

Particulate Mollusc Shells as Reinforcements for Aluminium Matrix Composites: A Review

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Received: 26.01.2021

Accepted: 06.05.2021

Published: 30.06.2021

Abstract: In recent times, the global need for high-performance, affordable and topnotch materials has necessitated a shift in research from unreinforced to composite materials. Metal matrix composites (MMCs) are relatively novel and highly promising materials that continually attract researchers' attention. They are constantly being developed, and their applications in various industries are on the increase. Aluminium matrix composites (AMCs) are the most commonly studied MMCs because of their many desirable properties, especially high strength and lightweight. These properties make them find extended use commercially. Also, AMCs can be manufactured via cost-effective and straightforward methods. However, the conventional ceramic reinforcements commonly used to reinforce aluminium matrix pose high costs and limited availability. This has triggered other renewable and cheap materials such as agro and industrial wastes as alternatives for reinforcing aluminium alloys. This work attempts to review some particulate mollusc shells that have been used as reinforcements for the fabrication of AMCs. The processes used for the fabrication of AMCs are briefly discussed. Finally, further research for the improvement of AMCs is suggested.

Keywords: composites, mollusc shells, aluminium, reinforcements

1. Introduction

Materials such as steel, aluminium, copper, cast iron, and many more have, over the years, been of high interest in the world of engineering. These materials find extensive use in automotive, aerospace, chemical processing, military, petroleum processing, biomedical, sports. And many other industries where they are used to suit different purposes. However, the global demand for reduced weight, top quality, high performance, and

low-cost materials is causing a shift of focus from these unreinforced materials to composite materials [1-4].

The new era engineering systems demand materials with a wide spectrum of properties, which are rarely met using unreinforced materials [5]. Also, the present reality regarding human material demands has necessitated industries to use materials of a better class that are sustainable and environmentally friendly [6]. Metal matrix

composites (MMCs) have been discovered to offer uniquely combined properties required in a wide range of engineering applications [5,7]. High specific strength, high thermal resistance, good damping capacities, improved wear resistance, high specific stiffness, and good corrosion resistance are some of the properties offered by MMCs [8-10]. Due to these much-improved properties, MMCs that were hitherto predominantly used in aerospace and automobile industries are now fast replacing metals. And their alloys in the vast majority of other applications, including defense, sports, marine, and recreation industries [11].

MMCs can be defined as metallic alloys reinforced mostly with hard ceramic material particles to produce desired specifications. Alloys of light metals like Aluminum (Al), Magnesium (Mg), and Titanium (Ti) are commonly used as based metals in the production of MMCs. However, other metals like copper (Cu) and Zinc (Zn) have also been used [12-16]. Despite having some drawbacks in their mechanical properties, aluminum and its alloys have been the most widely utilized as matrix materials in the fabrication of MMCs amongst the competing alternatives [8,11,13,17].

Aluminium matrix composites (AMCs) have a unique combination of physical, chemical, and mechanical properties. This property is rarely obtained when unreinforced materials are used to compete strongly with steel and other alloys in various engineering applications [18-21]. Also, AMCs have other advantages such as low cost of processing and being complaint to production using the techniques for processing monolithic metallic alloys [22,23].

Presently, mollusc farming on a commercial scale is an essential component of the aquaculture industry globally. It represents about 23% of the world's total aquaculture production. It is growing gradually because of its small investment requirement and relatively low energy consumption. Molluscs are sources

of cheap and healthy food for the growing world population [24]. Shells of some molluscs can amount to up to 75% of their total body weight. An estimated seven million tons of mollusc shells are disposed of yearly. These shells are usually dumped into public waters and landfills, making them pose some environmental discomfort [25]. Using these shells to reinforce metals will solve environmental problems and serve as a cheap alternative to synthetic reinforcements characterized by high cost and limited supply [26]. In this work, an effort has been made to review some particulate mollusc shells used as reinforcements for the fabrication of AMCs. However, they influence some properties in the composites

2 Materials used to Reinforce AMCs

Materials used to reinforce metal matrix play essential roles because they determine the fabrication methods used for the syntheses of AMCs and the nature of the alloy matrix being reinforced [12, 27-29]. Likewise, most parameters such as reinforcement type, shape, size, modulus of elasticity, hardness, and distribution in the matrix are strongly linked to reinforcing materials [30]

