



Research Article

High-pressure compacted recycled polymeric composite waste materials for marine applications

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Abstract

Abstract Options for recycling fiber composite polymer (FCP) materials are scarce, as these materials cannot be normally recycled and are toxic when improperly disposed. Additionally, reducing water usage is an increasing concern, as the concrete industry currently uses 10% of the world's industrial water. Therefore, building upon our previous work, this research explores the use of polymer hybrid carbon and glass composite waste products as reinforcements in high-pressure compacted cement. Our material used nearly 70% less water during manufacturing and exhibited improved durability and salt corrosion resistance. Compression strength tests were performed on high-pressure compacted materials containing 6.0 wt% recycled admixtures before and after saltwater aging, and the results showed that the material retained 90% of its original compression strength after aging, as it contained fewer pores and cavities. Our experimental work was supplemented by molecular dynamics. Simulations, which indicated that the synergetic effects of compaction and FCP admixture addition slowed the diffusion of corrosive salt ions by an average of 84%. Thus, our high-pressure compacted cement material may be suitable for extended use in marine environments, while also reducing the amount of commercial fiber composite polymer waste material that is sent to the landfill.

Article Highlights

- Fiber composite waste was successfully recycled into denser, high-pressure compacted ordinary Portland cement materials.
- High-pressure compacted cement samples containing 6% recycled admixtures retained 90% of their compression strength after salt aging.
- The high-pressure compaction method utilized 70% less water during specimen fabrication.

Keywords Recycled polymer waste · Cement · High pressure · Salt aging · Molecular dynamics

1 Introduction

Fiber composite polymers (FCPs) have many advantages for material applications, such as high durability, excellent strength to weight ratio, and extended service life, and are therefore frequently used by industry manufacturers. Worldwide mass production of polymer fiber composite

materials includes adhesives, plastic thermosets, and resins, however, most of these materials are unfortunately non-decomposable. Additionally, large amounts of composite waste are generated by the utility and engineering industries every year, most of which is sent to the landfill, and studies have shown that FCP material waste products will surpass 194 kilotons globally [1–5]. Unfortunately, with

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increasing production of FCP products containing carbon and glass fibers, the quantity of post-end of life materials being sent to landfills will only increase [1–5].

One source of FCP waste comes from power and electrical distribution utilities, which use composite conductor rods consisting of carbon and glass composite high-voltage conductor cores (HVCC) for their transmission lines. These HVCC rods contain hybridized carbon, glass fibers, and epoxy materials, which cannot be readily recycled. As a result, post-production waste, excessively damaged rods, and expired end-of-life HVCC materials are often discarded.

A major issue with these materials is the thermoset resin, which cannot be easily separated from the carbon and glass fibers. Recycling hybridized FCP is problematic because the methods used to separate the carbon or glass fibers and re-liquify the thermoplastic resin are limited to certain fibers, heat conditions, and mechanical constraints. Thus, large quantities of carbon and glass composites are not recycled because these procedures can mechanically degrade fiber quality. Fortunately, there are other ways to recycle FCP waste that have proven advantages such as physically or chemically recycling composite waste into existing materials such as cement and concrete structures, and these methods are more environmentally conscious than landfill disposal [3–5].

Presently, highly versatile corrosion-resistant cement matrix structures, such as pillar supports, wind energy structures, and coastal shore walls (barriers) are desired for marine water-based environments. A large majority of these industrial components frequently consist of cement-based concrete materials, which can withstand continuous impacts from ocean waves [6–10]. These structures are often based upon Portland cement, due to the material's chemical tolerance and high compressional high strength [6–9]. Unfortunately, even though Portland cement is tough and sturdy, it is not impenetrable to corrosive seawater environments, and must endure the destructive impacts of ocean water while being continuously exposed to salt-based corrosive compounds [11, 12]. Large concrete structures contain many open pores and internal gaps (spaces), anywhere between 10 and 20%, and these pores provide access points for water and saltwater, which can destroy the material over time [13–15]. As a result, a decline in mechanical strength often occurs within a few years due to surface degradation (pitting), development of large cracks, and internal expansions caused by water ingress [7–9, 16, 17], all of which physically compromise the overall structure [16–18].

The deterioration of various concrete structures can also be attributed to the fabrication and design quality of the component, which for cement-based materials is based upon the magnitude of surface and internal gaps or spaces [6–9, 13]. In some cases, severe deterioration due to seawater ingress may occur within a few years, resulting in surface defects and large cracks. These issues are

often combined with internal swelling caused by sulfate attack, chloride ingress, and freeze thaw expansion, which can lead to structural collapse or failure if not mitigated or prevented [6–14]. This phenomenon is due calcium hydroxide and aluminates hydrates reacting with sulfate ions, which can cause internal expansion due to the formation of calcium aluminate trisulfate, also known as ettringite. Deterioration from chemical sulfate expansion, ingress of chloride, and water expansion as a result of freezing can cause calcium leaching, which can further deteriorate and weaken the hardened cement paste [19]. Therefore, reducing the number of pores and voids in cement-based structures is crucial for structure longevity.

Previous studies have found that carbon and glass fibers in cement-based concrete can increase the qualities of the material, such as mechanical strength, life-span, and overall toughness [5, 20–23]. Thus, instead of being sent as waste to landfills, admixture FCP fillers may be used in high density cementitious materials, reducing the amount of water that needs to be used for mixing while also enhancing its pitch [5, 20–24]. Our previous work assessed the feasibility of introducing carbon fiber waste into standard ordinary Portland cement materials, and our results showed that certain FCP products improved the cement's resistance to salt aging, specifically due to the presence of the carbon and glass fiber material [5]. Therefore, to address the above-mentioned issues, we incorporated a high-pressure compaction machine to increase material density. Then, we assessed if high-pressure compacted materials containing reclaimed recycled HVCC materials can be used in highly corrosive environments, such as offshore and marine applications. Subsequently, our system utilized nearly 70% less water during fabrication, but created a denser material with fewer voids and cavities, which is critical for the concrete industry which currently uses 10% of the world's industrial fresh water [25].

2 Materials and methods

Cementitious materials can be improved through the addition of additives known as chemical admixtures, and these materials are specifically tailored to improve the mechanical performance of cement mixtures for certain construction applications. In this study, we combined Portland cement with various HVCC FCP admixtures, denoted in this study as recycle. We also used a high-pressure compaction system to decrease the porosity of the material, while also reducing the amount of water required during fabrication.

2.1 Ordinary portland cement (matrix)

Portland cement is composed of several main constituents, which are listed in Table 1. Cement is cured in several stages, regardless of whether it is commercially created in a factory and then transported to a construction site or mixed on-site and poured into molds or castings. While the hydration curing process results in a hardened paste cement within 24 h, the Portland cement will continue to cure long after its initial construction, even with non-hydrated compounds [16, 18, 25–27]. Not all types of cement are manufactured the same, and full hydration and maximum compression strengths may increase with age. Depending on setting (curing) environmental conditions, most nonblended cementitious based products in open air will reach maximum compression strength by 28 days, while for blended mixtures the maximum compression strength is achieved in 56 days. Therefore, it is standard practice to test strengths between 7 and 56 days after curing [16, 18, 25–27].

