

Assessing the use and quantity of contaminated plastic as reinforcement in cement mortar: Does biomineralizing the plastic influence the resulting strength?

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Highlights:

- Compressive strength tests were performed on mortar reinforced with waste plastics.
- Clean plastics at 20% replacement produced strengths adequate for many applications.
- A calcium carbonate mineral coating on clean plastics did not improve the strength.
- Mineral coating on oily plastics resulted in similar strengths as washing with water.

Abstract:

The demand for cement infrastructure and plastic products is continuously increasing, and the production of these materials generates greenhouse gas emissions. An additional problem is the low recycling rate of plastics — with only 9% of plastics ever produced being recycled. Instead, these waste plastics are accumulating in landfills and the environment. One contributing factor to low plastic recycling rates is that contamination from food waste and oil necessitates treatment steps that may increase the cost and reduce the quality of recycled plastic. Researchers have made headway against these challenges of increasing greenhouse gas emissions and low recycling rates by using waste plastic as reinforcement in cementitious materials. However, increasing the amount of plastic reinforcement leads to a decrease in composite compressive strength. Past studies indicate that using microbially induced calcium carbonate precipitation (MICP) to coat waste plastic in calcium carbonate may improve the strength of plastic-reinforced cementitious materials. The objective of this study was to increase the amount of clean and contaminated waste plastic that can be added to plastic-reinforced mortar and to assess whether a coating of MICP on the waste plastic enhances the strength. The performance of plastic-reinforced mortar cylinders was investigated using compressive strength tests at a 0%, 5%, 10%,

and 20% volume replacement for cement. Results indicate that even at a 20% replacement with untreated, clean post-use plastics (high density polyethylene (HDPE), polyvinyl chloride (PVC), low density polyethylene chips (LDPE1), and low density polyethylene granules (LDPE2) produced compressive strengths acceptable for several applications such as foundation walls, garages, and sidewalks. However, a coating of MICP on clean waste plastic did not significantly improve the compressive strength of the specimens. MICP treatment of oil-coated waste plastics (HDPE, LDPE1, and LDPE2) recovered the strength by 28.28% relative to cylinders containing untreated oil-coated plastics, on average. However, washing the same oil-coated plastics with water resulted in compressive strengths similar to that of MICP-treated, contaminated plastics. These results demonstrate that incorporating greater volumes of waste plastics into mortar at optimized replacement ratios could improve the sustainability of cementitious composites by the dual mechanisms of reduced cement production and repurposing plastic waste.

Keywords: Microbial-induced calcium carbonate precipitation, plastic-reinforced mortar, waste plastic, oil-contaminated plastic, compressive strength

1.0 Introduction

Cement is the second most-consumed resource after water, and its production generates 5-8% of global anthropogenic greenhouse gas emissions (Izumi et al., 2021; Naqi and Jang, 2019). The production of clinker, the binder in cement products, produces carbon dioxide as a byproduct in three different stages: (1) calcination of limestone, clay, or sand (2) fuel combustion from manufacturing in a rotary kiln, and (3) emissions from quarrying and transportation of products (Naqi and Jang, 2019). By replacing a fraction of the cement paste with waste materials, such as recycled plastic, the overall concrete composite has the potential to be more sustainable.

Between 1950 and 2015, a total of 6300 metric megatons of primary and secondary (recycled) plastic waste were generated (Geyer et al., 2017). Approximately 9% of this plastic was recycled, 12% was incinerated, and only 10% of recycled plastic has been recycled more than once (Geyer et al., 2017). If current production and waste management trends continue, by 2050 about 12,000 metric tons of plastic waste will end up in landfills or the environment (Geyer et al., 2017). An impediment to recycling is that many plastics are coated with food or oily residues, otherwise known as contaminated waste plastics. Plastics that are commonly recycled (e.g., polyethylene terephthalate (PET) or high density polyethylene (HDPE)) are rarely recycled if contaminated because the plastics must be sorted and washed prior to recycling (Ragaert et al.,

2017; Solis and Silveira, 2020). These additional steps are often not economically viable. Additionally, some recycling processes require high operating temperatures as various polymers have different melting temperatures, resulting in higher costs and energy input (Solis and Silveira, 2020). The quality of the final product may also be affected during reprocessing leading to fluctuations in the price of recycled materials (Awaja and Pavel, 2005; Ragaert et al., 2017).

Researchers have attempted to mitigate these problems of low recycling rates and greenhouse gas emissions by integrating waste plastics into cement. The addition of waste materials, including plastic, in reinforced cementitious materials has been extensively investigated (Afroughsabet et al., 2016; Babafemi et al., 2018; Kishore and Gupta, 2019; Naqi and Jang, 2019). However, these additions cause reductions in strength. Kim et al. (2010) found that the compressive strength of PET and polypropylene (PP) fiber reinforced concrete specimens decreased as the fiber volume increased. As little as 1.0% fiber volume fraction results in up to 9% and 10% loss in strength for PET and PP, respectively. This trend is consistent across several other studies (Babafemi et al., 2018; Hannawi et al., 2010; Sharma and Bansal, 2016). To combat the loss of strength, calcium carbonate coating on the waste plastic has been investigated as a method to improve PRM strength (Feng et al., 2021; Hao et al., 2018; Kane et al., 2021). The addition of calcium-carbonate-coated plastic increases the cement hydration, which may improve bond strength with fibers, ultimately increasing the overall composite strength (Feng et al., 2021; Hao et al., 2018).

One method to attach a calcium carbonate coating to plastic is through microbially induced calcium carbonate precipitation (MICP). MICP uses microorganisms, such as the common soil bacteria *Sporosarcina pasteurii* (*S. pasteurii*), to produce the enzyme urease. The hydrolysis of urea forms hydroxide and carbonic acid. The hydroxide shifts the pH forming bicarbonate and carbonate. The carbonate combined with the calcium has the potential to form calcium carbonate precipitate. The microbes produce the enzyme that catalyzes the chemical reaction to induce calcium carbonate precipitation and may become encased in the mineral (Bachmeier et al., 2002; Phillips et al., 2013; Stocks-Fischer et al., 1999).

Kane et al. (2021) evaluated the compressive strength of plastic-reinforced mortar (PRM) containing 5% weight replacement of cement with MICP-treated plastics (PET, polyvinyl chloride (PVC), low density polyethylene (LDPE), PP, polystyrene (PS), or acrylonitrile butadiene styrene (ABS)). While 5% replacement by untreated plastics significantly reduced PRM strength, the same quality replaced by MICP-treated PET, PVC, or mixed type plastics had compressive strengths comparable plastic-free mortar. These early results were encouraging; however, critical

gaps remain regarding the acceptable replacement volumes of waste plastics in PRM materials. First, the impact of MICP treatment on PRM strength has not been examined at greater replacement volumes (i.e., >5%). Next, whether MICP treatment could improve the strength of PRM prepared using contaminated plastics is unknown.

In the first experiment, the strength of PRM was investigated using four types of post-use plastic waste (HDPE chips, PVC chips, LDPE chips, and LDPE granules). These PRM samples were prepared with 5-20% volume replacement by plastics that were either treated with MICP or left untreated. Next, how vegetable oil (used to mimic contamination of plastics) influences calcium carbonate biomineral properties was explored. This experiment compared PRM strength between oily plastic, MICP-treated oily plastic, and oily plastic that had been washed with water. It was hypothesized that treating both clean and contaminated plastics with MICP treatment will improve the compressive strength relative to mortar containing untreated plastics. While the hypothesis did not hold, the results of these studies provide valuable information regarding the strength of PRM containing large replacement volumes of common waste plastics and suitable applications for oil-contaminated PRM. These studies advance the knowledge about the interaction between plastic type, oil contamination, and MICP treatment on PRM strength.

2. Materials and experimental methods

2.1 Materials

2.1.1 Plastic

Post-use, chipped plastic was obtained from Northwest Polymers (Molalla, OR, USA) for addition to plastic-reinforced mortar. The clean plastic types included HDPE, PVC, and two different kinds of LDPE (LDPE1 (chips), LDPE2 (granules)). Rubber residue in the PVC was separated from the chipped plastic and discarded. Sieves narrowed the size of the chipped plastic to + 4.75 mm - 9.52 mm. Photos of the waste plastic are shown in Figure 1.

