



Potential of distributed recycling from hybrid manufacturing of 3-D printing and injection molding of stamp sand and acrylonitrile styrene acrylate waste composite

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ABSTRACT

In the Upper Peninsula of Michigan, over 500 million tons of copper rich rock were removed from mines and treated in chemical baths to extract copper. Toxic substances have been seeping into the watersheds from the resultant waste stamp sands. Recent work on developing a circular economy using recycled plastic for distributed manufacturing technologies has proven promising, and this study investigates the potential to use this approach to form stamp sand and acrylonitrile styrene acrylate (ASA) composites. Specifically, this study found the maximum amount of stamp sand that was able to be added to waste ASA by mass with a single auger recyclebot system for compounding was below 40%. The mechanical properties of the composite were evaluated up to 40%, and the addition of stamp sand reduced the material's ultimate tensile strength by about half compared to the strength of raw recycled ASA, regardless of the percent stamp sand in the composite. However, this strength reduction plateaus and the tensile strength of the ASA and stamp sand composites can be compared favorably with acrylonitrile butadiene styrene (ABS) at any level. This makes waste ASA- stamp sand composites potential replacements for outdoor applications of ABS as well as some current ASA applications. These results are promising and call for future work to evaluate the technical, economic and environmental potential for waste ASA - stamp sand composites.

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1. Introduction

Between 1850 and 1968, copper and other ores were heavily mined in the Upper Peninsula of Michigan. Over 500 million tons of copper rich rock were removed from mines and treated in a chemical bath to extract more copper [1], then dumped into the areas surrounding Lake Superior and all of its inlets [2]. Copper, as well as other toxic substances¹ such as mercury and arsenic have been seeping into the watersheds from the stamp sands surrounding the Upper Peninsula since the mining era began [3,4]. These toxic substances have damaged ecosystems in and around Lake Superior by harming native fish and damming stream outlets [4,5] as well as being more harmful for amphibians [6]. When tests indicating that the levels of these minerals were dangerously high in areas such as Torch Lake, small amounts of stamp sand were excavated

[7]. However, stamp sands are unstable, highly mobile [5] and toxic materials [8], which remain an environmental scourge on the entire region as they contaminate the water including Lake Superior. For example, through littoral drift, an estimated 15 million cubic yards of stamp sands threaten the Buffalo Reef, which serves as a spawning habitat for whitefish and about a quarter of the annual lake trout yield from Lake Superior's Michigan waters [9]. Stamp sand beaches often have little vegetation as grass and shrubs are not able to grow in the toxic environment that stamp sand causes [1].

Various industries and research groups have attempted to find a use for stamp sands to enable their economic removal from the environment. Stamp sands are sometimes used in some Upper Peninsula communities for winter maintenance, but this contributes to contamination of the environment. Some applications of stamp sand such as additions of sand to Portland cement or flooring tiles were proved beneficial [10]. It has been proposed to be a fine aggregate in concrete [11] and the EPA has awarded a grant to investigate the potential of stamp sand to be used in roofing shingles [12]. However, because of the amount of stamp sand in the Upper Peninsula, no commercial application has been found to remove even a significant fraction of the waste material from contaminating nature. This is in part because although the cost

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¹For example, in areas like Torch Lake, where 200 million tons of stamp sand were deposited, cupric ammonium carbonate, lime, pyridine oil, coal-tar and wood creosotes, pine oil, and xanthates have also been found in soil samples [7].

of stamp sand is essentially its transportation cost (the material can be thought of as a free industrial waste), the Upper Peninsula is sparsely populated, geographically isolated and economically depressed in part because of high energy costs that reduce business viability [13]. A means to use more stamp sand in substantial quantities in such a rural isolated environment is needed.

Today new types of currently human-generated waste are threatening water systems [14,15] with 275 million metric tons (MT) of plastic waste generated in 192 coastal countries in 2010, with 4.8 to 12.7 million MT entering the ocean [16]. In addition, to the oceans, fresh water such as the Great Lakes have also been negatively affected by plastic pollution. One study detailing plastic particles on Lake Huron's beaches found 2984 pellets, 108 fragments, and 117 pieces of Styrofoam, over an area of only 85 m² [17]. In Lake Superior, there is also a significant amount of microplastics [18]. Perhaps most concerning are weather resistant plastics that can survive in the environment even longer than common plastics that last over 450 years [19–21]. One plastic, with particularly strong weather resistance is acrylonitrile styrene acrylate (ASA) [22–24]. ASA applications include exterior siding, signage, and side mirror housings on automobiles because it is chemically resistant [25], UV resistant, and resistant to color fading and aging [24]. As ASA plastic is weather resistant, it does not wear easily in outdoor environments and therefore would be expected to persist longer in the natural environment than other plastics creating similar problems of the long-life waste of stamp sand.