Several cement studies have noted that while water is a crucial ingredient in the initial mixing of cement, too much can hinder maximum compression strength, while too little can reduce workability and result in a weaker product [16, 18, 25, 27, 28]. Although water and ordinary Portland

cement are the primary ingredients, rocks (aggregate) are added to increase volume and lower cost, and when sand is added in addition to aggregate, it results in hydraulic concrete [16, 18, 25, 27, 28]. For basic construction purposes, standard high-strength concrete usually is ~60 to 80% coarse or fine aggregate [5, 18, 25, 27, 28]. Often, specialized additives are added to cement mixtures, such as silica-fume, fly-ash, small pebble (gravel), and larger diameter coarse or smooth rock aggregate.

Because the presence of open pores and internal gaps (spaces) can assist or accelerate moisture ingress and destroy the material over time, mitigating internal porosity and increasing the density of concrete products may be crucial for extending their service life in marine environments. With this in mind, we incorporated a high-pressure compaction system to reduce the porosity of the final ordinary Portland cement/HVCC powder composite material. Because it is vital to have a semi-liquid paste that can be easily poured and formed into molds, using the least amount of water possible, a compaction method created a final product with reduced pores or voids [16–18, 25, 27–29]. In this design, we followed the ASTM C150 standard and used commercial-grade Portland cement type I/II as the matrix with very low amounts of aluminates for moderate sulfate resistance, and recycled FCP materials were used as the admixtures [30].

Table 1 Composition of construction grade ordinary Portland cement type I/II by (wt%) [18]

| Constituent | Weight percentage (approx.) |
|--------------------------------|-----------------------------|
| CaO | 64.64 |
| SiO ₂ | 21.28 |
| Al ₂ O ₃ | 5.60 |
| Fe ₂ O ₃ | 3.36 |
| MgO | 2.06 |
| SO ₃ | 2.14 |
| Trace amounts (other) | <2.0 |

Common compounds noted by construction cement manufacturers

2.2 Background on the vibration dynamic compaction system

To create a dense cement product, a custom-designed high-pressure system was used (Fig. 1a). This vibratory dynamic compaction technology was adapted from the mining and tilling industry, and used twin 4000-Watt, 3-phase 50 Hz oscillating vibrators. This technology has been used to crush and compress geo-earth materials, as well as manufacture construction blocks and panels from coal fly ash, mining quarry tailing and rock, recycled concrete, granular industrial waste materials, and waste soils to form blocks, panels, and other geometries (Fig. 1b) [31–35].

Fig. 1 a, b. Dynamic high-pressure compaction system (a), high-pressure compacted Portland cement particle fiber mixed block (b)



This specific compaction system could compact Portland cement into tough durable blocks, making it ideal for creating large cementitious products [31–33]. In previous research conducted for the Department of Energy (DOE), this machine was successfully used to compact and stabilize contaminated soil, which allowed for safer storage, transportation, and disposal [31, 33].

The principle of dynamic vibration powder pressing relies on using repetitive pulverizing action. Because of the way the cement materials compress and cure, a lower kinetic energy pressure on the powder material will result in less compacted, higher void, and lower density material, while high-velocity impact compaction with an estimated peak impact force of ~2000 kN from the dynamic compaction system will result in a higher density surface to volume ratio. Therefore, when manufacturing a high-density material from a loosely packed powder, substantial density variations within the material's structure could be attributed to the high-velocity punching action of the upper anvil. To reduce this inhomogeneity problem within a compacted powder, oscillating vibratory forces from the lower steel table and mounted compaction mold were used to maximize the uniformity of material compression and the cementitious admixtures by ~40% by volume of the steel mold (Fig. 2b).

The main challenges of high-density compaction recycling of HVCC material are the mechanical material preparation and finding the optimal density to achieve high mechanical strengths and durability. The high-pressure compaction process ameliorated this by requiring less water than the traditional method for creating solid cement hardened products. While traditional concrete fabrication methods use around 33% water, only 10% water was used in this high-pressure compaction system (Table 2, Fig. 3). Thus, with the addition of microparticles from the Portland cement binder, and composite recycle particles averaging 52.85 μm in size, water usage

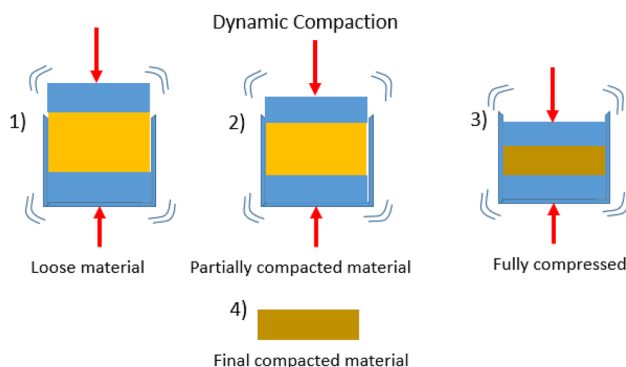


Fig. 2 Diagram of the dynamic high-pressure compaction system, showing a reduction of the cement by ~40% by volume in the fully pressed material

Table 2 Mixture proportions of cement powder (wt%)

| Compaction | Water ratio | Cement | Recyclate |
|------------|-------------|--------|-----------|
| HP | 10 | 100 | 6.0 |
| LP | 33.33 | 100 | 6.0 |

was reduced by 70% using the high-pressure compaction method, compared to the low-pressure, hand compaction method. Thus, if successful, this approach would help reduce overall water consumption in the cement industry [5].

2.2.1 Polymer composite fibers (admixture)

The HVCC polymer rod (also known as the conductor core) polymer matrix material used in this design was designed and manufactured by Composite Technology Corporation (CTC) Global. The composite matrix material was made of unidirectional hybrid carbon covered in a glass fiber shell. The matrix consisted of an epoxy resin, which is a high-heat resistant thermoplastic resistant to extreme temperature environments [5]. For the purposes of this study, the HVCC rod was cut and mechanically ground down to create a finer admixture pitch to include in the cement as the recyclate.

In this study, two recyclate HVCC admixtures were used: finely ground HVCC composite powder with an average particle size of ~52.85 μm , and crushed HVCC 'chips' which were ~8–10 mm in length and ~2–5 mm in width. The two admixtures are shown in Fig. 4a, which were made using the methods described in our previous work [5]. For comparison, virgin polyacrylonitrile (PAN) carbon fibers and glass fibers were also included in this study, to compare with the HVCC "recyclate". The PAN fibers were produced by Toray Industries, Inc., while the chopped E-glass fibers were attained from a commercial distributor. The FCP admixtures were also produced from newly manufactured HV conductor core rods.

In this work, 6.0 wt% of the "recyclate" admixture material was incorporated into the cement, and workability was evaluated by according to the ASTM standards [5, 30]. The recyclate admixture was combined with 10% water, after which the entire mixture was placed into a steel compaction mold (Fig. 4b), and then placed on the dynamic compaction base steel plate. The upper steel top plate with an anvil was then lowered to gently press the loose, moist cement powder mixture together (Fig. 5a). Dynamic compaction was then performed for 5–7 s, compressing the mixture into a dense solid.