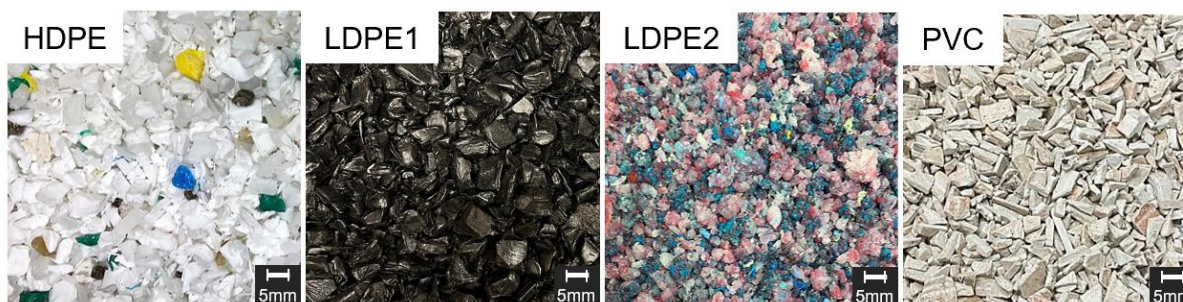


Figure 1. Clean waste plastic gifted by Northwest Polymers.

The density of the chipped plastic was determined using an analytical balance (Mettler Toledo XS205 Dual Range Balance, Density Kit MS-DNY-54). The density of each plastic type is shown in Table 1.

Table 1. Density determined for chipped waste plastic

Plastic Type	Density (g/cm ³)
HDPE	0.938
PVC	1.441
LDPE1	0.928
LDPE2	0.776

2.1.2 Biomineralization solutions

This formerly established MICP protocol has been used in previous studies (Kane et al., 2021; Phillips et al., 2013). Solutions used to perform biomineralization are as followed. The calcium-containing solution (CMM+) was composed of 3 g/L nutrient broth, 10 g/L ammonium chloride, 20 g/L urea, and 48 g/L calcium chloride dihydrate. The solution was brought up to volume, stirred to dissolve the chemicals and adjusted to pH 6.15 using NaOH or HCl prior to adding the calcium chloride dihydrate. The bacteria culture was grown in brain heart infusion (BHI) (37 g/L BHI, 20 g/L urea; BHI+urea). Both solutions were filtered with a 0.22 µm membrane. All chemicals were obtained from Fisher Scientific.

2.2 Biomineralization of plastic

2.2.1 Biomineralization protocol

A flask containing 100 mL of BHI was inoculated with the microbe *Sporosarcina pasteurii* (*S. pasteurii*). The culture was grown for 24 h in an orbital shaker at 30 °C and 150 rpm. Mesh bags (EcoWear-Amazon) containing 121 g of individual plastic types were submerged in 700 mL

of CMM+ in a 1 L beaker (n = 5). The beakers were placed on a stir plate and inoculated with 14 mL of the *S. pasteurii* culture. The beakers were covered loosely with aluminum foil and left to incubate at 135 rpm. After 48 h, the mesh bags were removed from the CMM+. Plastic was removed and air dried on paper towels for 72 h.

2.2.2 Biomineral precipitation on plastic

The change in mass was recorded for plastic fibers after biomineralization. Plastic fibers were used to weigh the precipitate rather than chipped plastic to control for geometry between plastic types. Plastic filament with a diameter of 1.75 mm was purchased from <https://Filaments.ca> (Mississauga, ON, Canada). Plastic types included PET (PETG Filament—White), HDPE (Filament—Natural), PVC (Filamentum Vinyl 303 PVC—Black), LDPE (LLDPE Filament—Natural). Filament was cut to a goal length of 8 mm. Fibers were biomineralized as described in Section 2.2. The mass of the fibers was recorded following biomineralization and air drying for 72 h.

2.2.3 Biomineral precipitation on oil-contaminated plastic

The change in mass was recorded with the addition of vegetable oil to plastic fibers to determine the effect of oil on biomineral deposition. Plastic fibers soaked in Crisco pure vegetable oil (125 mL per each 121 g plastic batch) for 10 minutes (n = 5). Fibers were manually shaken for 15 s to ensure an even coating of oil on all fibers. Fibers were transferred from the plastic container to a mesh bag and submerged into the mineralization solution (Section 2.2). The mass of the fibers was recorded following biomineralization and air drying for 48 h.

2.3 Determination of compressive strength for biomineralized plastics at varying replacement percentages

Mortar cylinders were produced with a mix ratio of 0.46: 1.0: 2.0 by weight of tap water, Portland cement (Quickrete, Commercial Grade Type I/II), and sand (ASTM-Type C778 Graded Sand, U.S. Silica Company) respectively, consistent with ASTM C109 (C109/109M-16a, 2016). Portland cement was substituted by volume with MICP-treated and untreated chipped plastic (HDPE, PVC, LDPE1, LDPE2) at 5%, 10%, and 20% (density determined, where an example mix is presented in Table 2). A consistent volume of plastic was added to the mix by using a volume replacement opposed to weight replacement as the densities of the plastics varied. The chipped plastic was subjected to biomineralization and no biomineralization (n = 5 / group). These components were mixed in a rotary planetary mixer following ASTM International C305 (ANON,

1995). An expanded table of the mortar mix design for all volume replacements is shown in Table S2.

The mortar was cast into cylindrical molds (2 in D x 4 in H; Bio-cylinder, Deslauriers Inc.) using the procedure described in ASTM C192 (ASTM, 2007). The specimens were cast in two layers. Each layer was compacted with tamping and tapping techniques. The specimens were stored in a concrete curing room at 100% relative humidity for 24 h. After initial curing, the specimens were demolded and cured for an additional 27 days in the curing room.

Table 2. Representative mortar mix design for HDPE plastic at 10% cement replacement. Additional mix ratios are shown in Table S2.

Components	Mass (g) per in ³ of mortar	Total (g) (n=5)
Cement	19.405	1219
Sand	9.2	578
Water	40	2513
HDPE Plastic	0.595	37

Compressive strength tests were performed after 28 days for cured mortar cylinders. The specimen height and area were recorded before placing neoprene caps on both sides of the cylinder to ensure an even load rate. The specimens were subjected to compression until failure using a constant load rate of 0.127 mm/s (0.005 in/s) on an MTS Criterion Model 64. A total of five replicates were tested for each plastic type and treatment.

2.4 Mineralogical characterization of biomineral precipitated with and without oil contamination

Biomineral crystalline structures were examined using X-ray diffraction (XRD, Bruker D8 Advance Powder X-ray Diffractometer). Plastic fibers were used to characterize and image the biomineral to control for geometry between plastic types. Mineral was scraped 7 days and 28+ days off plastic fibers and pulverized using a mortar and pestle to obtain homogeneity. The fine powder was mixed with isopropyl alcohol and dried on a glass slide. Samples were analyzed from 3 to 75° 2 θ . Peaks were identified using JADE software. High-resolution images of mineralized and oil-coated, mineralized plastic fibers were obtained by a Zeiss Supra 55VP field emission scanning electron microscope (FESEM) with an SE2 detector at 2–3 kV and a working distance of 5.5–8.5 mm. Prior to analysis, fibers were fixed onto carbon fiber tape and sputter-coated with gold (Emitech K-875X Sputter Coater).

2.5 Evaluating the impact of biomineralization versus washing on mortar cylinder strength

Mortar cylinders were prepared to determine whether compressive strength is generated from the biomineral treatment or washing of fibers, that occurs incidentally to biomineralization. Oil-treated fibers were subjected to washing and/or biomineralization. Oil-contaminated HDPE, LDPE1, and LDPE2 were subjected to water or *S. pasteurii* and CMM+ (n = 5 / group). Mortar specimens were produced with these fibers at a moderate volume replacement of 10% and tested for compressive strength, as described in Section 2.4. All specimens were tested after 75 days of curing. Despite a longer curing time, strengths are not expected to appreciably change after 28 days (Strength, 1926).

2.6 Data analysis

All analyses were performed in Minitab (ver. 19.2020.1, Minitab LLC, State College, PA, USA). The criteria for statistical significance was set *a priori* to 0.05. The effects of plastic type, biomineralization treatment or oil coating, and the interaction between the variables on precipitate mass were examined. One-factor and two-factor ANOVA were used to evaluate the effects of plastic type and oil on the amount of mineral precipitated on the fibers due to biomineralization. Three-factor ANOVA tested the effects of plastic type, biomineralization, and replacement percentage as well as the interaction between these variables on the compressive strength of the mortar. Two-factor ANOVA was also used to evaluate the effects of plastic type, biomineralization treatment or washing, and the interaction between the variables on compressive strength with contaminated waste plastics. In the case of significant interactions, post hoc testing was performed by Tukey multiple pairwise comparisons to distinguish simple effects. All models were checked for residual normality and homoscedasticity.

