous structure and more voids, therefore reduction in thermal conductivity [50].

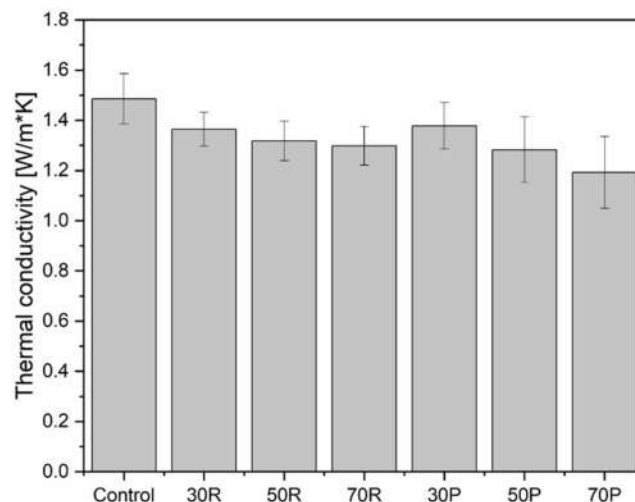


Fig. 5. Thermal conductivity of AAMs with PVC and rubber at different replacement ratios.

3.2.2. Water absorption

All the composites showed a clear upward trend in water absorption over time (see Fig. 6). The control sample had the highest water absorption rate, whereas the P70 sample showed the lowest (Fig. 6). After 72 h, the water absorption for the control sample was around 5.5%, while P70 was about 1.2%, resulting in a 78% water absorption decline.

R30 and R70 had almost the same values and demonstrated the second lowest water absorption, which dropped to around 3% after 72 h, resulting in a 45% decline compared to the control sample. The reduction in water absorption rate may come from their lower specific volume of rubber and PVC aggregates compared to natural aggregates, resulting in inhomogeneous rubber and PVC particle distribution in the composites structure [47]. Another reason could be that water absorption in the mortar was shown to be higher in lamellar and angulated plastic aggregates than in spherical and regular aggregates, presumably due to the weaker bond between aggregates and matrix (see Fig. 4). Li et al. (2020) and Teh et al. (2017) [52,53] reported comparable results for rubber in OPC mortar where water absorption decreases at a rubber content below 35% (size: 1.44 mm–2.83 mm) and increases somewhat above this level. Another study supporting the results of this research [42] found that the initial absorption of standard concrete was reduced by 40.5%, 39.6%, 45.8%, and 62.8% after the replacement of natural aggregates with 20%, 30%, 40%, and 50% RPVC (recycled PVC), respectively. Another recent study reported that as the amount of rubber in the mix increases, the amount of water absorbed drops. Rubber particles absorb less water than natural aggregates, resulting in a smaller effective area spanned by water [54]. Even though the integration of PVC and rubber aggregates creates voids within the AAMs composites, the hydrophobic properties of PVC and rubber work on reducing water absorption.

3.2.3. The volume of permeable void

The voids in porous materials are usually classified as open or closed porosity [28]. The open voids in cement-based materials are further separated into permeable, very narrow, and dead-end pores. The volume of permeable voids (VPV) in cementitious mortars is used to approximately predict the volume of capillary voids [28]. Fig. 7 shows that the VPV values for both AAMs containing rubber and PVC aggregates at all replacement ratios were lower than the control sample. The control sample had the highest VPV value, around 16%, indicating more voids and hence more water absorption. A comparable VPV values (i.e., around 12%) were registered for 30P and 30R, while PVC and rubber particles had the opposite behaviour inside the composites beyond this point. The VPV of AAM samples containing PVC aggregates increased gradually when the volume fractions of recycled aggregates were increased from 30% to 50% and 70%, reaching 13.7% and 13.8%, respectively. However, the opposite trend was registered for the samples containing rubber aggregates. The VPV of 50R and 70R decreased by 1.6% and 8.3%, respectively, compared to the 30R. The opposite trend in the VPV of AAMs containing PVC and rubber aggregates is likely due to the rubber's higher density than PVC. The relatively lower VPV of rubberised AAMs is due to the higher density of rubber particles than PVC (1.5 g/cm^3 for rubber versus 1.1 g/cm^3 for PVC), which indicates that at the same volume replacement ratios, more rubber aggregates are present in the mixture, resulting in more rubber particles forming blocked dense hydrophobic barriers prevent water uptake. To summarise our findings, plastic and rubber are hydrophobic materials [15] and therefore when these aggregates are introduced into the AAM composites, the water absorption was reduced.