A total of four different rectangular blocks were created using the dynamic compaction system, as shown in Fig. 6a: (1) 6.0 wt% virgin carbon/glass fiber + Portland cement,

Fig. 3 Comparison of total mass percentages of materials for mixing designs high-pressure vs. low-pressure compaction

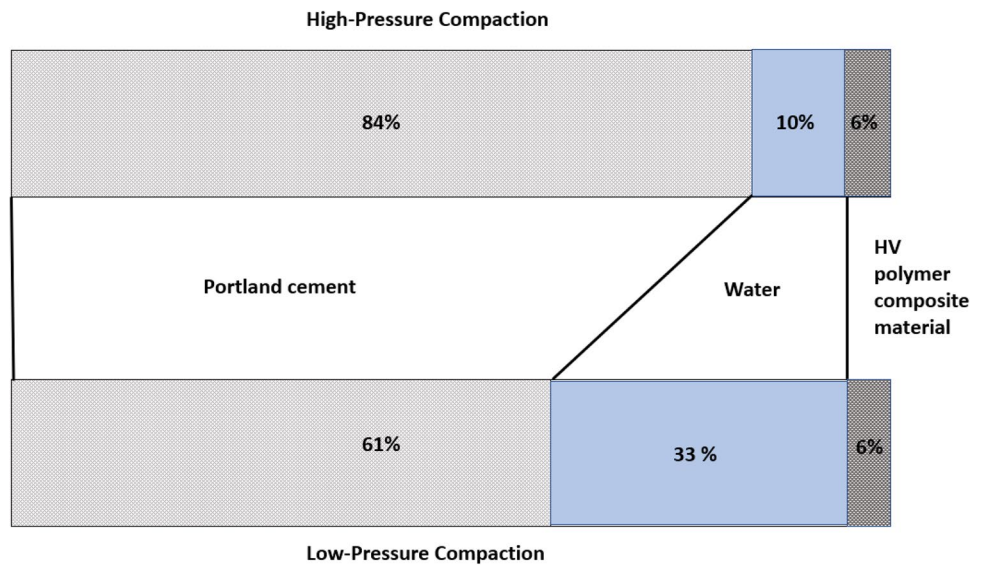
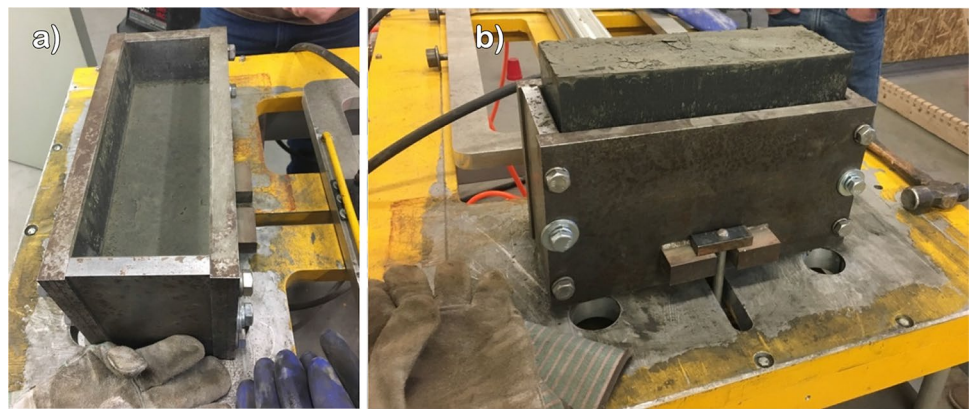


Fig. 4 **a** High voltage hybrid FCP admixtures that were processed into chips (middle) and a powder ($\sim 52.85 \mu\text{m}$) (far right), and then **b** mixed with Portland cement and water and high-pressure compacted into a solid rectangular brick-shaped material



Fig. 5 **a, b.** Steel high-pressure compaction rectangular mold used in the dynamic compaction system after compaction **a** and after extraction **b**

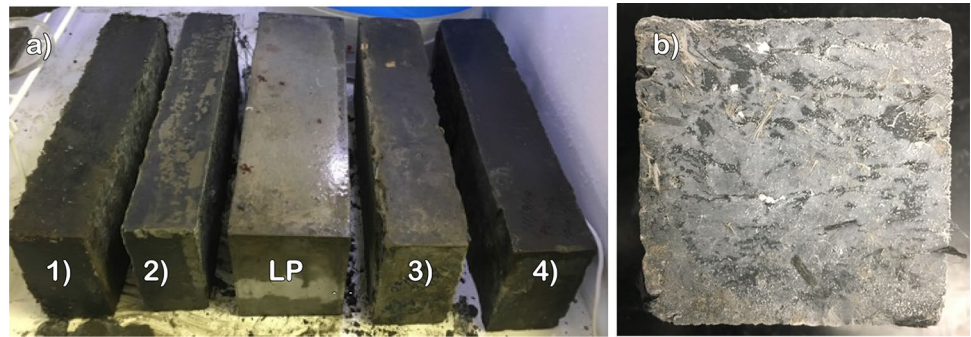


(2) neat Portland cement, (3) 6.0 wt% HVCC "chips" + Portland cement, (4) 6.0 wt% HVCC particle fiber powder (PFP) + Portland cement. One low pressure (LP) neat Portland cement sample was also included, for comparison. The low-pressure Portland cement specimens underwent standard hand compaction (tamping) with a rubber tool, using between 25 and 30 N of force to set the cement paste.

After compaction, the high-pressure compacted rectangular $31.5 \times 10 \times 6.8$ cm blocks were pre-cured for

24 h before being removed from the steel compaction mold. The four rectangular blocks were then submerged in tap water for 180 days at room temperature ($\sim 23^\circ\text{C}$), before being sliced into 5.0 cm^3 cubes (Fig. 6b), after which saltwater aging experiments were performed on individually prepared cubes. A total of 24 Portland cement cube specimens were thus created for a total of three samples per specimen type.

Fig. 6 a, b. High-pressure compacted rectangular blocks compressed at 2000 kN (1–4), along with regular low-pressure cement compressed at 25–30 N (LP, middle), before curing (a) and a sliced cube with 6.0 wt% of virgin carbon/glass fibers before compression testing (b)



2.2.2 Saltwater aging

Saltwater aging on 12 of the 24 cubic samples was conducted to observe the saltwater resistance of the selected cement admixtures. Concrete structures typically experience long-term exposure in seawater, as they are exposed to salt and the ocean moisture, which contain chlorine and other corrosive compounds [14, 19, 26, 36]. In real-world scenarios, mechanical wear and impact damage may occur by oceanwater wave impaction, or by the sand and gravel they carry, however below the surface, the seawater will ingress into the cement pores and surface voids [7, 8]. Thus, salt aging corrosion is more likely to degrade low-pressure cured cement with more voids than high-pressure compacted cement containing fewer voids [7, 8, 11–15, 37]. We used ImageJ to analyze the dry sample surface areas of the voids in the microscopy images, which were estimated to be spherical in shape. The chord length (diameter) was measured to fall between 50 and 700 μm , with noticeable exceptions, as some of the voids where > 1.0 mm. However, the voids in the high-pressure compacted cement decreased to ~ 7 –50 μm in size.

For the saltwater aging set, samples were situated in a 3.0% artificial aqueous solution that simulated ocean water at elevated temperatures of 83 $^{\circ}\text{C}$ for 30 days, to simulate accelerated aging in oceanic conditions. The aqueous seawater solution contained roughly 25 g/L of sodium, magnesium, and calcium chlorides. Other known constituents of < 1 g/L were also included in the artificial salt solution based on the ASTM D1141 standard [38], as noted in Table 3.