3 Results

3.1 Biomineral can be deposited on clean and oil-coated plastics

Calcium carbonate mineral precipitated on all plastic types, regardless of an addition of oil. An increase in mineral on oil-coated LDPE (16.07 ± 0.40 g) was observed relative to clean LDPE (13.5 ± 1.35 g) (Figure 2). A significant interaction ($p < 0.05$) was found between plastic type and oil coating on the amount of precipitate accumulated. The mean precipitated mineral varied from 9.9 g on oil-coated PET to 16.5 g on oil-coated LDPE. Post hoc analysis found that a coating of oil on LDPE plastic had a significant effect on the amount of mineral precipitated ($p < 0.05$, 19.01% increase), while for all other plastic types (PET, HDPE, PVC) an oil coating prior to biomineralization did not significantly affect the mineral coating weight ($p > 0.05$).

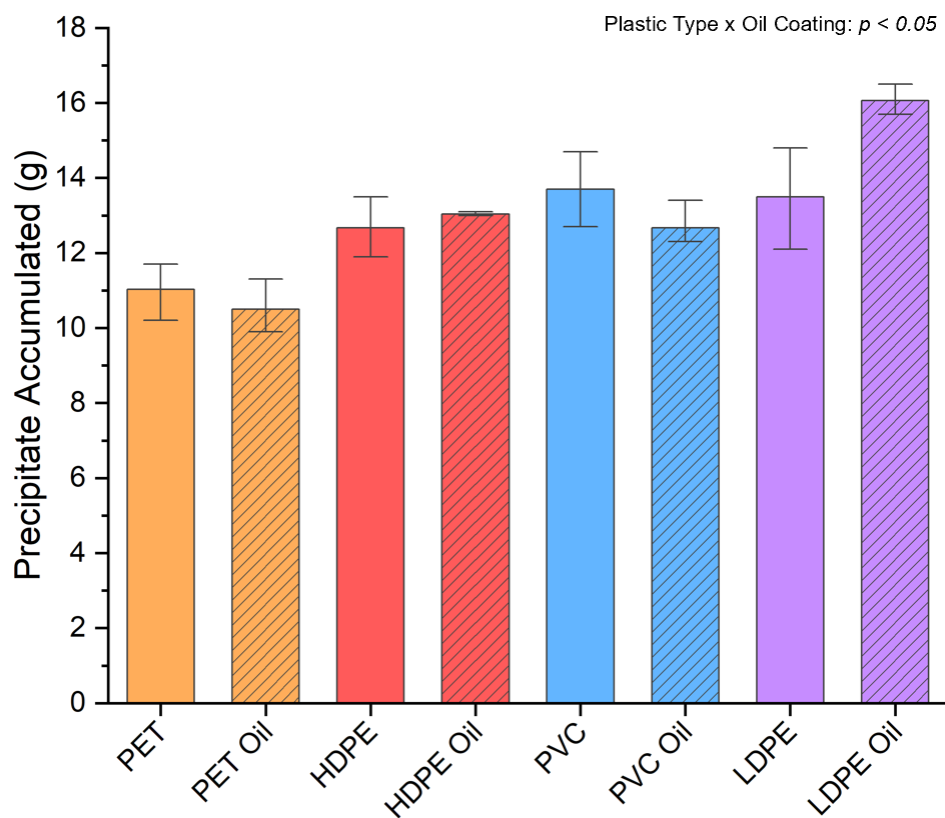


Figure 2. Mass of calcium carbonate attached to 121 g of each plastic type. The bar heights represent the mean, and the error bars indicate one standard deviation.

3.2 Oil contamination of plastic changes biomineral polymorph and geometry

XRD shows that MICP produced two calcium carbonate polymorphs (Figure 3). The predominate mineral on clean plastic fibers was calcite with a minor phase of vaterite. Calcite and vaterite peaks were also observed for oil-coated plastic fibers, although the vaterite signature was larger, suggesting that this phase was more prominent for plastics with oil.

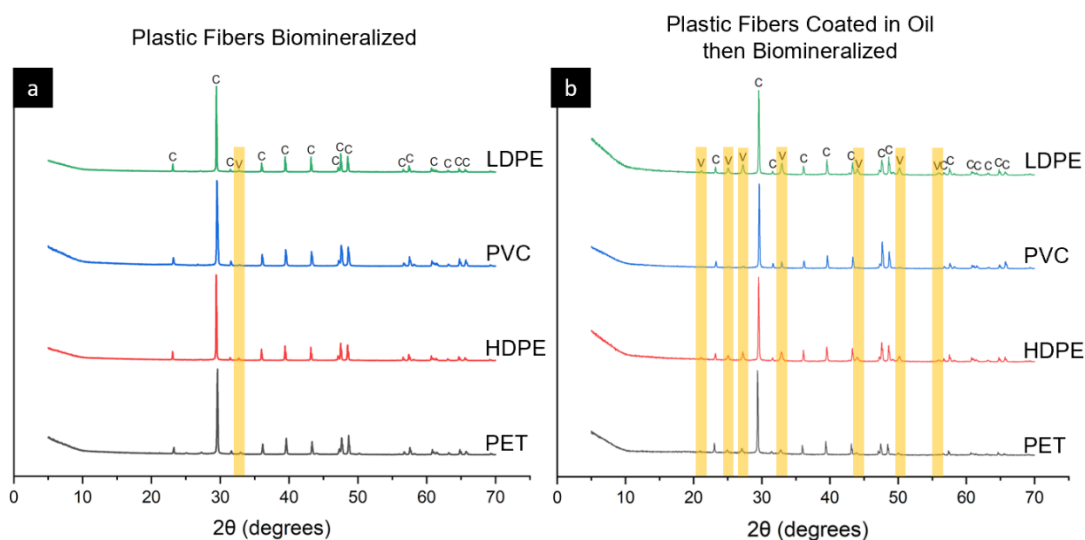


Figure 3. Mineralogy of all samples. **(a)** Mineralized plastic with no oil coating **(b)** Oil-soaked and mineralized plastic. Vaterite peaks are highlighted.

Past work has shown that the metastable vaterite undergoes a phase change to calcite over time (Heveran et al., 2019; Sondi and Salopek-Sondi, 2005). Vaterite was still identified on the oil-coated plastics after 145 ± 5 days of aging (Figure S3). These results possibly indicate that biomineralized oil-coated plastic fibers will have similar properties to freshly biomineralized oil-coated fibers, even after an extended period of time.

The mineral coverage varied for all plastic types with and without oil (Figure 4). The calcium carbonate mineral on plastic with no oil coating yielded a rhombohedral morphology (Figure 4a, c, e, g). This shape corresponds to the mineral calcite, agreeing with the results obtained by the XRD analysis (Figure 3). Hexagonal and spherical minerals were observed on oil-coated HDPE and PVC which exhibited higher vaterite content (Figure 4d, f). The vaterite mineral shape was not distinct on oil-coated plastics PET and LDPE (Figure 4b, h).