Considering the literature studied while preparing this review, different reinforcing materials that have been used in the development of AMCs can be classified into three broad categories. These are synthetic ceramic particulates, industrial wastes, and agro-waste derivatives [30]. Silicon carbide, silicon nitride, silica, aluminium oxide (alumina), titanium carbide, titanium nitride, graphite, zirconium, boron carbide, tungsten carbide, carbon nanotubes are some of the synthetic ceramic particulates that have been widely studied [31,32]. Nonetheless, silicon carbide and alumina have been the most utilized compared to other synthetic reinforcing particulates [32]. Fly ash and red mud, which are the waste products of power plants and aluminium industries, are the

industrial wastes that have been investigated by various authors [33]. Among many other agro-wastes, bamboo leaf ash (BLA), rice husk ash (RHA), sugarcane bagasse ash (SCBA), Maize stalk ash (MSA), groundnut shell ash (GSA), and coconut shell ash have been used respectively by [34],[35],[36],[37],[38], and [39] to reinforce various aluminium alloys. Seashell wastes that are products of aquaculture or mariculture in recent times are fast gaining interest in the use as reinforcement for aluminium matrix. They are the major focus of this review.

3 Using Particulate Mollusk Shells as Reinforcement phase

Particulate mollusc shells have over the years been used to serve different reinforcement purposes. Various authors have widely studied their reinforcement ability, most especially in concrete and polymers. Mollusc shells are materials that offer advantages of modulus improvement and cost reduction without notably increasing the specific gravity of the composites when compared with the commonly used inorganic fillers [40].

Many authors, including [41],[42],[43], and [44], have used mollusc powder to reinforce polymers such as polypropylene and polyester, and they all affirmed improved mechanical properties. Efforts have been made to use mollusc shells in the reinforcement of AMCs. Based on the published articles studied, the most investigated mollusc shells in aluminium matrixes are those of land snails, periwinkles, mussels, and oysters which are considered in this review

3.1 Snail Shells

The main constituent of the shell is calcium carbonates which can be one of two crystalline forms; calcite or aragonite. The remainder is organic matrixes that constitute a protein known as conchiolin, which usually makes up to 5% of the shell [45-47]. Snail shells are

usually present as discarded bio-waste remnants of restaurants, households, eateries, and snail sellers. They constitute environmental threats with little or no economic significance [48,49]. As observed by [49], the Snail shell has about 9.4-25.9 lesser density compared with other agro or industrial reinforcement materials such as fly ash, coconut shell ash, bagasse, and maize husk.

Kolawole et al. [49] have studied the potentials of snail shells as a low-cost reinforcement material in AMCs utilizing a characterization technique. The mineralogical composition and physical properties of particulate snail shells were carried out using density determination. And the thermogravimetric analysis (TGA), refractoriness, dispersive energy X-ray (SEM/EDX), X-ray fluorescent (XRF). The X-ray diffraction (XRD) analyses at 0°C, 800°C, 850°C and 900°C calcination temperatures for 3hrs. The results obtained affirmed that the particulate snail shell had chemical hard phase oxides such as Al₂O₃, Fe₂O₃, SiO₂, CaO, Cr₂O₃, MnO, and NiO. Therefore, at all calcination temperature values with the maximum amount of these phases formed at a 900°C calcination temperature. The XRD analysis indicated the presence of lime (Ca₄O₄), calcite (Ca₆C₆O₈), and portlandite (CaO₂H₂) as major hard phases of the particulate snail shells at 900°C calcination temperature. The study also indicated that the density and refractoriness temperature of the snail shell particles are 1.63 g/cm³ and 1400°C, respectively. The result of the TGA showed that the particulate snail shells attained their thermal stability at 840°C. It was concluded from the results that particulate snail shell was a promising reinforcing material in the production of lightweight metal matrix composites at low costs. Additionally, the high refractoriness temperature of the snail shell particle indicated that it was a suitable reinforcement material in the production of MMCs that are stable at elevated temperatures. And could find use in

automotive components such as pistons connecting rods.

Asafa et al. [50] experimentally studied the potentials of particulate snail shells as reinforcement for discarded aluminum-based materials. Particulate snail shells of 16 to 48 wt% and 200, 400, and 600 μm were added to discarded aluminium pistons using a double stir-casting method. After casting, a microstructural study was carried out on the resultant composites using a metallurgical microscope. Also, the hardness and tensile strength of the composites were examined. The microstructure of the unreinforced sample indicated significant pores of different sizes and shapes. The reinforced samples, however, showed snail shell particles uniformly distributed in the aluminium matrix. The sample containing 48 wt% and 600 μm particle size gave the maximum tensile strength and hardness with the values of 236 MPa and 48.3 HRF, respectively. In contrast, the unreinforced sample gave 92.4 MPa in tensile strength and hardness of 29.2 HRF. The notable increase in tensile strength and hardness was attributed to the uniform distribution of snail shells in the aluminium matrix.