What if two wrongs could make a right – by combining these two wastes into a new material that can be used for manufacturing products locally? Recent work on developing a circular economy using recycled plastic for additive manufacturing (AM) technologies has proven promising [26,27] for a wide range of plastics including: polylactic acid (PLA) [28–33], acrylonitrile butadiene styrene (ABS) [33–37], high-density polyethylene (HDPE) [38–40], polypropylene (PP) [33,40], polystyrene (PS) [40], polyethylene terephthalate (PET) [33,41], linear low density polyethylene (LLDPE) and low density polyethylene (LDPE) [42], polycarbonate (PC) [43] and elastomers [28,44]. These studies all used single material waste streams, but other studies have studied composites including various types of waste wood or waste cellulose (paper and cardboard) with various plastics [45–47]. There is an established hierarchy of distributed recycling processing techniques [48] that include first mechanical grinding, then the particles are 1) the particles are directly printed with fused particle fabrication (FPF) or a heated syringe printer, if not amenable then 2) converted to filament using a recyclebot and 3-D printed using fused filament fabrication (FFF), and again if not amenable then 3) the particles are injection molded in a custom 3-D printed mold [43]. Thus, distributed recycling can be used in distributed manufacturing of bespoke products. Following that work, this study investigates the potential to combine two harmful waste materials (stamp sand and ASA) into a useful material, which can be injection molded into 3-D printed PC molds allowing for distributed manufacturing. The particle size distribution of stamp sand and waste ASA were quantified and then formulations of 0–20% stamp sand by mass were compounded using a recyclebot waste plastic extruder, pelletized, and then injection molded into ASTM type IV tensile bars. As the higher stamp sand percentages were not able to be mixed uniformly by the recyclebot, they were dry mixed and further mixed in the injection molder. The mechanical properties of the waste stamp sand recycled ASA were quantified and the break surfaces were analyzed using optical microscopy. The results are presented and discussed in the context of eliminating stamp sand in the Upper Peninsula of Michigan by using it waste polymer composites.

2. Materials and methods

2.1. Materials

Stamp sand was acquired from North Canal Township Park, MI. Black recycled ASA was provided by McDonnough Plastics, Fenton

MI for \$1/lb. (\$2.20/kg). The particle size characteristics of the stamp sand and recycled ASA starting material were quantified using digital imaging and the open source Fiji/ImageJ following a standard methodology [43] to determine the size distributions of the particles.

2.2. Methods

Mixtures of stamp sand and recycled ASA were made of 0%, 10%, 20% 40% stamp sand: ASA. The percentages were determined by mass on a scale with certainty ± 0.1 g. Mixtures were vacuum dried in a vacuum oven at 70 °C for 8 h and mechanically mixed. Mixtures below 40% stamp sand were compounded using a recyclebot waste plastic extruder [38], which was the limit of percentage limit of sand able to be used on the machine. During operation of the recyclebot, the stamp sand and ASA plastic mixtures were poured into a 3-D printed funnel apparatus connected to the top of the recyclebot's auger. The recyclebot operated at 280 °C and rotated approximately 7.75 rotations per minute while extruding. The resultant filaments were pelletized [49]. Particle size characterization following the method described above was also utilized for the pellets. 40% stamp sand and ASA was mixed dry and directly used.

To find the density of the stamp sand a Micromeritics AccuPyc 1330 certain to ± 0.0001 g/cc was used. A small amount of stamp sand is placed in a cylinder, massed, and then placed into the machine. The machine uses the known volume of the cylinder and measures the amount of helium gas needed to fill the cylinder entirely to calculate the space that the material takes up and uses the inputted mass of the sample to give the density. For this data point, 5 tests were done to minimize error.