Furthermore, Figs. 6 and 7 showed that the control sample had the highest water absorption and VPV rates compared to the PVC and rubber integrated composites, indicating that the hydrophobicity of PVC and rubber opposes water absorption and water capillary force. Moreover, the probable filling impact of smaller sized PVC and rubber aggregates at low volume seems to have a favourable effect on the void filling and reducing water absorption. However, increased aggregates contents tend to entrap more air, leaving a higher number of pores behind [37].

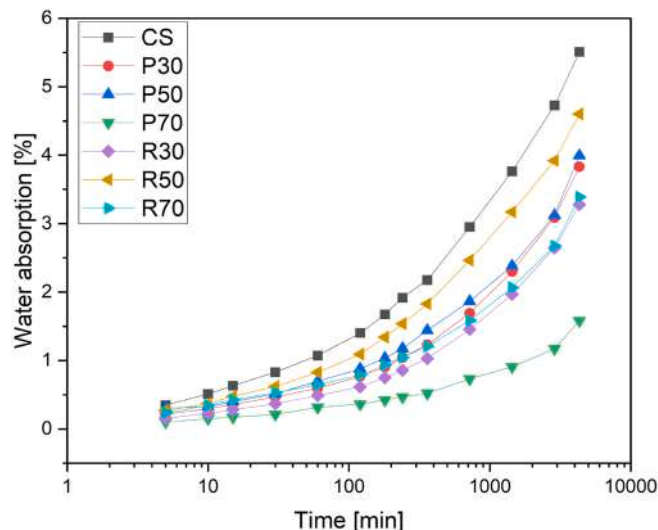


Fig. 6. Water absorption of AAMs with PVC and rubber at different replacement ratios.

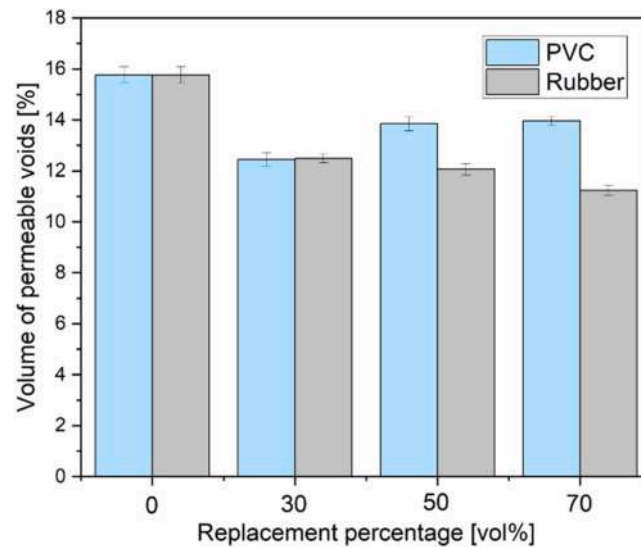


Fig. 7. Volume of permeable voids of AAMs with PVC and rubber at different replacement ratios.

3.2.4. Acoustic insulation

Massive materials represent a good acoustic solution in terms of insulation performance, ensuring effective sound barrier properties due to the high density and mechanical stiffness. Therefore, the control mix could be expected to provide significantly higher noise attenuation performance over the AAM mixes incorporating lightweight particles. SRI results in Fig. 8 show a completely different trend, highlighting comparable and, for certain replacement levels of rubber and PVC fraction, higher SRI values for the modified mixes than the reference sample. Polymer aggregates induce an additional acoustic dissipation mechanism, resulting from their viscoelastic nature and dissipative ability against vibro-acoustic phenomena [55]. Furthermore, the permeable porosity reduction following the addition of the polymeric particles (see Fig. 7) would ensure greater compaction of the AAM matrix and its sound-insulating behaviour, reducing the transmissibility of the acoustic waves through the material. In the two frequency ranges under examination, the different acoustic response was detected. In the low-middle frequency range (<500 Hz), the weakest insulating performance was noted, and the acoustic response of the investigated samples was almost identical, ranging from 2.60 dB in the 50P sample to 3.18 dB in the 70P sample. Limited by the mass law, lightweight materials have a fatal drawback: the low-frequency sound insulation performance is abysmal. To reduce the low frequency noise, a relatively thicker material is needed to realise an optimal reflection of low-frequency sound and breaking the mass [56]. At high frequencies, the effect of the polymer fraction on the acoustic resistivity of AAM samples was more evident. As a function of the type of polymer and polymer fraction, SRI values followed a poor clear trend. Regarding the influence of PVC aggregates, an ameliorative acoustic insulation effect occurred for the lowest sand-polymer