2.2.3 Compression strength testing

After manufacturing and curing of the cubic specimens for 180 days the high-pressure unaged and salt-water aged cement cubes containing either recycle, virgin fibers, or no recycle filler materials were strength tested using a high-strength compression frame system. A total of 12 cubic unaged samples were tested for compression strength, and 12 cubic specimens were exposed to

Table 3 Manufacturer specifications for simulated sea salt

| | | |
|--------------------|---------------------------------|-----------------|
| Sodium Chloride | NaCl | 24.53 g/L |
| Magnesium Chloride | MgCl ₂ | 5.20 g/L |
| Sodium Sulfate | Na ₂ SO ₄ | 4.09 g/L |
| Calcium Chloride | CaCl ₂ | 1.16 g/L |
| Potassium Chloride | KCl | 0.695 g/L |
| Sodium Bicarbonate | NaHCO ₃ | 0.201 g/L |
| Potassium Bromide | KBr | 0.101 g/L |
| Boric Acid | H ₃ BO ₃ | 0.027 g/L |
| Strontium Chloride | SrCl ₂ | 0.0025 g/L |
| Sodium Fluoride | NaF | 0.003 g/L |
| Water | H ₂ O | 988.968 g/L |
| | | Total: 1025 g/L |

saltwater conditions and then tested for compression as aged specimens. Compression testing was conducted on select cubes in accordance with the ASTM C109/C109M standard [30].

3 Experimental results and discussion

The maximum compression strengths of the unaged samples are shown in Fig. 7. All the high-pressure samples cured in water for 180 days had maximum compression strengths between 30 and 60 MPa (Fig. 7). These relatively high compression strength results were likely due to the decreased porosity and lower permeability of the high-pressure Portland cement, compared to low-pressure self-compacted cement. In cement, most surface/internal voids and pores are the result of insufficient compaction, mixing, and placement of the created cement paste during curing, and as a result low-pressure compacted cements will have comparably more voids in their structures (Fig. 8a) [14, 25, 27, 39]. Surface magnification of the high-pressure compact cement indicated fewer, and smaller voids after compaction compared to low-pressure compacted cement, however the high-pressure compacted cement sample did

Fig. 7 Compression strengths of the high-pressure compacted unaged cement samples with and without recycled admixtures cured for 180 days

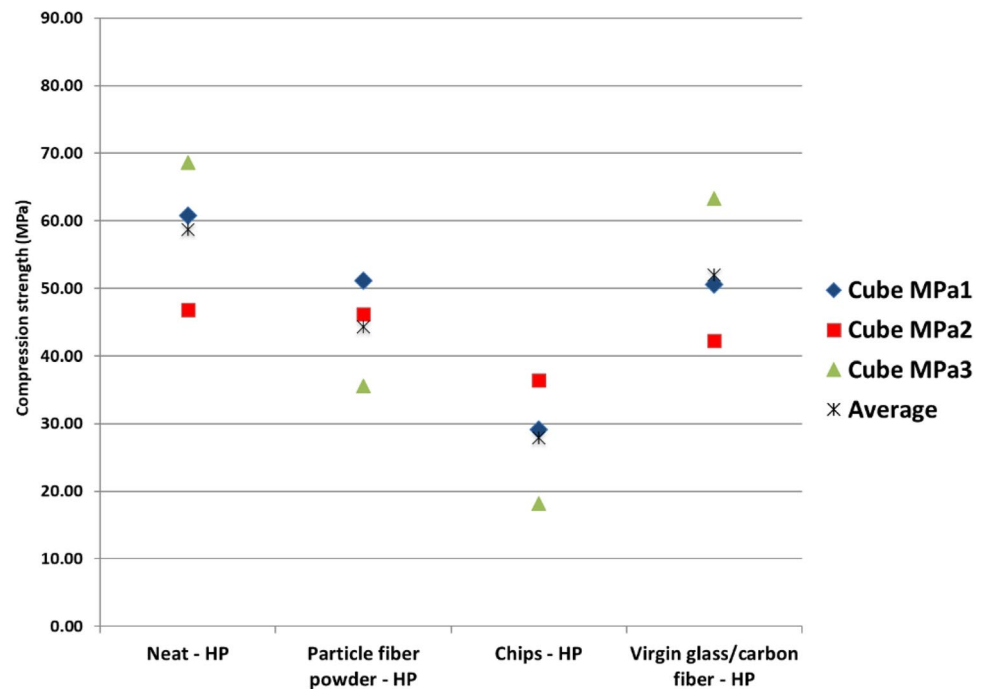
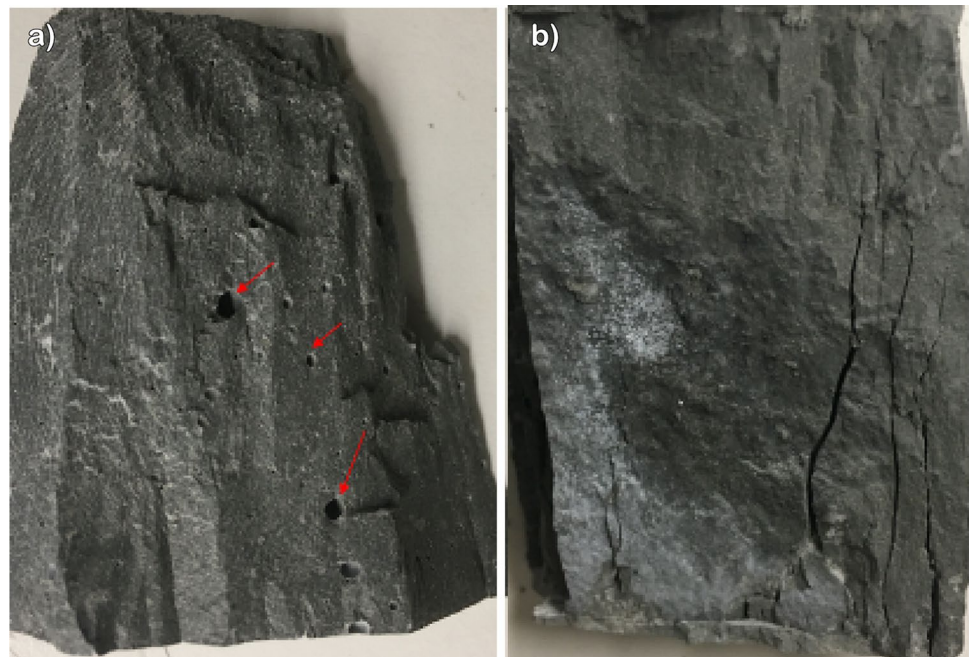


Fig. 8 a, b. Post compression test analysis of the fractured prism specimens: low-pressure compacted cement with noticeably more internal voids compared to the high-pressure compacted cement **b**, which also contained fracture cracks as a result of compression testing



contain post-compression fracture stress cracks (Figs. 8a, b and 9a, b).

Surface microscopy analysis of the neat low-pressure compacted cement, neat high-pressure compacted cement, and high-pressure compacted cement samples containing the recycle showed notable differences in surface characteristics. Numerous microscopic voids and cavities were visible in the neat low-pressure Portland

cement samples (Fig. 10a) but absent in the high-pressure compacted sample (Fig. 10b). In the high-pressure compacted sample containing 6.0 wt% of HVCC particle fiber powder (PFP), individual fibers were visible as well as darker areas where the powder material was incorporated into the high-pressure compacted cement (yellow arrows), and overall fewer voids and cavities were observed compared to the low-pressure compacted cement.