ASTM D638 Type 4 standard tensile bars [50] molds were fabricated with a Lulzbot TAZ 6 open source 3-D printer (Aleph Objects, Loveland CO) with 2.85 mm PC filament (Gizmodorks, Temple City, CA). The PC molds were then covered in an aluminum tape to give its internal faces a smooth molding surface and increase its lifespan. A small amount of the mixture was poured into the heating tube of an open source injection molding system shown in Fig. 1. As in Fig. 1 a tube, heated with a nichrome wire coil, is filled with the ASA mixture and a plunger is manually operated to inject the melted mixture into the mold. The mold of the ASTM type IV tensile bars consists on a top and bottom layer of aluminum sheet, and a middle layer that was 3-D printed out of polycarbonate. The injection molding system was set to

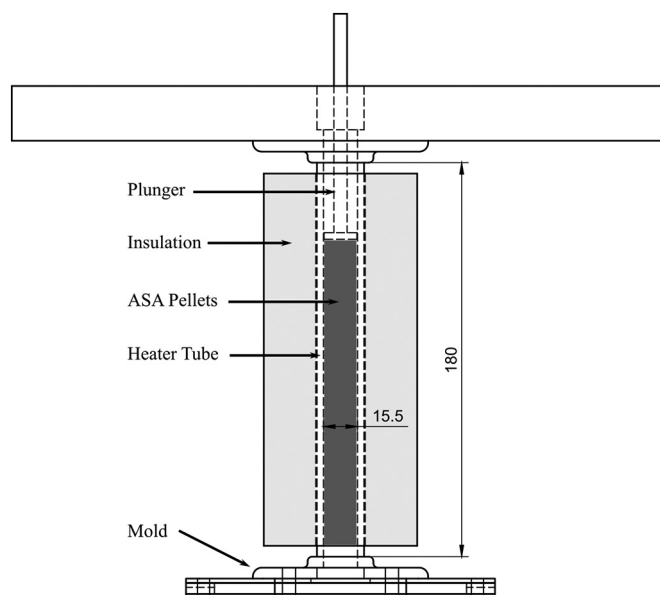


Fig. 1. Schematic of open source injection molding system.

280 °C, and the composite material was heated for approximately 15 min. A stir rod was used to further mix the stamp sand and ASA together once it was melted, and then the mold was screwed firmly to the end of the heating tube. A push rod was then pressed into the top of the tube, extruding the composite material into the mold. 100% Extra Virgin Olive Oil Cooking Spray (Essential Everyday brand) was used as a mold removing agent. After the tensile bars were molded, they were removed from the molds and let cool in the air at room temperature for approximately 10 min. They were then washed in cold water to remove the cooking oil residue.

Tensile testing was completed five times for each sample percent of stamp sand ASA composite material. The specimens were pulled until failure using a 10,000 lb. load cell (Model LCF455). The strain data was captured using the crosshead extension on the Universal Testing Machine. The crosshead speed used was 5 mm/min. The tests were displacement controlled.

Fracture surfaces were evaluated with a microscope (Leica MZ8) and images were captured using a Sony ILCE-5000. The average percentage area covered by sand grains was calculated using ImageJ software. This was then compared with the percentage composition by volume of sand in the composite using the density of stamp sand from the Micromeritics AccuPyc. In addition, the grain sizes on the cross section of the specimens were analyzed for the highest stamp sand density composites (40%). The size of the grains were measured using Image J and then the values were converted to millimeters using the cross section area as reference, which was measured using digital calipers.

Finally, the ability to coat the exterior surface of ASA-stamp sand composites was tested by coating the surface with acetone and then dip coating in stamp sand.

3. Results

The particle sizes of the stamp sand and starting polymers are shown in Figs. 2 and 3, respectively. Stamp sand has an approximate optical area of 0.7–0.8 mm², while the ASA pellets had an area ranging from 6.0–10.2 mm², approximately ten times larger than that of the sand. After compounding with the recyclebot extruded and cut the resultant thin column-like granules are shown in Fig. 4. These composite granules were then further processed in the injection molding system.

In low percentages (until around 40%) the sand tends to stick to the plastic pellets and remain relatively well mixed. This allowed the tensile bars to be more reliable because the mixture was homogeneous and the sand did not settle to the bottom of the molds or pool within the melted plastic.

The density of the stamp sand was measured as 2.955 g/cm³ and the well described density of ASA is about one third that of the stamp sand at ~1.060 g/cm³.