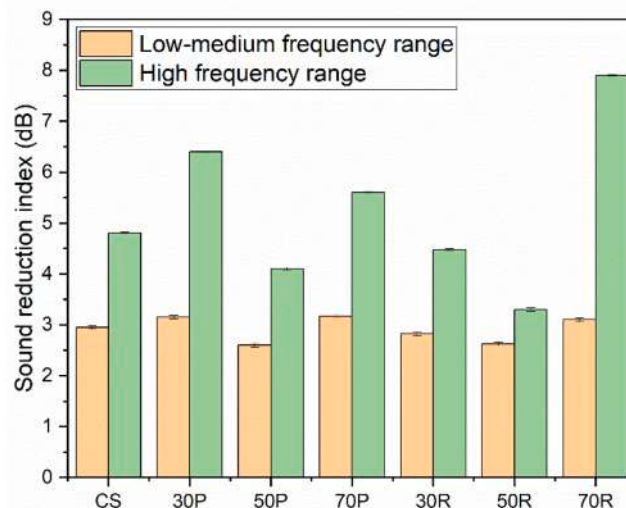


Fig. 8. Sound reduction index at different replacement.

replacement level. Indeed, the maximum increase in SRI (+33.3%) was found in the 30P mix compared to the control. As previously discussed, PVC content was lower than that of rubber fraction at the same sand-polymer substitution level. Therefore, it can be assumed that for low PVC dosage (30P mix), the insulation is still strongly influenced by the high stiffness and density of the AAM matrix, which adds the dissipative contribution of the polymeric part. For the rubberised formulations, the most effective contribution was verified for the highest polymer content. 70R sample showed better acoustic performance, providing an increment in SRI of about 64.5% with respect to control. Comparing this result with that of 70P mix, a clear difference in sound attenuation can be deduced: an increase of 41% in SRI-value was detected in the rubber-based formulation compared to the PVC-based counterpart. In addition to the discrepancy in the overall content of recycled aggregate functionalising the material, the effect of sound wave-particle interaction induced by the different macromolecular structures of the two polymers should also be considered [57]. When sound wave interacts with tyre rubber particles, the elastomeric chains are highly prone to deformation and displacement, as the rubber has better elasticity (Young's modulus of 2–10 MPa) and a glass transition temperature (T_g) below room temperature. Compared to stiff PVC particles (Young's modulus of 2700–3000 MPa; T_g of 80–90 °C), the elastic rubber more easily changes into new structural conformations when subjected to an acoustic wave, relieving the vibration energy, and increasing the acoustic damping, so to achieve better sound insulation performance.