Fig. 9 a, b. 70× surface magnification of low-pressure compacted ordinary Portland cement a), and high-pressure Portland cement with 6.0 wt% of particle high-voltage conductor material b)

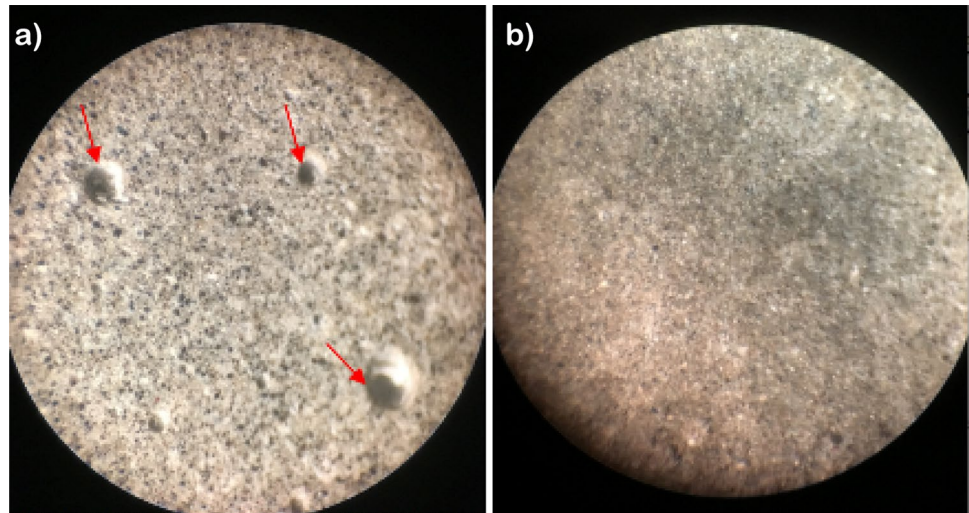
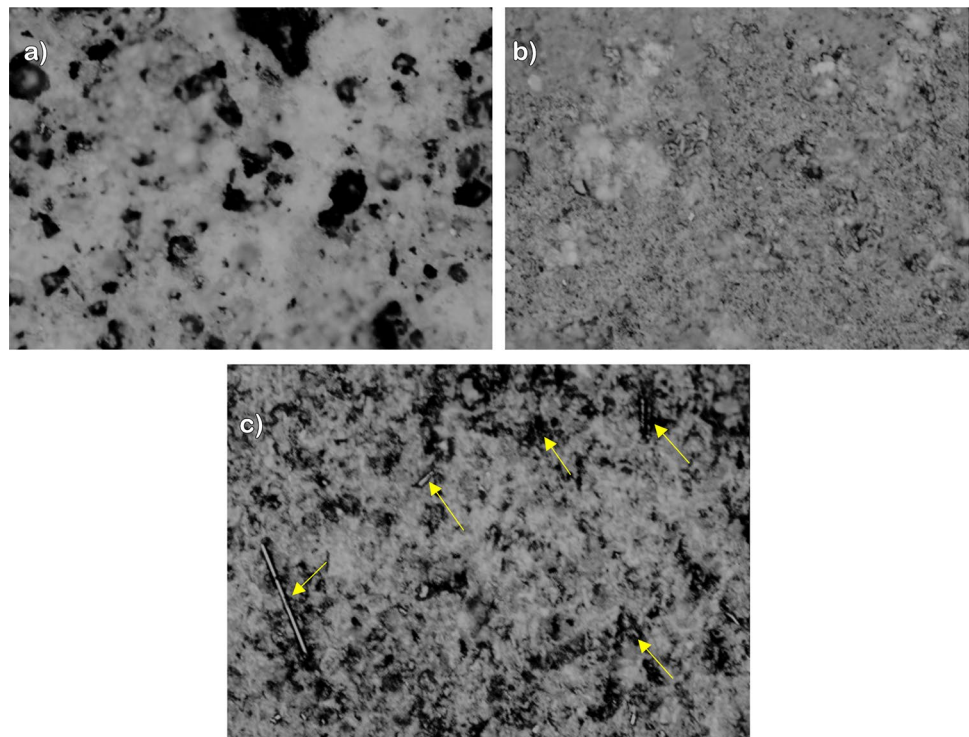


Fig. 10 a–c. 200× magnification of low pressure compacted Portland cement (a), high-pressure compacted Portland cement (b), and high-pressure compacted cement with 6.0 wt% of PFP (c)



The salt-aged samples also showed marked differences in visual appearance, and visual observations of the high-pressure compacted samples indicated visible surface efflorescence from salt-aging, as shown in Fig. 11a–e. This surface effect was similar to the surface efflorescence observed on the low-pressure compacted cement in our previous study. Large cracks and efflorescence were also noticeable in the high-pressure compacted 6.0 wt% HV chips sample (Fig. 11c).

The compression test results of the salt aged samples showed that high-pressure compressed materials containing the recycle admixtures had average compression

strengths that were comparable to standard hydraulic concrete (Figs. 8, 12). The compression strength of the neat high-pressure compacted cement, which was water cured for 180 days, was roughly 10% of its average compression strength value after saltwater aging for 30 days. However, the cement specimens with the particle fiber powder admixture lost only ~7% average maximum compression strength after saltwater aging, for a final value of 41 MPa (Fig. 12). This result was comparable to the averaged maximum compression strength of the unaged, non-saltwater treated material, which was 44 MPa. However, compression strength values of the neat high-pressure

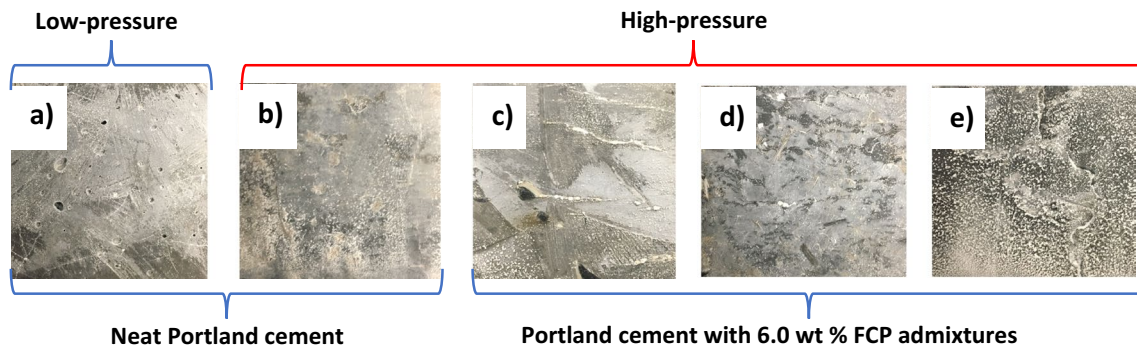
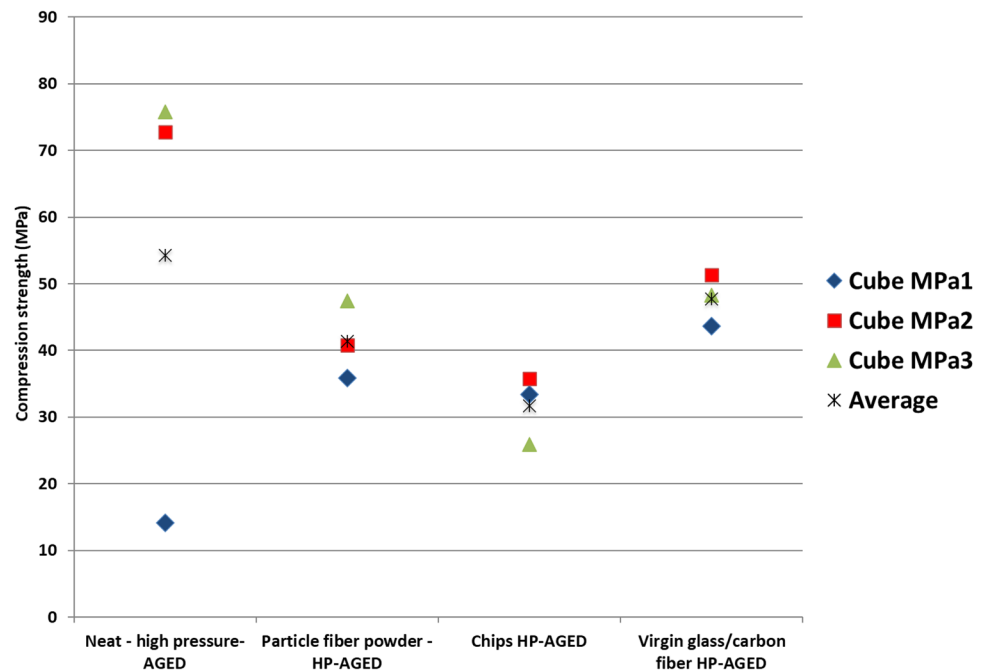