The stress vs strain curves are shown for pure ASA, 10%, 20% and 40% stamp sand in Fig. 5.

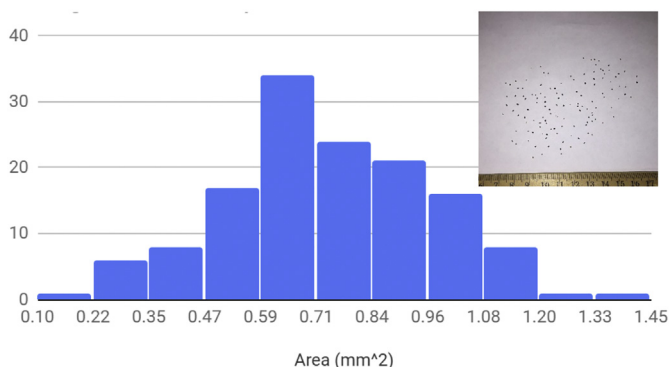


Fig. 2. Particle size distribution of stamp sand.

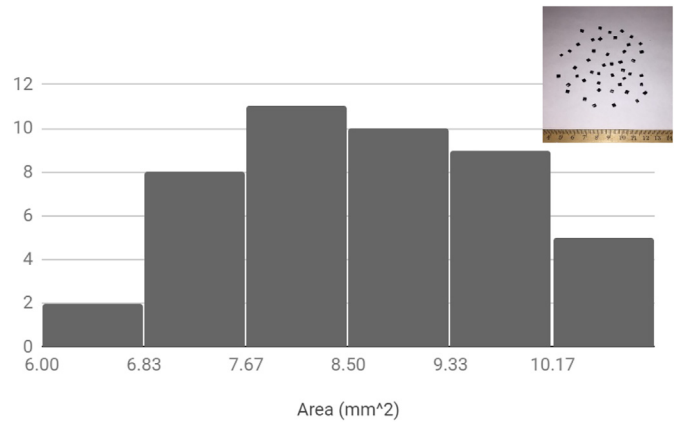


Fig. 3. Particle size distribution of ASA raw pellets.

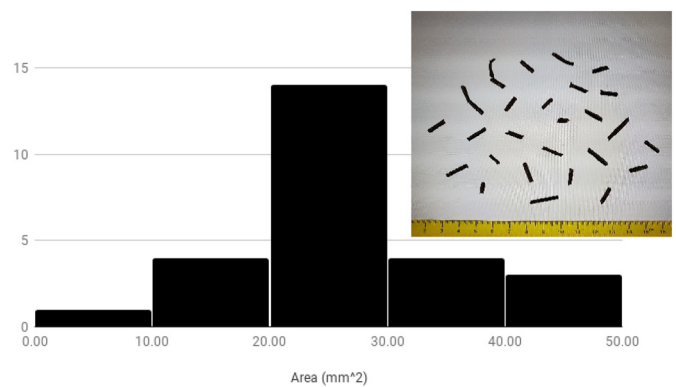


Fig. 4. Particle size distribution of pelletized stamp sand and ASA composite after recyclebot processing and cutting.

The ultimate tensile strength (UTS) is shown in Fig. 6. As can be seen for Fig. 6, the tensile strength is reduced with the inclusion of stamp sand, but then saturates as more stamp sand is added until the compounding limit is reached on the recyclebot.

Fig. 7 shows the cross section of each of the ASA stamp sand composite compounds under the microscope. The voids, which are visible for the composite cross-sections, were generally not created during the injection molding process or air voids. The voids on one part of the specimen had corresponding grains on the other part of the specimen (e.g. the voids were the size and shape of grain locations).

Fig. 8 shows the sand grain size distribution in the 40% stamp sand ASA composite. The average particle size from image J was found to be 0.325 mm² with a standard deviation of 0.208 mm².

4. Discussion

4.1. Mixing and upscaling the process

Upscaling this processing system is important to utilize the large quantities of recycled ASA and stamp sand available. ASA plastic can be granulated at the industrial scale, but also as the small and medium size enterprise (SME) scale using open source waste plastic granulators [51]. Preparation and mixing of ASA and stamp sand can be done on large scales using industrial mixers like ones used for cement. If the dry materials are mixed recently before melting, the stamp sand will not settle towards the bottom, and a uniform composite can be made based off the observations made in composites with up to 40% stamp sand.