3.2.5. Environmental concerns of PVC incorporation and chloride leaching evaluation

PVC is the third most extensively used synthetic plastic polymer after Polyethylene (PE) and Polypropylene (PP) [58]. When incinerated, PVC produces high amounts of chlorine due to the generation of chlorinated dioxins, as 57% of PVC's chemical constitution is chloride [5]. Moreover, when PVC waste is burned, a large amount of hydrochloric acid (HCl) is produced, resulting in the production of trace amounts of other dangerous gases such as organohalogen compounds, potentially causing pollution [59,60]. Moreover, contamination of waste plastic by PVC generates many problems, one of which is PVC dechlorination, which is a significant impediment to recycling. In the recycling process of PVC-containing polymers, dechlorination is an inescapable step [61]. Polyvinylchloride (PVC) is one of the most environmentally destructive polymers, yet it is also one of the most affordable and widely employed, but when it breaks down, it has a significant negative environmental impact. PVC releases phthalate plasticizers and chlorine-containing hydrocarbons (dioxins) during its deterioration in landfills or thermal treatment. Hydrochloric acid (HCl), tar, and a benzene-containing liquid fraction are among the thermal-degradation products. PVC deterioration is auto catalysed in the presence of HCl [48]. The authors believe that employing recycled PVC as aggregates in AAMs could be a promising way to avoid chlorine leaching into the environment and causing harm to the ecosystem. EDX was performed on the PVC/AAM interface in this study to validate the chloride leaching phenomenon. Due to the subjective nature of EDX analyses, different spots were randomly selected to verify the hypothesis. Fig. 9 illustrates the two spots located inside the PVC/AAM interface, one on the PVC aggregates and the other on the AAM paste. According to the EDX results, PVC particles can be safely integrated into the AAMs with minimal chlorine leaching beyond the matrix structure, preventing any toxic chlorine from leaching into the environment.

3.2.6. Carbon footprint estimation and life cycle assessment

Prior to presenting the ECO2 results of the AAM composites, it is important to compare the environmental and energy impacts of recycling tyre rubber and PVC by examining the flowcharts from LCA analysis (see Fig. 10). Regarding the PVC recycling process (see Fig. 10 a), 2.6 MJ of input energy for the grinding system were taken into account. This had an effect on the global warming potential (GWP) category and resulted in a cost of 0.23 kg of CO₂-equivalent. The Ecoinvent 9.3 databases' existing PVC manufacturing method using suspension polymerization served as the model's avoided product. In the form adopted for the study, this process is typical of all PVC manufacturing processes. This flow has a negative impact on the life cycle of AAM composites because it uses trash from a previous life cycle rather than a production method that has an adverse effect on the environment, which results in an impact of 1.89 kg of CO₂-equivalent. Overall, the production of PVC using the investigated process results in a reduction of 1.66 kg of CO₂-equivalent to the environment since the effect of power usage does not make up for the negative value of the avoided product. Similar considerations

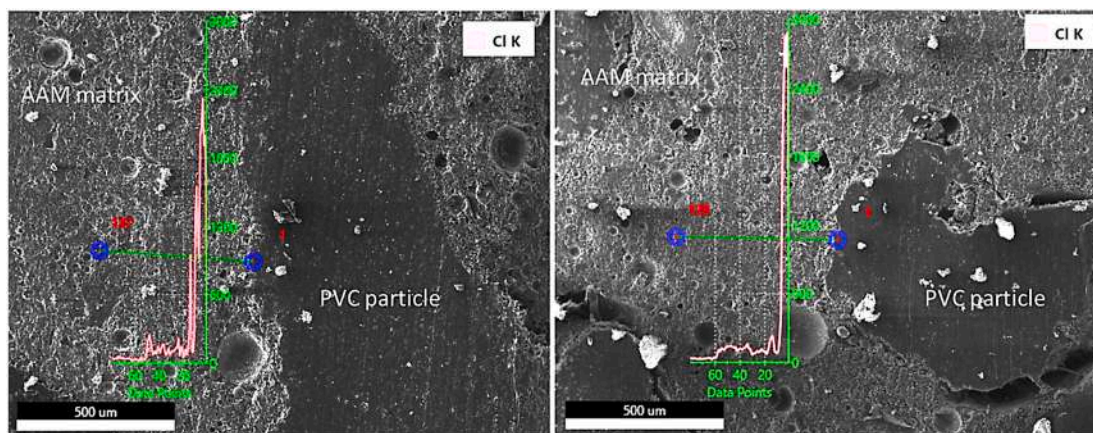


Fig. 9. EDX images of Chloride (Cl) detecting in the AAM composite.