Fig. 11 a–e. Cement mixtures of low-pressure and high-pressure compacted fiber mixtures

Fig. 12 Compression strengths of the high-pressure compacted cement with and without PFP admixtures after salt-aging for 30 days



compacted exhibited greater variations in strength, likely due to embrittlement after saltwater aging. The neat high-pressure compacted had the lowest single performing test value, but the highest overall compression strength. The combined virgin carbon and glass fiber admixture also lost very little compression strength, from 51 to 48 MPa, and the small decrease in strength was likely due to the non-pozzolanic effects of the carbon fiber particles. Although the newly manufactured (virgin) carbon/glass fiber admixture exhibited slightly average maximum compression strength after aging, they were not a recycled waste composite material, thus a slightly higher average compression strength was expected. Furthermore, although the recycled composite core chip admixture samples showed a notable increase in compression strength after aging, they exhibited the lowest performance, with an average compression strength decrease of 27%. Therefore, from

these mechanical compression testing results, we concluded that the addition of recycled particle fiber powder admixture noticeably mitigated the deleterious effects of saltwater aging such as salt (magnesium and sodium) sulfate/chloride attacks, allowing it to maintain its compression strength and effectively making the material more resistant to saltwater corrosion. This effect will be further examined in Sect. 4 with the assistance of molecular dynamics.

4 Molecular dynamics

Moisture ingress is a serious concern in cement-based materials, especially in saltwater environments, as moisture can carry damaging particles such as magnesium, sodium, and chlorine compounds, into the interior of the

cement. Thus, a material that can limit moisture diffusion may consequently delay aging. To better recognize and evaluate the experimental results, a molecular dynamics model was created based on our previous research to illustrate ion diffusion through high-pressure cement [5]. This analysis was conducted by using Mean Squared Displacement (MSD) to determine molecular interactions with Portland cement, and HVCC particle fiber powder materials, under simulated low and high-pressure compaction conditions [5, 40, 41].

4.1 Molecular dynamics methods

Simulated nanoscale-sized cement particles and fiber polymer blocks were created using Materials Studio's molecular dynamics software, using the same methods used in our previous work, with the addition of small fiber fragments (carbon and glass). These glass and carbon fragments were constructed as pre-constructed compounds using the Nanostructure module in Materials Studio [5]. To simulate the resin used in the experimental study, manually constructed epoxy fragments were also included in the form of bisphenol A (BPA), as these compounds are prevalent in many epoxies.

A total of three nanoscale Portland cement models were created: low-pressure standard density Portland cement (PCLP) as a baseline comparison, high-pressure high-density Portland cement (PCHP), and high-pressure high-density Portland cement containing the FCP admixture (PCpfHP). Finite-sized nanoscale molecular compounds of SiO_2 , Fe_2O_3 , Al_2O_3 , and CaO , which make up a large portion of Portland cement, were also included in each model. The structures were placed in an Amorphous Cell lattice box ($7.8 \times 7.8 \times 7.8$ nm), at a weight percentage consistent to ordinary Portland cement (Table 1). The third high-pressure model contained nanoscale carbon

and glass fibers, and epoxy (BPA) compounds, to simulate the experimental samples created in this study containing recycled admixtures [5, 40–42]. Each individual lattice box contained either seven Na^+ , Mg^{2+} , or Cl^- ions, to simulate the ingress of sodium chloride and magnesium chloride, as these compounds are present in seawater and are known to cause deleterious effects to cement-based materials in marine environments. The movements of these compounds through the lattices were assessed using MSD.

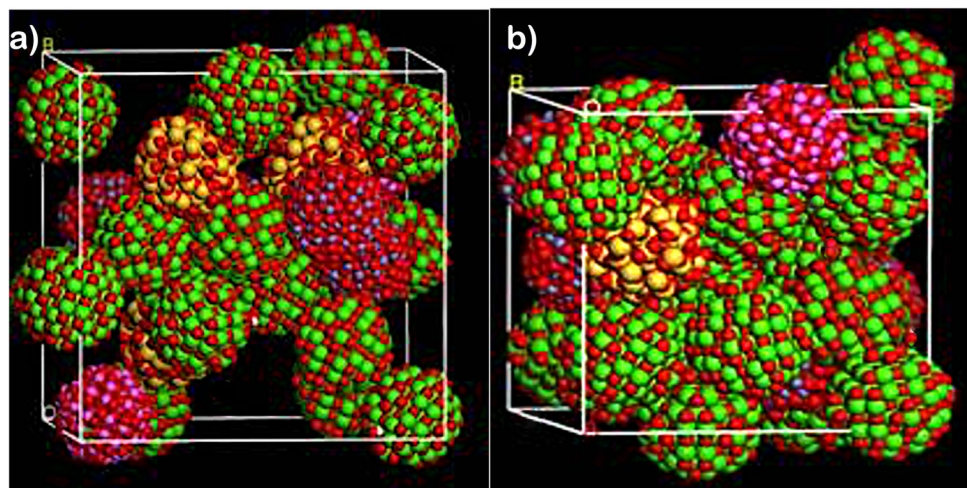
After creating the initial models using Amorphous cell, the two-high pressure/compacted models were 'compacted' using molecular dynamics NPT—the persistent or 'constant' number (N) of molecules, pressure (P), and the temperature (T)—for 20 picoseconds at 3.0 GPa of pressure to reduce the cell density to approximately 3.4 g/cm³. [5] The temperature, volume, and number of molecules were held constant during NVT Forcite dynamics, after which analysis of corrosive ion diffusion through the cement and FCP materials was conducted using MSD (Eq. 1) [5]. The three molecular dynamics models (PCLP, PCHP, and PCpfHP) are shown in Figs. 13a, b and 14b, and the overall molecular dynamics build process is shown in Fig. 15.