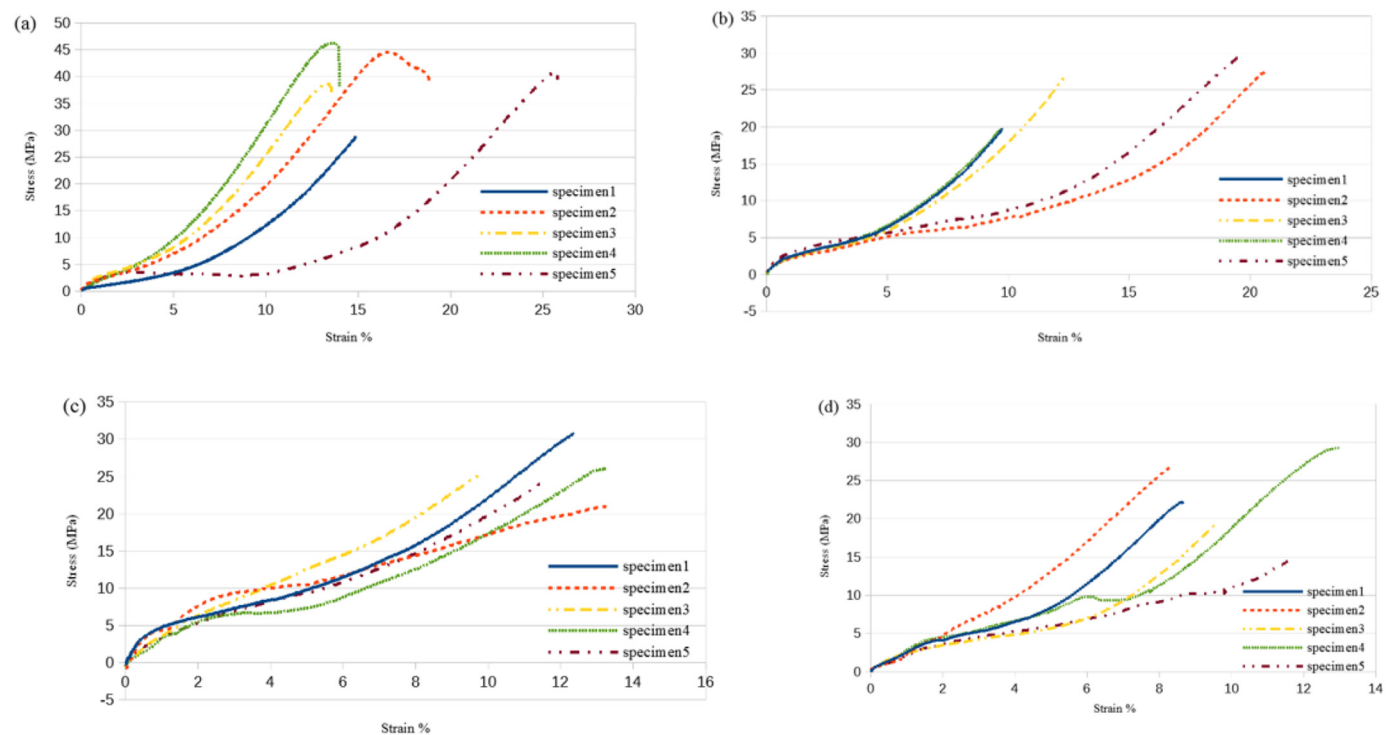


Fig. 5. Stress strain curves for each set a) 0%, b) 10%, c) 20%, and d) 40% stamp sand in ASA.