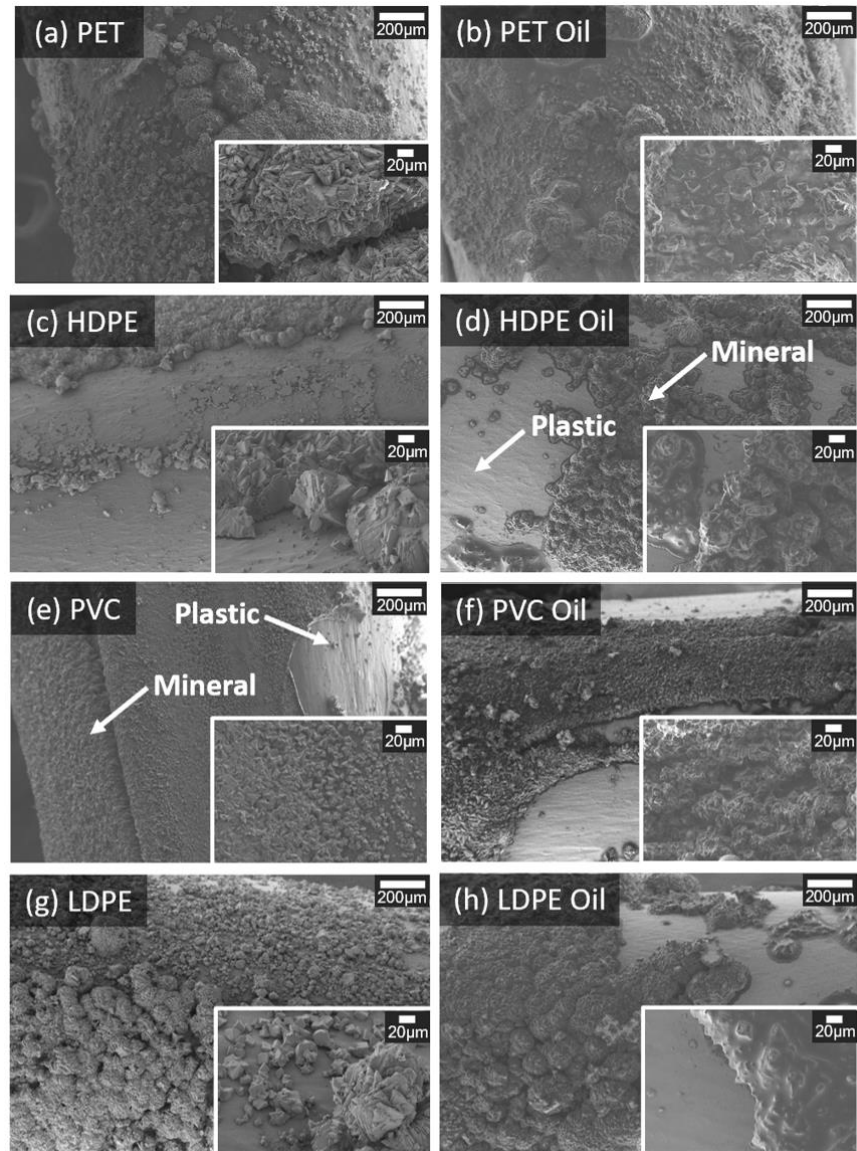


Figure 4. Scanning electron micrographs of biomaterialized plastic types with (a, c, e, and g) and without (b, d, f, and h) the addition of oil at magnifications of 150x and inset images at magnification of 1000x.

3.3 Plastic type and replacement percentage influences mortar strength

Larger decreases in mean compressive strength were observed with increasing untreated plastic content. The mean compressive strengths of PRM prepared with untreated plastic were 11.02% (51.29 MPa), 17.64% (48.50 MPa), and 27.67% (42.60 MPa) less than the mean strength of plastic-free mortar at replacement percentages of 5%, 10%, and 20%, respectively (Figure 5). The 28-day compressive strength decreased 0.57 MPa for every additional 1% of untreated

plastic ($R^2 = 0.70$, $p < 0.05$). Subsequently, it was tested whether MICP would improve PRM strength at each of these replacement quantities.

MICP treatment did not significantly affect the compressive strength of PRM ($p > 0.05$). However, there was a significant interaction ($p < 0.05$) between the effects of plastic type and the percent plastic replacement on the PRM compressive strength. Post hoc analysis showed that cylinders with 10% PVC and 10% LDPE1 both exhibited significantly higher compressive strengths than either PET or LDPE2 at a 10% replacement. Mortar cylinders containing PVC at a 20% replacement had significantly higher compressive strength than any of the other plastic types at this replacement percentage ($p < 0.05$) (Figure 5).

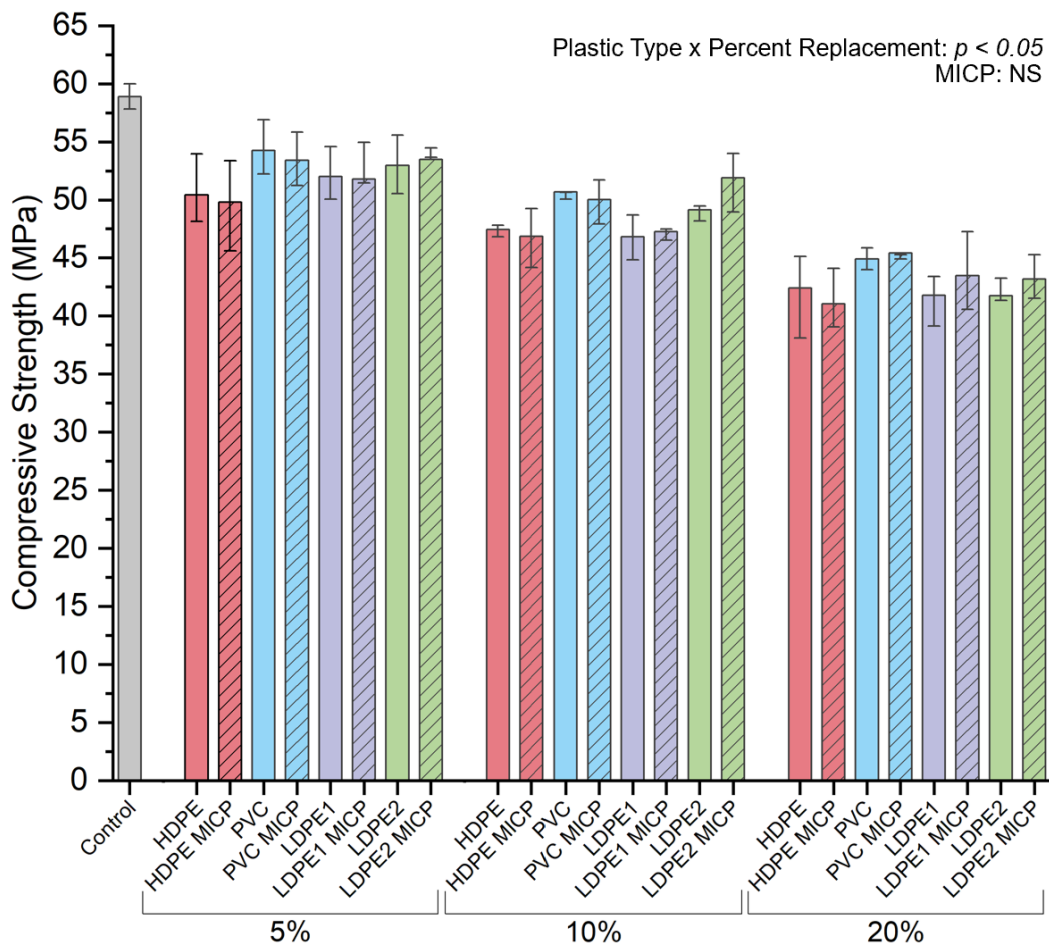


Figure 5. Compressive strengths at replacement percentages 5-20% with waste plastic and no treatment (solid) and MICP-treated waste plastic (striped). Each color indicates a different type of waste plastic: HDPE (red), PVC (blue), LDPE1 (purple), and LDPE2 (green). The bar heights represent the mean, and the error bars indicate one standard deviation.

Our results demonstrate that even at a 20% replacement with untreated, waste plastic (HDPE, PVC, LDPE1, and LDPE2), the resulting mortar showed promising compressive strengths, adequate for several applications, shown in Figure 6.

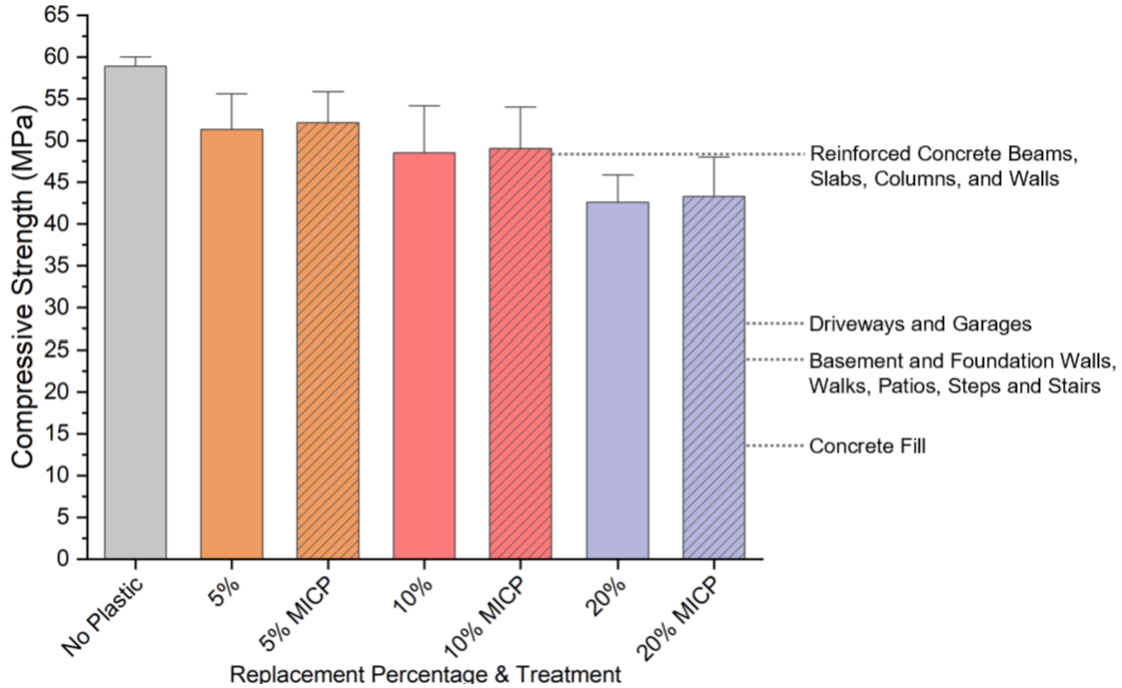


Figure 6. Compressive strengths at replacement percentages 5-20% with waste plastic and no treatment (solid) and MICP-treated waste plastic (striped). Strengths shown are averages across HDPE, PVC, LDPE1, LDPE2 at the given replacement percentage. The bar heights represent the mean, and the error bars indicate one standard deviation. General compressive strength requirements for each type or location of concrete construction is from the International Code Council Concrete Manual: Based on the 2015 IBC® and ACI 318-14 (Strength, 1926).