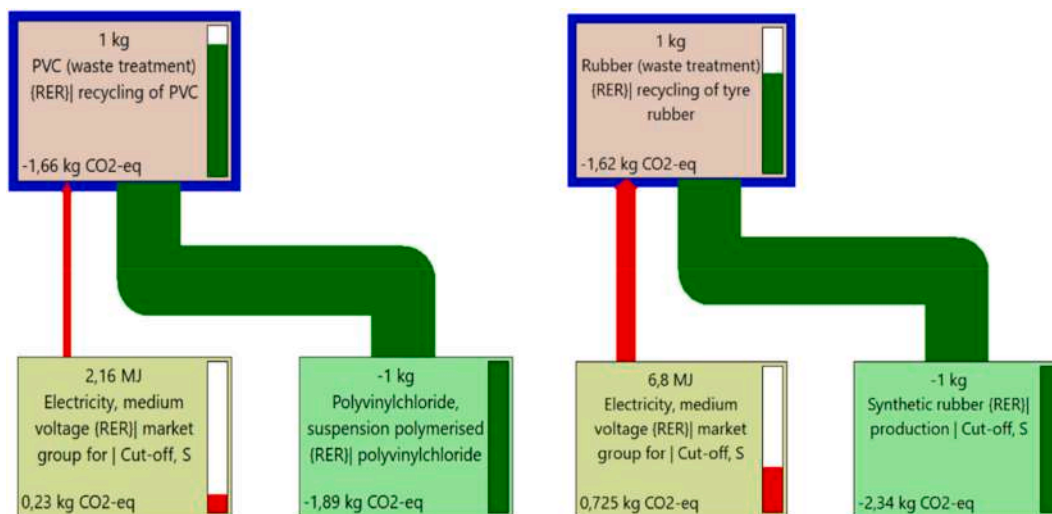


Fig. 10. LCA flows of recycling process: (a) PVC and (b) tyre rubber.

were made while recycling tyre rubber (see Fig. 10 b), which used 6.8 MJ of power and resulted in an impact of 0.725 kg of CO₂-equivalent. Synthetic rubber manufacture was regarded as an avoided product when it was included to the Ecoinvent database. In terms of GWP, the latter method results in a benefit with an impact of 2.34 kg of CO₂-equivalent. The recycling of tyre rubber has a negative environmental impact of 1.62 kg CO₂-equivalent. It is mentioned in relation to the PVC processing outcome that the energy flow and avoided product bring lesser impact values compared to those connected with the rubber recovery procedure. The CO₂-equivalent measurements were quite comparable and were used to quantify the environmental impact of the two polymer fractions in the production of AAM composites.

Fig. 11 shows a visualisation of the AAM composites' ECO₂ values. The carbon footprint was gradually decreased by using the two recycled parts in place of the natural aggregate. By using recycled PVC at 30 and 50% by volume, respectively, the decrease rates were 52 and 93% when compared to the Control mix (230 kgCO₂/ton). Reduction rates of 46 and 84% (for substitution levels of 30 and 50% by volume, respectively) were observed in relation to tyre rubber's effect. However, the highest amount of sand-recycled aggregate replacement yielded the most intriguing results (i.e., 70% by volume). A negative ECO₂ value was found regardless of the kind of polymer fraction employed as aggregate in the AAM mix design. According to this data, recovering waste materials from earlier processes (such as PVC from electrical cables and rubber from used tyres) and avoiding the manufacture of virgin raw materials (such as sand) results in high-value environmental benefits. Consequently, a reduced carbon emission is produced during both the creation of the composite material and the final disposal of the original waste [62]. Although an increase in recycled fraction incorporation results in a gain for the environment, it is important to carefully consider the loss of some engineering characteristics (primarily mechanical strength properties) to choose the best replacement level for scaling the final material in practical applications. Finally, there are two factors that contribute to the different effects of the two polymer aggregates on the overall carbon footprint of AAM: (1) a slight variation in the CO₂-equivalent index associated with the respective processing, and (2) the percentage of recycled fraction included in the design mix. At the same amount of replacement, the PVC content is larger than that of rubber since the two polymers have different densities. This results in greater environmental advantages.

The encouraging results of the current environmental impact analysis can serve as a standard for more thorough and comprehensive LCA analyses.