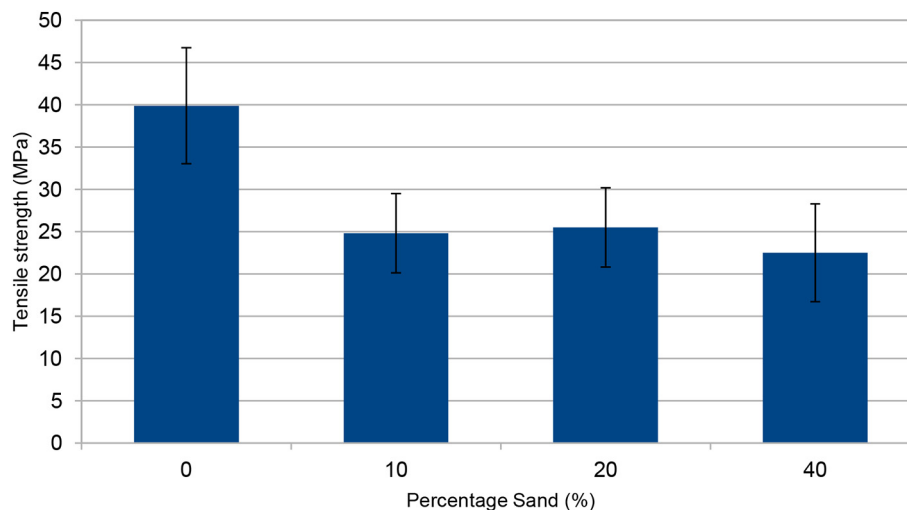


Fig. 6. UTS as a function of percentage of stamp sand in composite.

Unfortunately, industrial injection molding machines would not be able to handle the grit of the stamp sand. They are designed for pure plastics, not composites, and the stamp sand would clog or damage the mold and machines. The open source injection molding system used for this project is too small to use on a large scale, but a similar system can be designed and used to injection mold larger parts or a series of such systems can manufacture many parts in parallel.

The most efficient way to scale up production of stamp sand ASA composites would be to move towards a compression molding or temperature molding system and is left for future work. Large molds can be made out of sheet metal or 3-D printed on high temperature printers, and dry mixtures of stamp sand and ASA can be poured in and placed in an oven to melt into a solid shape. Bricks or sidewalk tiles can be produced in large quantities with enough molds and industrial ovens. This

process would greatly increase output of stamp sand and ASA composites, which would remove those waste materials from the environment at a faster pace.

4.2. Mechanical properties of stamp sand + ASA composites

The ASA stamp sand composite has about half the strength of raw recycled ASA as shown in Fig. 6. The cross-section images in Fig. 7 show that most of the specimens broke at one of the larger grains of sand. The size of the largest grains and the strength of those grains would determine the actual tensile strength of the composite. Hence, significantly more variation is observed within each sample than between the 10%, 20% and 40% samples as shown in Fig. 5. The tensile strength of the ASA and stamp sand composite can be compared

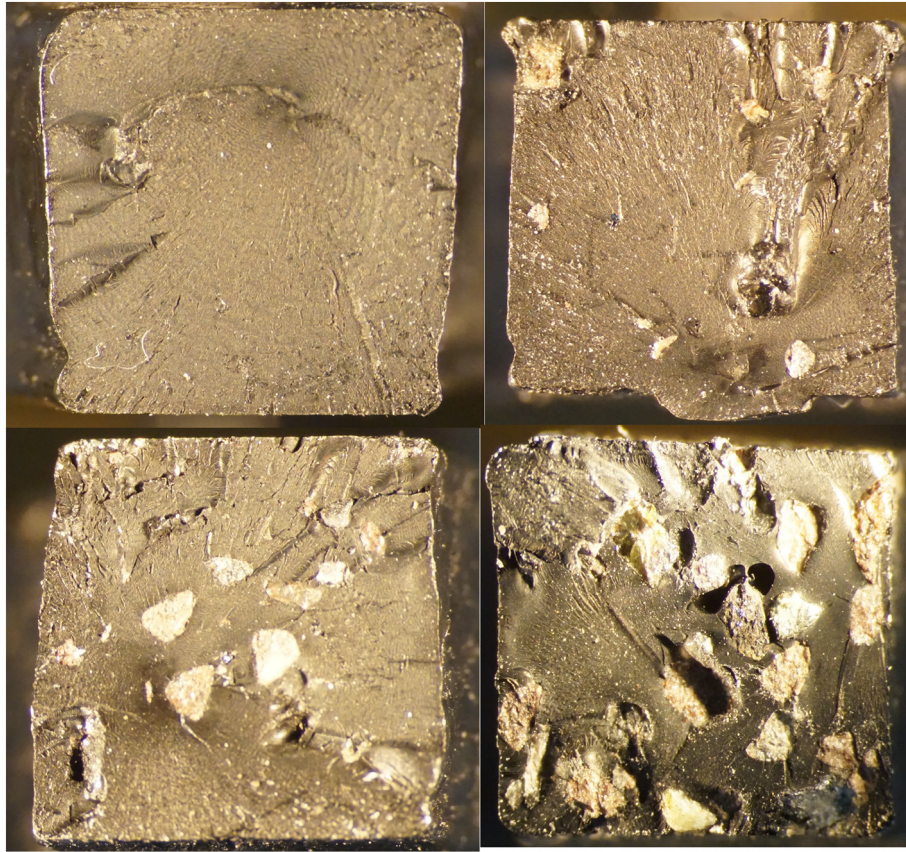


Fig. 7. Microscope image of a) 0%, b) 10%, c) 20%, and d) 40% stamp sand in ASA.