3.4 PRM strength is reduced by oil unless treated with biomineralization or washing

To examine whether treating oil-coated plastics with MICP or washing will rescue PRM strength, compressive tests were carried out at 10% volume replacement (Figure 7). There was a significant interaction ($p < 0.05$) between plastic type and treatment on the compressive strength of the cylinders. Post hoc testing shows the addition of biomineral or washing of oil-coated HDPE exhibited the greatest strengths for all plastic types coated in oil. For each plastic type (LDPE1, LDPE2, HDPE) the MICP treatment or washing the oil-coated plastic with water showed a similar increase in compressive strength ($p < 0.05$). On average, biomineralization and washing of the oily plastics increased the compressive strength by 28.28% (48.37 MPa) and 27.14% (47.94

MPa), respectively, when compared to cylinders containing untreated, oil-coated plastics (37.71 MPa).

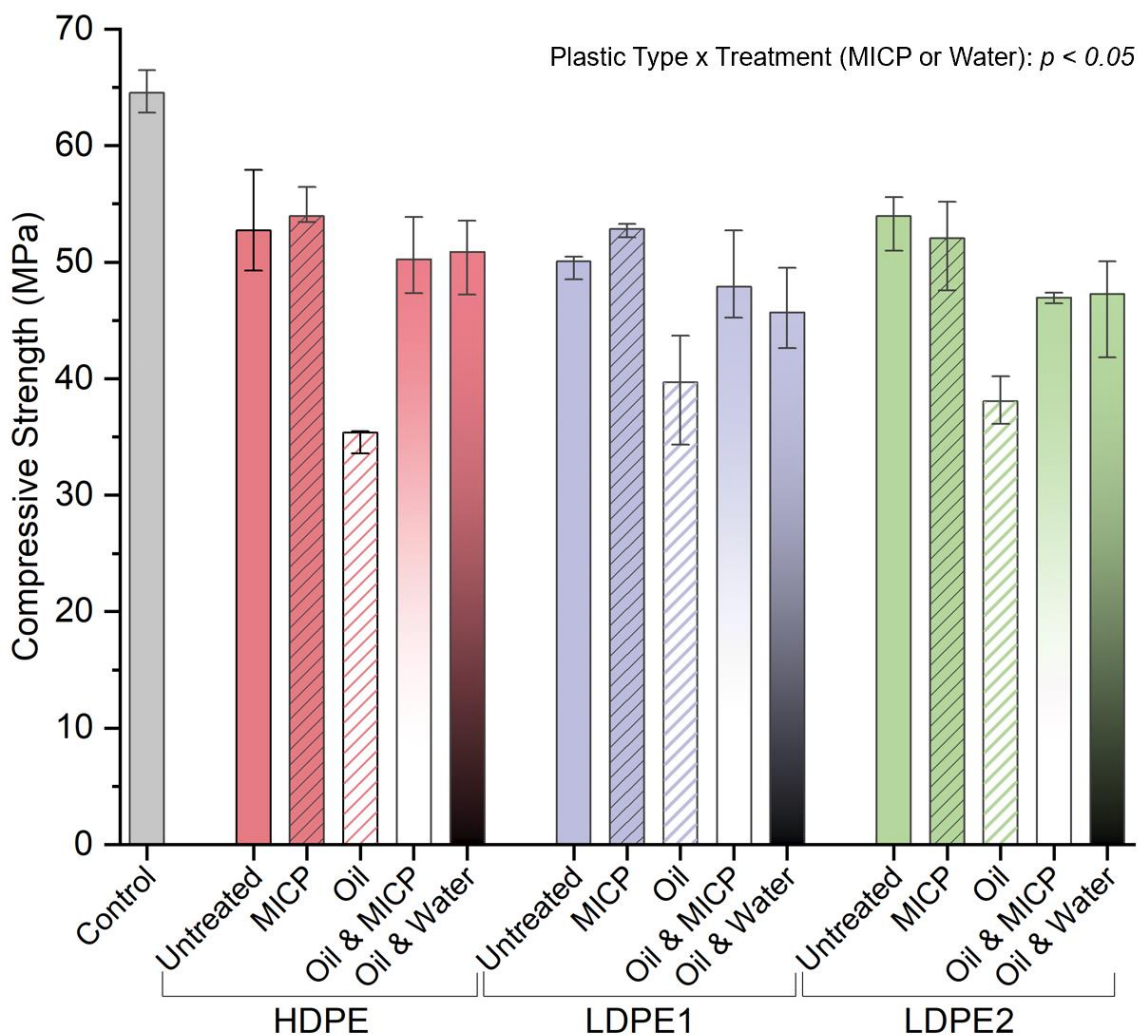


Figure 7. Compressive strengths at 10% volume replacement with waste plastic types HDPE (red), LDPE1 (purple), and LDPE2 (green). Each type of plastic was subjected to five treatments: no treatment (solid), MICP-treated plastic (solid and striped), oil-coated plastic (white and striped), oil-coated and MICP-treated plastic (white to solid), oil-coated and washed with water plastic (black to solid). The bars represent the mean values, and the error bars indicate one standard deviation.

4 Discussion

The objective of this research was to increase the quantity of clean and contaminated waste plastic that can be added to PRM while maintaining adequate composite strength for common low-strength applications where more sustainable alternatives to traditional

cementitious materials are desired. While the addition of plastic as a replacement for aggregate or low volumes of cement has been explored, it is not well understood how greater volume replacements of cement with plastic will impact the compressive strength (Al-Tulaian et al., 2016; Bendjillali et al., 2017; Kane et al., 2021; Kaur and Pavia, 2020; Kim et al., 2021; Safi et al., 2013; Senhadji et al., 2019). Additionally, we sought to determine if MICP treatment of the plastic would improve the strength of the resulting mortar.

First, the impact of cement substitution with waste plastic at 5%, 10%, and 20% replacement volumes on compressive strength was assessed. The 28-day compressive strength results indicated that there was an inverse, linear relationship between the amount of plastic added and PRM strength. Despite the differences in plastic geometry and mix composition, the compressive strengths at a 5% replacement reported in the present study compare well with the strengths of mortar containing 5% untreated plastics (PVC, LDPE, PP) reported by Kane et al. (2021). At a 20% replacement with untreated, waste plastic, the resulting mortar showed compressive strengths adequate for applications such as sidewalks, foundation walls, driveways, and garages (Figure 6) (Strength, 1926). Adding waste plastic to commonly used applications could translate to large sustainability benefits. For example, sidewalks account for nearly 7.2% (6 km²) of the total developed area in Barcelona, Spain as of 2006. Every year in Barcelona, 130,000 m² of the sidewalk area is dug up and replaced, requiring over 14,000 metric tons of cement. (Oliver-Solà et al., 2009). If 20% of the cement used to repair the sidewalks was replaced with waste plastic, at least 2,797 metric tons of cement would be saved annually, which would therefore reduce carbon dioxide emissions by around 20% while saving 913 metric tons (assuming a density of 1.028 g/cm³) of plastic from entering landfills (Monteiro et al., 2017). This plastic waste reuse would represent a significant portion of the currently non-recoverable plastic waste generated in Barcelona (Medina-Mijangos et al., 2021). This simple example demonstrates the potential for large-scale impact for utilizing large volumes of waste plastics in common, lower-load applications.

The choice of waste plastic type influenced PRM strength. The highest compressive strengths were found for LDPE2 and PVC. The differences in compressive strength may relate to different bond strengths between the plastics and the cement mortar. Espinal et al. (2022) found that the bond strength was influenced by the type of plastic and more specifically the surface energies of the plastic opposed to surface roughness or fiber tensile strength. PVC was found to have high bond strength (Espinal et al., 2022), which is in alignment with the high compressive strength of PVC-containing PRM of the current study. LDPE studied by Espinal et al. did not have

high bond strength, which agrees with the low compressive strength found for PRM made with the chipped LDPE1 in the current study. By contrast, PRM made with the granular LDPE2 had high compressive strength, likely signifying a relationship between the geometry of the plastic and the resulting mortar strength. Past studies identified a relationship between fiber shape or size and mechanical strength. Kim et al. (2008) found that the geometry of recycled PET fibers influenced the mechanical bond strength due to the difference in surface energy of the fibers and friction resistance during pullout. Furthermore, Pereira-de-Oliveira et al. (2012) reported that the compressive strength of cement mortar decreases when the acrylic fibers added to the mixture increase in length.