4. Conclusion

Using recycled rubber and PVC aggregates derived from end-of-life tyres and wire coatings in AAM is a solution that aligns with the construction industry's current eco-sustainable approaches. Therefore, this study aimed to verify the potential contribution of PVC and rubber waste as aggregates in low-carbon cementitious composites. The microstructure, mechanical performance, and physical properties were thoroughly investigated to draw the following conclusive statements regarding waste aggregates to replace virgin raw materials. The outcome of this study suggests that the specimens incorporated with PVC particles achieved better mechanical performance (both in flexural and compressive strengths) at all replacement ratios than their rubber-incorporated counterparts. PVC demonstrated better surface characteristics, achieving stronger bonding with the matrix paste. The highest compressive (i.e., 36 MPa) and flexural (i.e., 9 MPa) strengths values were registered for the 30P sample. Inclusion of both PVC and rubber aggregates in all replacement fractions improved the composites' thermal conductivity. The lowest thermal conductivities were 1.192 and 1.299 w/mk for 70P and 70R specimens, respectively. The integration of PVC and rubber aggregates with AAM materials decreased water absorption and the volume of permeable voids (VPV). After 72hr, the lowest water absorption recorded for 70P with a reduction of 72% compared to the control sample. The 70R mixture exhibited the lowest VPV value, 34% lower than the control sample. The addition of PVC and rubber aggregates positively influences the high-frequency acoustic insulation performance of the AAM matrices. Due to the

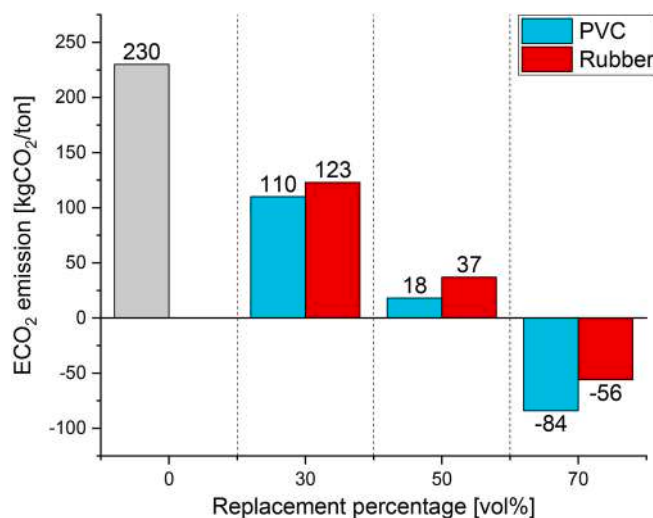


Fig. 11. Carbon footprint estimation in AAM composites incorporating PVC and tyre rubber aggregates.

dissipative effect of polymer aggregates, an improvement in sound reduction index ranging from 33.3% (for specimen 30P) to 64.5% (for specimen 70R) compared to the control sample can be found. This study also confirms that the PVC particles can be employed safely as aggregates in the AAM materials with no hazardous environmental issues concerning chloride leaching. In terms of environmental impact, by replacing sand with the polymeric fractions reduces the carbon footprint of AAM composites. For high levels of substitution (70% by volume), negative ECO₂ emissions were estimated, indicating how the recovery and reuse of a recycled fraction to replace a virgin one greatly strengthens the material's sustainability.

This research proved the promising potential of using recycled PVC and rubber aggregates as an eco-friendly alternative to natural aggregates to produce low-carbon cementitious composites with enhanced durability, thermal resistivity, and acoustic insulation. This study has highlighted that additional research is still needed to enhance the composites' mechanical characteristics to increase the replacement ratios of waste aggregates up to 100% replacement and widen their applications. Moreover, further investigation on the effects of highly alkaline environment of AAM matrix on the potential surface modification of PVC aggregates is also required.

Data support

The data that support the findings of this study are available from corresponding author Seyed Hamidreza Ghaffar, upon reasonable request.

CRediT authorship contribution statement

Eslam El-Seidy: Data curation, Methodology, Writing - review & editing.

Matteo Sambucci: Data curation, Methodology, Writing - review & editing.

Mehdi Chougan: Data curation, Methodology, Writing - review & editing.

Mazen J. Al-Kheetan: Writing - review & editing.

Ilario Biblioteca: Data curation, Methodology, Writing - review & editing.

Marco Valente: Writing - review & editing.

Seyed Hamidreza Ghaffar: Funding acquisition, Supervision, Conceptualization, Methodology, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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