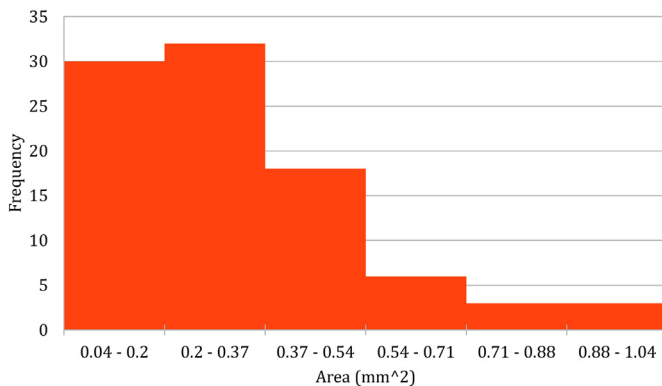


Fig. 8. Sand grain size distribution in the 40% stamp sand ASA composite after processing and cross sectioning.

favorably with AM ABS, which has a tensile strength of 25–40Mpa [52–55]. However, injection molded ABS has a tensile strength of about 47Mpa [56], which indicates that for conventional injection molded ABS products the composite may not be appropriate.

ABS is one of the most common thermopolymers used for material extrusion-based AM, which makes up the majority of the global AM market and is expected to grow to US\$ 36–41 billion/year by 2027 from US\$ 9.3 billion/year in 2018 [57,58]. In 2018 and polymeric additive manufacturing has reached nearly US\$5.5 billion and if the materials fraction of the budget grows as expected it will reach about US \$10 billion by 2027 [58]. Even though the composite developed in this study would only be an appropriate replacement for a fraction of this market, it represents a substantial method to reduce stamp-sand in

the U.P. For example, the world's first 3-D-printed car, Local Motors Strati used ABS plastic [59,60]. The reduced tensile strength of the stamp sand+ASA composite as compared to injection molded ABS may be acceptable for some applications particularly those for outdoors, which should be investigated in future work.

As expected, the percentage of stamp sand by mass (input) controlled the volume of sand observed in the composites as well as the amount of stamp sand in the cross-sectional areas at breaking as summarized in Table 1. The volume percentage was calculated using the measured density of the stamp sand of 2.95449 g/cm^3 (with a standard deviation of 0.0002 g/cm^3) and the density of ASA of 1.060 g/cm^3 . The error for the percentage area of sand was calculated using imageJ to quantify the cross section and the mass has error based on the measuring instrument used and the density is accurate to three significant figures.

The slightly lower percentages measured on the cross section (Table 1) are because of stamp sand grains that broke from the test specimens. When comparing the grain size distribution in the fractured composite (Fig. 8) from the raw material sand grain size distribution (Fig. 2), it is clear that the grain area size distribution has been reduced by processing, which could be due to the grains breaking during the manufacturing process, a preference for larger grain sizes, or due to the smaller grains accumulating in the reduced section of the specimen and the larger grains accumulating in the grip section of the specimen. This, however is of secondary importance as the number of stamp sand grains that broke within the specimen were low. Hence, the failure location was not due to a high concentration of sand grains simply the presence of some of them. Overall, the addition of some stamp sand to recycled ASA was a small enough impact on its UTS to make the composite a promising material for outdoor applications for replacing ABS particularly if it is AM manufactured ABS. ABS is a poor outdoor polymer

Table 1
Percent volume and area of stamp sand.