Contrary to the hypothesis of this study, a coating of MICP treatment on the plastics did not significantly increase PRM strength. These findings disagree with work by Kane et al. (2021), which showed that at a 5% weight replacement of cement paste, MICP treatment significantly increased compressive strength for PET and PVC short plastic fibers. Factors that may have caused differences in MICP effectiveness may include plastic geometry and size. Other studies observed a slight increase in flexural strength through the use of MICP treatment (Feng et al., 2021; Hao et al., 2018). However, the current study agrees with findings from several studies. Hao et al. (2018) reported that MICP treatment of PP fibers at a 1% by volume replacement did not significantly affect the compressive strength. Espinal et al. (2022) found that MICP treatment did not improve the interfacial bond strengths, frictional bond strengths, chemical bond energies, or work to pullout measures. Characteristics of the MICP coating may impact whether this treatment improves PRM material properties. Hao et al. (2018) reported that PP with an average coating of 0.094 g CaCO_3/g plastic resulted in an increased fiber pull-out strength, thus improving the interfacial bonding between the plastic and the cement matrix. This fiber pullout strength was higher than for plastic coatings of 0.026 CaCO_3/g plastic and 0.374 g CaCO_3/g plastic (Hao et al., 2018). However, the current study found that the average precipitate on the plastics ranged from 0.091 g CaCO_3/g plastic for PET to 0.113 g CaCO_3/g PVC, which are similar to the coatings achieved by Hao and co-authors. Further investigation of the exact mechanisms by which CaCO_3 coating impacts the strength of PRM materials is needed to determine optimal biomineral coating parameters to improve PRM strength.

Whether the strength of PRM made using oil-contaminated plastic could be rescued through MICP treatment was also investigated. As expected, oil contamination dramatically decreased PRM strength (e.g., -22.06% for a 10% replacement with untreated, oil coated plastic compared to 10% replacement of clean plastic). MICP treatment was successful in coating the

oily plastic and did improve the strength of PRM, but these benefits were not different than simply washing the oily plastic with water. These results suggest that at the volume replacement studied, it is more appropriate to wash contaminated plastic as this treatment method does not require the production of media and the disposal of bacteria waste.

There were several limitations present in this study. Plastic filaments of uniform diameter were used to characterize, image, and weigh the biomineralized fibers opposed to chipped waste plastic because of their uniform geometry. While vegetable oil was used to mimic contaminated waste plastics, a coating of food or other oils may produce differing results than those found in this study. Additional testing is required to determine how MICP treatment affects other valuable mechanical properties of these composites such as durability and flexural strength.

5 Conclusion

Experimental data indicates that PRM manufactured with a 20% replacement of untreated plastics produced compressive strengths acceptable for several applications such as driveways, foundation walls and slabs, and concrete fill. A key finding of this study was that MICP coating on several types of common waste plastic does not significantly improve the PRM compressive strength at replacement volumes of 5-20%. MICP was able to rescue the strength of PRM made from oil-contaminated plastic, but the benefit to strength was not greater than for simply washing the plastic. Together, these findings suggest that by substituting a portion of the cement in mortar with clean and contaminated plastics, the sustainability of cementitious composites could be improved through the dual mechanisms of reduced cement production and repurposing plastic waste.

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380 The authors declare no competing interests in this publication.

381 **Author contributions**

382 **Kylee Rux:** Conceptualization, Methodology, Formal Analysis, Investigation, Data Curation,
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387 Review & Editing, Funding acquisition **Chelsea Heveran:** Conceptualization, Methodology,
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References

- Afroughsabet, V., Biolzi, L., Ozbakkaloglu, T., 2016. High-performance fiber-reinforced concrete: a review, *Journal of Materials Science*. Springer US.
<https://doi.org/10.1007/s10853-016-9917-4>
- Al-Tulaian, B.S., Al-Shannag, M.J., Al-Hozaimy, A.R., 2016. Recycled plastic waste fibers for reinforcing Portland cement mortar. *Constr. Build. Mater.* 127, 102–110.
<https://doi.org/10.1016/j.conbuildmat.2016.09.131>
- ANON, 1995. Standard practice for shotcrete 14–16. <https://doi.org/10.1520/C0305-20.2>
- ASTM, 2007. ASTM 6433 Standard Practice for Roads and Parking Lots Pavement Condition Index Surveys 1–8. <https://doi.org/10.1520/C0192>
- Awaja, F., Pavel, D., 2005. Recycling of PET. *Eur. Polym. J.* 41, 1453–1477.
<https://doi.org/10.1016/j.eurpolymj.2005.02.005>
- Babafemi, A.J., Šavija, B., Paul, S.C., Anggraini, V., 2018. Engineering properties of concrete with waste recycled plastic: A review. *Sustain.* 10. <https://doi.org/10.3390/su10113875>
- Bachmeier, K.L., Williams, A.E., Warmington, J.R., Bang, S.S., 2002. Urease activity in microbiologically-induced calcite precipitation. *J. Biotechnol.* 93, 171–181.
[https://doi.org/10.1016/S0168-1656\(01\)00393-5](https://doi.org/10.1016/S0168-1656(01)00393-5)
- Bendjillali, K., Chemrouk, M., Boulekbache, B., 2017. European Journal of Environmental and Civil Engineering Performances of cementitious mortars containing recycled synthetic fibres under hot-dry climate Performances of cementitious mortars containing recycled synthetic fibres under hot-dry climate. <https://doi.org/10.1080/19648189.2017.1344152>
- C109/109M-16a, A., 2016. Standard test method for compressive strength of hydraulic cement mortars (Using 2-in. or cube specimens). *Annu. B. ASTM Stand.* 04, 1–10.
<https://doi.org/10.1520/C0109>
- Espinal, M., Kane, S., Ryan, C., Phillips, A., Heveran, C., n.d. Evaluation of the bonding properties between low-value plastic fibers treated with 1 microbially-induced calcium carbonate precipitation and cement mortar 2 3.
- Feng, J., Yang, F., Qian, S., 2021. Improving the bond between polypropylene fiber and cement matrix by nano calcium carbonate modification. *Constr. Build. Mater.* 269, 121249.

419 <https://doi.org/10.1016/j.conbuildmat.2020.121249>

420 Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made.
 421 Sci. Adv. 3, 3–8. <https://doi.org/10.1126/sciadv.1700782>

422 Hannawi, K., Kamali-Bernard, S., Prince, W., 2010. Physical and mechanical properties of
 423 mortars containing PET and PC waste aggregates. Waste Manag. 30, 2312–2320.
 424 <https://doi.org/10.1016/j.wasman.2010.03.028>

425 Hao, Y., Cheng, L., Hao, H., Shahin, M.A., 2018. Enhancing fiber/matrix bonding in
 426 polypropylene fiber reinforced cementitious composites by microbially induced calcite
 427 precipitation pre-treatment. Cem. Concr. Compos. 88, 1–7.
 428 <https://doi.org/10.1016/j.cemconcomp.2018.01.001>

429 Heveran, C.M., Liang, L., Nagarajan, A., Hubler, M.H., Gill, R., Cameron, J.C., Cook, S.M.,
 430 Srubar, W. V., 2019. Engineered Ureolytic Microorganisms Can Tailor the Morphology and
 431 Nanomechanical Properties of Microbial-Precipitated Calcium Carbonate. Sci. Rep. 9, 1–
 432 13. <https://doi.org/10.1038/s41598-019-51133-9>

433 Izumi, Y., Iizuka, A., Ho, H.J., 2021. Calculation of greenhouse gas emissions for a carbon
 434 recycling system using mineral carbon capture and utilization technology in the cement
 435 industry. J. Clean. Prod. 312, 127618. <https://doi.org/10.1016/j.jclepro.2021.127618>

436 Kane, S., Thane, A., Espinal, M., Lunday, K., Armağan, H., Phillips, A., Heveran, C., Ryan, C.,
 437 2021. Biomineralization of plastic waste to improve the strength of plastic-reinforced
 438 cement mortar. Materials (Basel). 14, 1–19. <https://doi.org/10.3390/ma14081949>

439 Kaur, G., Pavia, S., 2020. Physical properties and microstructure of plastic aggregate mortars
 440 made with acrylonitrile-butadiene-styrene (ABS), polycarbonate (PC), polyoxymethylene
 441 (POM) and ABS/PC blend waste. J. Build. Eng. 31, 101341.
 442 <https://doi.org/10.1016/j.jobbe.2020.101341>