After initial molecular dynamics, the MSD analysis tool was used to assess how quickly the ions moved throughout each system. This tool determined the number of molecules (Eq. 1) in each system, represented by N for the diffusivity coefficient (D) in a group of fixed atoms in calculating MSD [33, 40, 42].

$$D = \frac{1}{6N_a} \lim_{t \rightarrow \infty} \frac{d}{dt} \sum_{i=1}^{N_a} [r_i(t) - r_i(0)]^2 \quad (1)$$

Using Materials Studio, we utilized the MSD method to evaluate the molecular interactions of the Cl^- Na^+ ions as they collided with one another and observed their

Fig. 13 a, b Molecular dynamics models of low pressure (PCLP), and high-pressure compacted Portland cement with particle fiber material (PCpfHP)



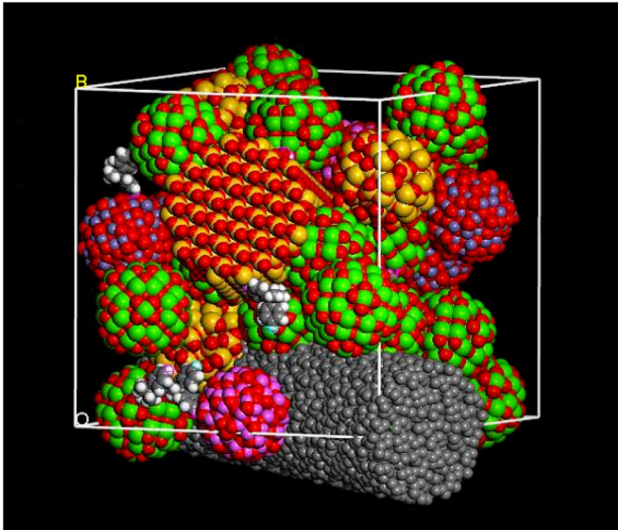


Fig. 14 Molecular dynamics model of high-pressure compacted Portland cement with particle fiber material (PCpfHP)

movements through the accessible or presented volume [33]. In MSD, even though there is a non-directional motion, a molecule will not maintain its proximity to its original location. Therefore, molecules will often meander or ‘wander’ at a certain rate, as determined by their charge, the density of the material, and the relative temperature during the process of diffusion [32, 33]. Thus, from this process, the rate of diffusion could be estimated by the speed of molecular movement.

4.2 Molecular dynamics results and Discussion

The diffusion rates of the Cl^- , Mg^{2+} , and Na^+ ions within the low and high-density cement models are illustrated in Figs. 16, 18, as a function of MSD over 10 ps. The results showed that the average MSD Cl^- , Mg^{2+} , and Na^+ ion diffusion rates in the low-pressure Portland cement (containing no composite admixture components) were significantly higher after 10 ps, compared to the high-pressure PCHP and PCpfHP models. This indicated that Cl^- , Mg^{2+} , and Na^+ ions would be more likely to diffuse, and would diffuse faster, through standard low-pressure cement material. By contrast, the high-pressure compacted models showed lower overall MSD rates for the Cl^- , Mg^{2+} , and Na^+ ions. Notably, high pressure compaction alone did not reduce diffusion nearly as much as the simulated compacted model containing admixture components. Thus, while the high-pressure simulations had lower rates of Cl^- , Mg^{2+} , and Na^+ ion diffusion, the high-pressure models containing carbon and glass fibers with epoxy components had the lowest diffusion rates, with final MSD values of < 50 for the PCpfHP models. As a result, Cl^- ion diffusion in the

PCpfHP model decreased by $\sim 88\%$ compared to the PCLP model, Mg^{2+} ion diffusion decreased by $\sim 85\%$, and Na^+ ion diffusion decreased by $\sim 80\%$ in the PCpfHP model, compared to the PCLP model. Therefore, in all simulations, the Cl^- , Mg^{2+} , and Na^+ ions diffused through the higher density Portland cement materials containing admixtures at slower rates, with an average of 84% decrease (Fig. 16, 17, 18).

Because damage to Portland cement structures can be exacerbated by moisture (water) ingress, especially in high chloride solutions such as marine environments, lower rates of Na^+ , Mg^{2+} , and Cl^- (MgCl and NaCl) species diffusion are preferable. Notably, a comparison of Mg^{2+} and Na^+ diffusion rates showed that while Mg^{2+} diffused $\sim 12\%$ faster than Na^+ in the PCLP model, the diffusion of Mg^{2+} was slower than Na^+ in the PCHP and PCpfHP models, by $\sim 11\%$ and $\sim 14\%$. Therefore, these results indicated that the combined effects of high pressure and the fiber and resin components helped to slow the diffusion of Mg^{2+} ions. This is significant, as MgCl is known to have deleterious effects on cement and concrete materials, even compared to NaCl , in marine environments.

Thus, our molecular dynamics results indicated that the presence of fiber and resin components, combined with a denser overall material, endowed the material with fewer migratory pathways for the corroding ionic compounds, effectively delaying ionic migration. While Portland cement is a typically porous that typically assists in ion mobility, the molecular dynamics results verified that high pressure compaction could significantly delay salt aging, and supported the compression strength results obtained in this work, showing that the high-pressure compacted materials with admixtures retained 90% of their strength. Of note, while the molecular dynamics results showed that corrosive ion diffusion was delayed, standard water diffusion may also be affected, which is required for curing. Therefore, curing times for high-pressure compacted samples in real-world circumstances may need to be extended, as standard moisture ingress would require more time to travel to the interior of the material and initiate the heat of hydration reaction. As a result, this may be a limitation of this material in some applications, and this topic will be investigated in our future work.

5 Conclusions

Of the various FCP admixtures and Portland cement mixture combinations evaluated in this study, the 6.0 wt% high-pressure compacted ground particle fiber powder sample retained 90% of its average maximum compression strength after aging in saltwater for 30 days, compared to the neat ordinary Portland cement on average.

Fig. 15 Build process for the molecular dynamics models, which examined Na^+ , Mg^{2+} , Cl^- ion movement through high and low pressure compacted nanoscale Portland cement with and without FCP admixtures