Percentage of Stamp Sand by Mass	10	20	40
Percent Volume of Stamp Sand in the composite	3.83	8.23	19.30
Percentage Area of Stamp Sand at the Cross Section	2.94 (SD 0.93)	6.76 (SD 2.21)	18.24 (SD 4.05)

because the surface “chalks”, which not only discolors but also reduces the impact strength dramatically due to micro-crack initiation [61–64]. Although ABS can be UV stabilized, the impact modifier in ABS is polybutadiene, which is susceptible to oxidation from the double bonds, rendering the polymer unstable [65,66]. Despite these shortcomings ABS is often used in outdoor applications with pigments that shield it against UV radiation [67].

4.3. Applications

If processes such as the one described in this study are taken advantage of, two environmentally-harmful waste materials could be removed from the environment and used to replace already necessary materials. This study has shown that distributed manufacturing methods (combining 3-D printing with small batch injection molding) could make waste ASA-stamp stand composites viable materials for a range of applications. Although the ability to 3-D print bespoke molds and then inject a waste material [43] has numerous benefits, the preliminary investigation of the waste composite indicates that the mechanical properties may also be of interest to large-scale manufacturing of conventional manufactured products. Waste ASA composites are potentially ideal for outdoor applications to begin with and adding a toxic sand would indicate that outdoor applications are also the optimal option. This is because the materials are highly likely to be toxic themselves so would be inappropriate for products to be used in food gardens, children's toys, and other products that would come in contact with humans directly or indirectly through ingestion. These applications need to be away from drinking water and food production areas (e.g. farms and food gardens). However, because of stamp sand's herbicidal properties, maintenance for many outdoor applications may be reduced or not required at all [1]. ASA stamp sand composites could be used to replace existing outdoor applications of ABS like electronic boxes or marine applications. The stamp sand ASA composite would be more resistant to outdoor weathering while at the same time the stamp sand's ability to limit biological growth could reduce cleaning and maintenance requirements. These properties of the composite need to be tested in future work. The composite could also be used for a stand in for ABS on energy conservation applications like air deflectors, which are used in transportation to reduce wind resistance for vehicles improving their fuel economy [68]. Such waste composite air deflectors would be expected to reduce cleaning. In addition, ASA-stamp stand composites could be used for traditional applications of ASA like siding material, outdoor seating, and giant outdoor chess sets [22,69]. Large volume applications are necessary to remove all of the polluting stamp sand. Thus, stamp sand - ASA composites could also be investigated to be used for non-traditional applications like making tombstones. These composites would again provide for easier maintenance and less cutting of grass directly around the tombstones would be required if the biological properties of the stamp sand are retained in the composite. In addition, the 3-D printing mold making capabilities shown here could allow for easy customization as compared to traditional stone milling used for tombstones. One possible application of an ASA plastic and stamp sand composite is production of sidewalk paving bricks. Many brick-paved pathways require maintenance to prevent weeds and grasses from growing in the cracks between bricks. These

types of applications are necessary in order to drive the large market needed to eliminate stamp sand waste repositories in the upper peninsula of Michigan from further polluting Lake Superior.

Future work is needed to evaluate the technical, economic and environmental potential for waste ASA - stamp sand composites for both applications of ABS material replacement as well as for ASA applications. New tests are needed to evaluate the compressive strength as well as impact resistance of these materials. The latter is necessary as the fracture surface shown in Fig. 6 displays little ductility, and thus the impact strength may limit applications. To gain a better understanding of the mechanical properties future work could probe the distribution of sand within the polymer using computed tomography. In addition, the anti-biological properties of the material composite need to be evaluated if the composite should be thought of as means to safely encapsulate stamp sand filler (e.g. a way to make ASA more dense for specific applications) or if the anti-biological properties would reduce maintenance for outdoor applications by removing the need to clean algae, biofilms, etc.

5. Conclusions

This study found potential to combine the two harmful waste materials of stamp sand and waste ASA into a useful material, which can be injection molded into 3-D printed PC molds allowing for distributed manufacturing of custom products. Stamp sand was able to be added to waste ASA up to 40% by mass using a single auger recyclebot system for compounding. The ultimate tensile strength of the resultant composites reduced about half the strength of raw recycled ASA after even 10% sand. However, this strength reduction plateaus and the tensile strength of the ASA and stamp sand composites can be compared favorably with ABS. This makes waste ASA- stamp sand composites potential replacements for outdoor applications of ABS as well as some current ASA applications. These results are promising and call for future work to evaluate the technical, economic and environmental potential for waste ASA - stamp sand composites.

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.

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