443 Kim, J.H.J., Park, C.G., Lee, Si Won, Lee, Sang Woo, Won, J.P., 2008. Effects of the geometry
 444 of recycled PET fiber reinforcement on shrinkage cracking of cement-based composites.
 445 Compos. Part B Eng. 39, 442–450. <https://doi.org/10.1016/j.compositesb.2007.05.001>

446 Kim, M.O., Lee, H.K., Kim, H.K., 2021. Cost and environmental effects of ocean-borne plastic
 447 flakes in cement mortar considering equivalent-strength mix design. Constr. Build. Mater.
 448 291, 123267. <https://doi.org/10.1016/j.conbuildmat.2021.123267>

449 Kim, S.B., Yi, N.H., Kim, H.Y., Kim, J.H.J., Song, Y.C., 2010. Material and structural
 450 performance evaluation of recycled PET fiber reinforced concrete. *Cem. Concr. Compos.*
 451 32, 232–240. <https://doi.org/10.1016/j.cemconcomp.2009.11.002>

452 Kishore, K., Gupta, N., 2019. Application of domestic & industrial waste materials in concrete: A
 453 review. *Mater. Today Proc.* 26, 2926–2931. <https://doi.org/10.1016/j.matpr.2020.02.604>

454 Medina-Mijangos, R., Ajour, S., Zein, E., Guerrero-García-Rojas, H., Seguí-Amórtégui, L., 2021.
 455 The economic assessment of the environmental and social impacts generated by a light
 456 packaging and bulky waste sorting and treatment facility in Spain: a circular economy
 457 example. <https://doi.org/10.1186/s12302-021-00519-6>

458 Monteiro, P.J.M., Miller, S.A., Horvath, A., 2017. Towards sustainable concrete. *Nat. Mater.* 16,
 459 698–699. <https://doi.org/10.1038/nmat4930>

460 Naqi, A., Jang, J.G., 2019. Recent progress in green cement technology utilizing low-carbon
 461 emission fuels and raw materials: A review. *Sustain.* 11.
 462 <https://doi.org/10.3390/su11020537>

463 Oliver-Solà, J., Josa, A., Rieradevall, J., Gabarrell, X., 2009. Environmental optimization of
 464 concrete sidewalks in urban areas. *Int. J. Life Cycle Assess.* 14, 302–312.
 465 <https://doi.org/10.1007/s11367-009-0083-7>

466 Pereira-De-Oliveira, L.A., Castro-Gomes, J.P., Nepomuceno, M.C.S., 2012. Effect of acrylic
 467 fibres geometry on physical, mechanical and durability properties of cement mortars.
 468 *Constr. Build. Mater.* 27, 189–196. <https://doi.org/10.1016/j.conbuildmat.2011.07.061>

469 Phillips, A.J., Gerlach, R., Lauchnor, E., Mitchell, A.C., Cunningham, A.B., Spangler, L., 2013.
 470 Engineered applications of ureolytic biomineralization: A review. *Biofouling* 29, 715–733.
 471 <https://doi.org/10.1080/08927014.2013.796550>

472 Ragaert, K., Delva, L., Van Geem, K., 2017. Mechanical and chemical recycling of solid plastic
 473 waste. *Waste Manag.* 69, 24–58. <https://doi.org/10.1016/j.wasman.2017.07.044>

474 Safi, B., Saidi, M., Aboutaleb, D., Maallem, M., 2013. The use of plastic waste as fine aggregate
 475 in the self-compacting mortars: Effect on physical and mechanical properties. *Constr. Build.*
 476 *Mater.* 43, 436–442. <https://doi.org/10.1016/j.conbuildmat.2013.02.049>

477 Senhadji, Y., Siad, H., Escadeillas, G., Benosman, A.S., Chihaoui, R., Mouli, M., Lachemi, M.,
 478 2019. Physical, mechanical and thermal properties of lightweight composite mortars

containing recycled polyvinyl chloride. *Constr. Build. Mater.* 195, 198–207.
<https://doi.org/10.1016/j.conbuildmat.2018.11.070>

Sharma, R., Bansal, P.P., 2016. Use of different forms of waste plastic in concrete - A review. *J. Clean. Prod.* 112, 473–482. <https://doi.org/10.1016/j.jclepro.2015.08.042>

Solis, M., Silveira, S., 2020. Technologies for chemical recycling of household plastics – A technical review and TRL assessment. *Waste Manag.* 105, 128–138.
<https://doi.org/10.1016/j.wasman.2020.01.038>

Sondi, I., Salopek-Sondi, B., 2005. Influence of the primary structure of enzymes on the formation of CaCO_3 polymorphs: A comparison of plant (*Canavalia ensiformis*) and bacterial (*Bacillus pasteurii*) ureases. *Langmuir* 21, 8876–8882.
<https://doi.org/10.1021/la051129v>

Stocks-Fischer, S., Galinat, J.K., Bang, S.S., 1999. Microbiological precipitation of CaCO_3 . *Soil Biol. Biochem.* 31, 1563–1571. [https://doi.org/10.1016/S0038-0717\(99\)00082-6](https://doi.org/10.1016/S0038-0717(99)00082-6)

Strength, K.O.F., 1926. The Strength of Concrete. *Science* (80-.). 64.
<https://doi.org/10.1126/science.64.1660.xii.t>

Supplementary Materials: The Influence of Biomineralized Contaminated Waste Plastics in Reinforced Cement Mortar

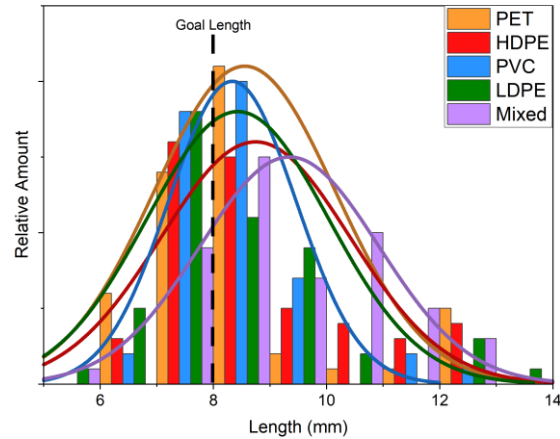


Figure S1. Distribution of fiber lengths tested in this study with a goal length of 8 mm. Filament was cut using a paper cutter fitted with a 3D-printed cutting jig. 50 fibers of each plastic type were selected randomly and measured with calipers.

Table S1. Density determined for all printer filament plastic types. Density of the plastic was calculated by recording the mass and determining the volume using caliper measurements. The resulting density was found by averaging a total of 30 plastic fibers for each plastic type.

Plastic Type	Density
PET	1.178
HDPE	0.913
PVC	1.292
LDPE	0.919
Mixed	1.027

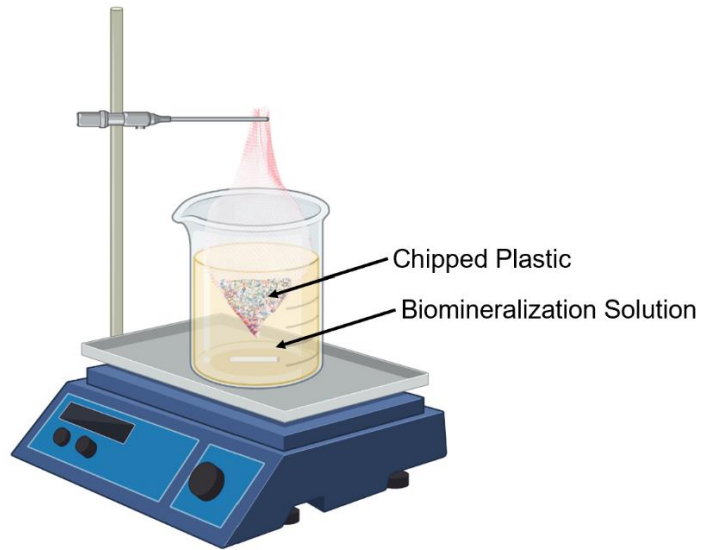


Figure S2. Biomineralization setup for precipitating calcium carbonate mineral on chipped plastic

Table S2. Mortar mix design for all plastic types at 5%, 10%, and 20% volume replacements using plastic densities shown in Table 1.