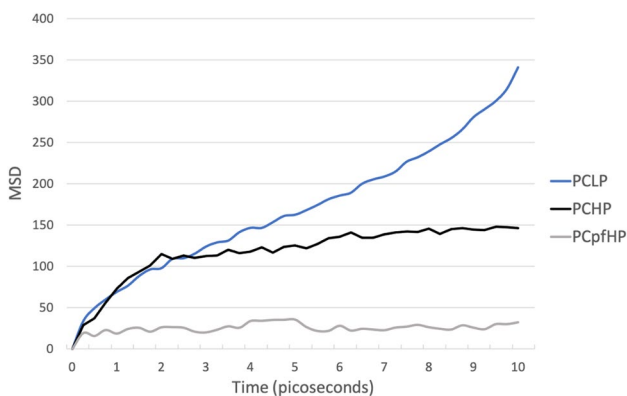
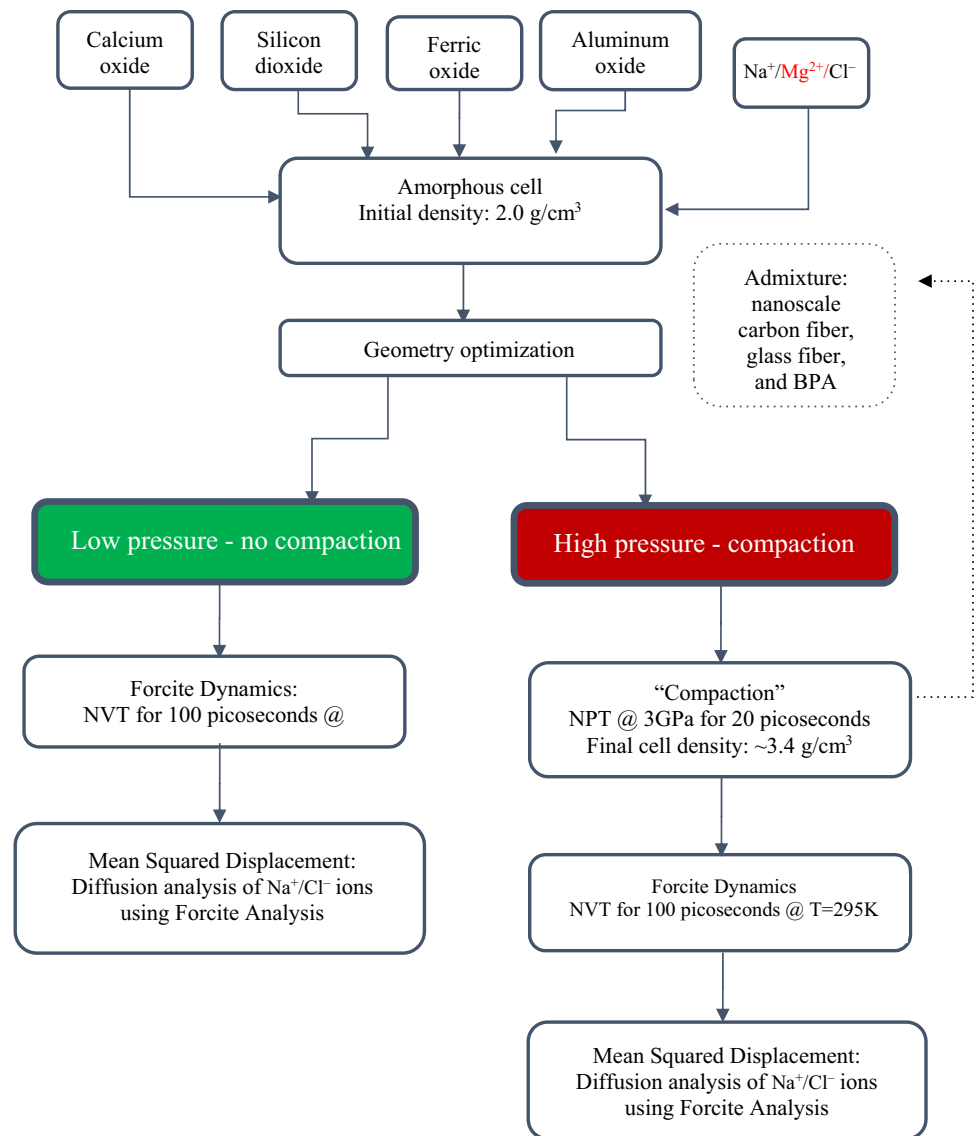


Fig. 16 Diffusion of Cl^- ions through low-pressure Portland cement (PCLP) in blue, high-pressure Portland cement (PCHP) in grey, and Portland cement with particle hybrid glass and carbon fiber FCP material (PCpfHP) in black

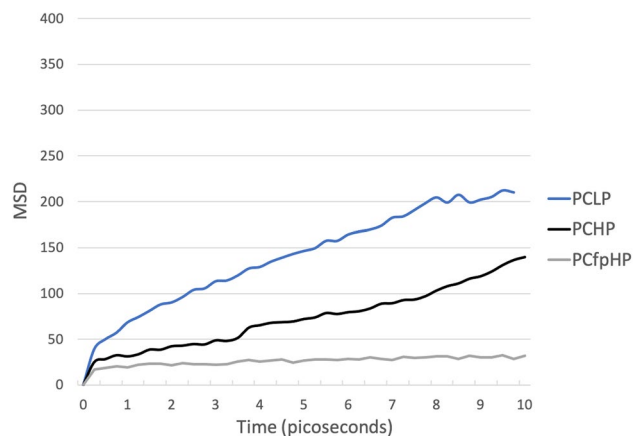


Fig. 17 Diffusion of Na^+ ions through low-pressure Portland cement (PCLP) in blue, high-pressure Portland cement (PCHP) in black, and Portland cement with particle hybrid glass and carbon fiber FCP material (PCpfHP) in grey

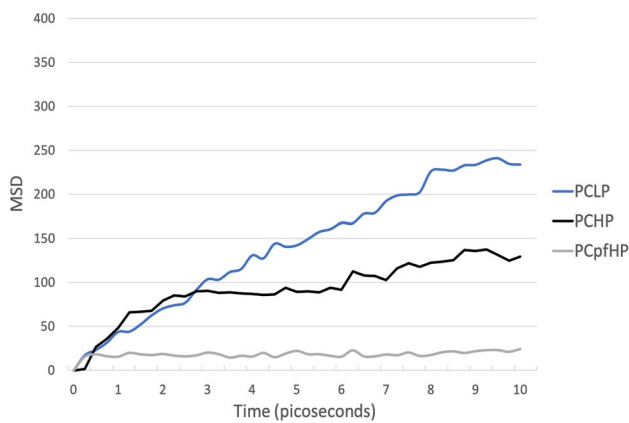


Fig. 18 Diffusion of Mg^{2+} ions through low-pressure Portland cement (PCLP) in blue, high-pressure Portland cement (PCHP) in black, and Portland cement with particle hybrid glass and carbon fiber FCP material (PCpfHP) in grey

Although the average compression strength of the neat Portland cement and virgin glass fiber admixture samples were higher, they experienced more deleterious effects on compression strength compared to the particle fiber powder sample. Therefore, specifically mixing the ground fiber composite waste material into cement could be used to retard water ingress and delay the diffusion of reactive ions in highly compacted hydraulic Portland cement.

The averaged maximum compression strength retention of the high-pressure compacted cement, specifically the heated saltwater aged particle fiber powder recycle admixture, supported the possibility of recycling powder FCP waste materials into cement-based materials structures for highly saline, corrosive submersion environments. This study showed that finely ground, recycled FCP waste can be used in high-pressure compacted Portland cement to improve density and reduce porosity, creating a material that can substantially mitigate diffusion of deleterious compounds in saltwater (Cl^- , Mg^{2+} , and Na^+ ions) by an average of 84%. Therefore, the high-pressure compacted cementitious material with recycled polymer fiber waste may be advantageous for use in some structural applications that require extended service life in saltwater, as it retained most of its average compression strength even after accelerated salt aging.

While the experimental findings and numerical models in this study supported the possibility of recycling carbon and glass fiber polymer HVCC waste into concrete, this study still contained a few limitations. For example, while the high-pressure compacted material showed promise, further investigations will be needed to assess its use as a building material in real-life situations in offshore environments. Additionally, while the high-pressure vibratory compaction system was beneficial in reducing voids and

increasing the density of the cement samples, its relatively high cost, limited mobility, and extended curing time could limit the applications of these cement structures. Nevertheless, we showed that with proper treatment and processing of waste FCP materials, recycling of this waste into cement-based materials may be a valuable alternative to disposal in landfills.

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Declarations

Conflict of interest Dr. Edward Clark received a research grant from CTC Global through the NSF Grant Opportunities for Academic Liaison with Industry program.

Data availability Data available on request from the authors.

Code availability Code available on request from the authors.

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