Percent Replacement	Plastic Type	Components	Total (g) (n=5)
5%	HDPE	Cement	1237.93
		Sand	2513.27
		Water	578.05
		Plastic	18.71
	PVC	Cement	1227.28
		Sand	2513.27
		Water	578.05
		Plastic	29.36
	LDPE1	Cement	1238.13
		Sand	2513.27
		Water	578.05
		Plastic	18.51
	LDPE2	Cement	1241.15
		Sand	2513.27
		Water	578.05
		Plastic	15.49

10%	HDPE	Cement	1219.22
		Sand	2513.27
		Water	578.05
		Plastic	37.41
	PVC	Cement	1197.91
		Sand	2513.27
		Water	578.05
		Plastic	58.72
	LDPE1	Cement	1219.62
		Sand	2513.27
		Water	578.05
		Plastic	37.02
	LDPE2	Cement	1225.66
		Sand	2513.27
		Water	578.05
		Plastic	61.95
20%	HDPE	Cement	1181.81
		Sand	2513.27
		Water	578.05
		Plastic	74.82
	PVC	Cement	1139.19
		Sand	2513.27
		Water	578.05
		Plastic	117.45
	LDPE1	Cement	1182.59
		Sand	2513.27
		Water	578.05
		Plastic	74.04
	LDPE2	Cement	1194.69
		Sand	2513.27
		Water	578.05
		Plastic	61.95

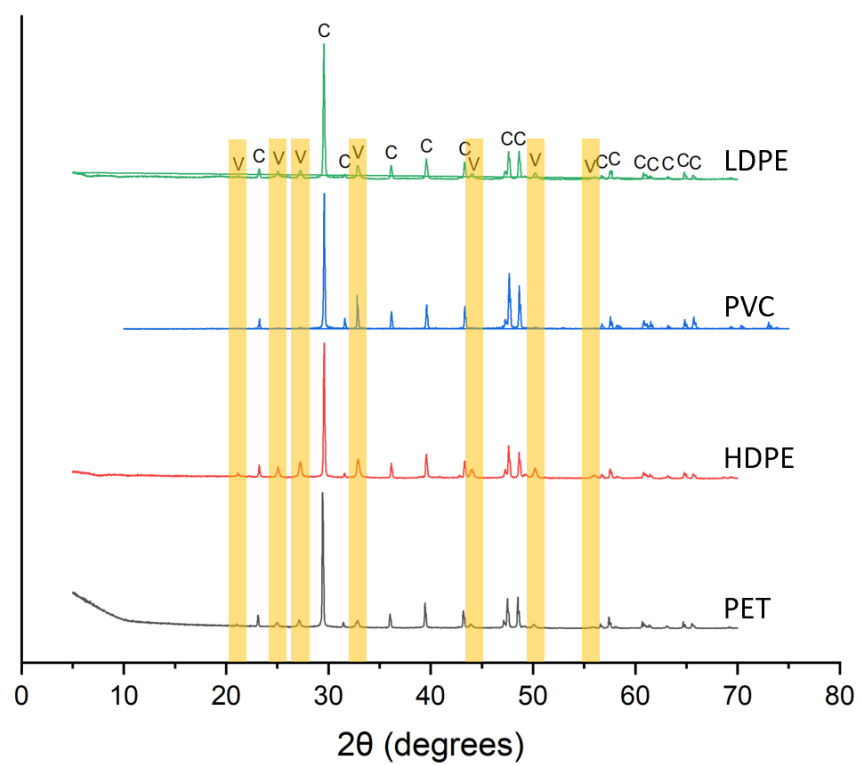


Figure S3. Vaterite is still present on oil-coated and biomineralized fibers after 145 ± 5 days of drying. Vaterite peaks are highlighted.

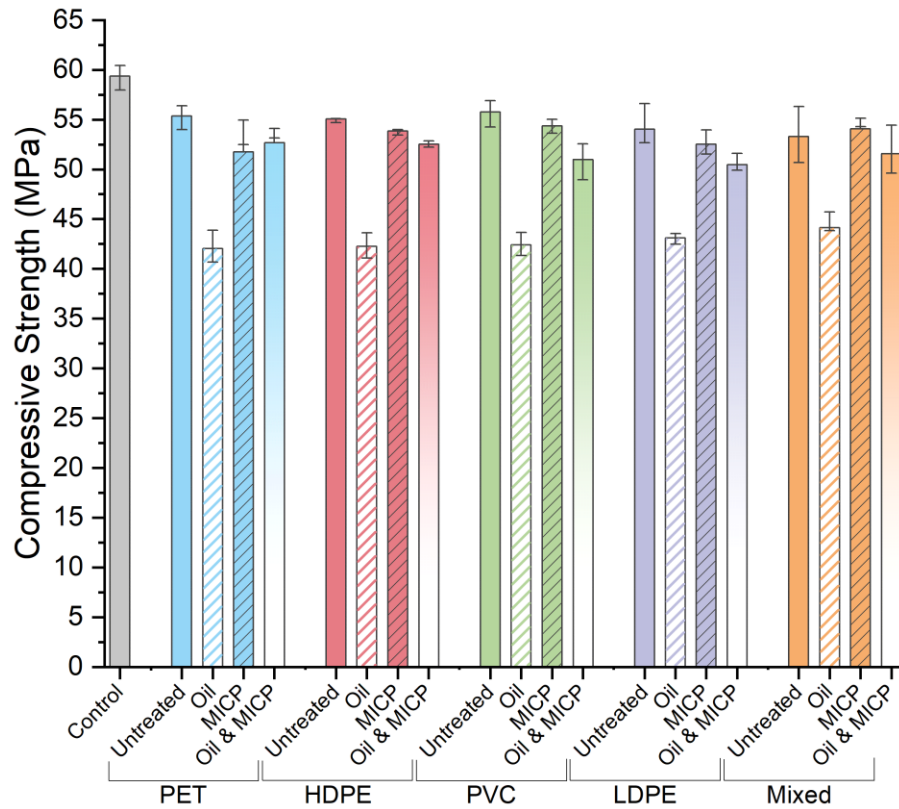


Figure S4. Preliminary experiment showing compressive strengths at 5% volume replacement with 3D printer filament (1) plastic and no treatment, (2) oil-coated plastic, (3) MICP-treated plastic, (4) oil coated, MICP-treated plastic. The bars represent the mean values, and the error bars indicate one standard deviation. Biom mineralization of plastics resulted in similar strength as untreated plastics.

Table S3. Tukey post hoc results and groupings of precipitate mass and compressive strengths. Groupings of different letters signify statistically different mean values. Post hoc tests were only conducted for significant main effects and interactions.

Mass of Calcium Carbonate Precipitate Tukey Pairwise Comparisons: Plastic Type*Oil (y/n)			
Interaction (g)			

Plastic*Oil (y/n)	N	Mean	Grouping		
LDPE y	3	16.0667	A		
PVC n	3	13.7000	B		
LDPE n	3	13.5000	B		
HDPE y	3	13.0333	B	C	
HDPE n	3	12.6667	B	C	D

PVC y	3	12.6667	B	C	D
PET n	3	11.0333		C	D
PET y	3	10.5000			D

Compressive Strengths at Various Replacement Volumes Tukey Pairwise Comparisons:
Plastic Type*Percentage (MPa)

Plastic*Percentage	N	Mean	Grouping		
LDPE2 5%	10	53.2213	A		
LDPE1 5%	10	51.8772	A	B	
PVC 5%	10	51.5902	A	B	
LDPE2 10%	10	50.5012		B	
PVC 10%	10	50.3482		B	
HDPE 5%	10	50.1092		B	
HDPE 10%	10	47.1483			C
LDPE1 10%	10	47.0334			C D
PVC 20%	10	45.1685			D
LDPE1 20%	10	42.6273			E
LDPE2 20%	10	42.4575			E
HDPE 20%	10	41.71981			E

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Compressive Strengths at 10% Replacement with Different Treatments Tukey Pairwise
Comparisons: Plastic Type*Treatment (MPa)

Plastic*Treatment	N	Mean	Grouping		
HDPE No oil, MICP, No water	5	54.0087	A		
LDPE1 No oil, MICP, No water	5	53.5532	A	B	
LDPE2 No oil, MICP, No water	5	53.1711	A	B	
HDPE No oil, No MICP, No water	5	52.9027	A	B	
LDPE2 No oil, No MICP, No water	5	52.8802	A	B	
HDPE Oil, No MICP, Water	5	50.8947	A	B	C
HDPE Oil, MICP, No water	5	50.2544		B	C D
LDPE1 No oil, No MICP, No water	5	50.0748		B	C D

LDPE1 Oil, MICP, No water	5	47.8976	C	D	E		
LDPE2 Oil, No MICP, Water	5	47.2583		D	E		
LDPE2, Oil, MICP, No water	5	46.9637		D	E		
LDPE1 Oil, No MICP, Water	5	45.6700			E		
LDPE1 Oil, No MICP, No water	5	39.6759				F	
LDPE2 Oil, No MICP, No water	5	38.0632				F	G
HDPE Oil, No MICP, No water	5	35.3832					G

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