Olawuni et al. [51], to reduce the cost of reinforcement and minimize waste disposal challenges, used discarded aluminium piston reinforced with alumina and snail shells to produce hybrid composite piston material. Discarded motorcycle pistons were melted and then reinforced with alumina and snail shells at different proportions using the D-optimal approach of the Design of Experiment. The microstructure is shown in Figure 1, and elemental composition was determined using a Scanning Electron Microscope (SEM). Mechanical tests such as tensile strength, hardness, and wear were conducted. A corrosion test was also done. The results indicated that hardness, yield point, and tensile strength increased with increased snail shell particles to aluminium alloy. Also, wear loss was greatly minimized as a result of improved mechanical properties. However, the corrosion rate of the composite was higher than that of unreinforced alloy. The reason for this was not stated. It was concluded that alumina and snail shells improved aluminum pistons' mechanical properties, which can be helpful in various engineering applications.

Table1. Chemical Composition of Snail shell ash obtained at 800°C [52]

Constituent	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	TiO ₂	Na ₂ O	P ₂ O ₅	MnO ₃	K ₂ O
wt%	10.20	4.81	3.15	61.95	0.18	0.03	0.05	0.04	0.01	0.01	0.01

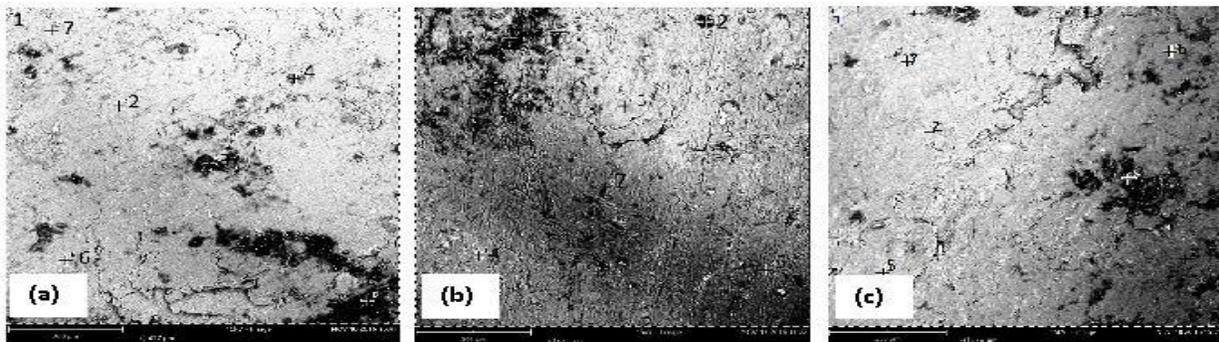


Figure1. SEM Micrographs of (a) Aluminium alloy + 10% Al₂O₃ + 10% snail shell (b) Aluminium alloy + 5% Al₂O₃ + 20% Snail shell (c) Unreinforced Aluminium alloy. Source [51]

3.2 Periwinkle Shells

Periwinkle shells are hard, generally light, and readily available in sizeable quantities. They are also inexpensive [53]. Umunakwe et al. [54] have accessed some mechanical properties and microstructure of particulate periwinkle shell-aluminum 6063 metal matrix composite (PPS-AIMMC) produced through a two-step casting route. After being washed and boiled in water for 40 minutes at 100°C, Periwinkle shells were subsequently sundried. They were further dried in an oven at 110°C for 30mins to remove all moistures. The shells were crushed, pulverized, and sieved to 75µm and 150µm particle sizes. A software-driven optical metallurgical microscope was used to study the microstructure of the specimens. The tensile and hardness tests were also conducted. The microstructural examination results indicated that particulate periwinkle shells distributed uniformly in AA6063 alloy and refined grains from coarse grains to fine grains at smaller particle sizes. Consequently, due to the ability of particulate periwinkle shells to refine AA6063 alloy grains, the strength and hardness of the composites were improved when compared with the unreinforced alloy. It was deduced that the composites could be used in areas where lighter weight and higher strength were required.

The effect of isothermal heat treatment on the hardness and microstructure of aluminum-periwinkle shell ash biocomposite was experimentally studied by Umaru et al. [55]. Using a stir casting method, the aluminium alloy was reinforced with periwinkle shell ash (PSA) from 5% - 25% in step 5. The composites were subsequently heat treated. It was revealed that periwinkle shell ash distributed homogenously in some aluminium specimens, as shown in Figure 2. These composites had better thermal conductivity and hardness compared to the unreinforced specimen.

The density and mechanical properties of particulate periwinkle shell aluminium 6063 metal matrix composite (PPS-AIMMC) were also investigated by Umunakwe et al. [56]. Particulate periwinkle shell sizes of 75µm and 150µm were used to produce the composite at 1,5,10 and 15wt% using two-step casting. It was observed that the filler was distributed uniformly in the aluminium matrix. The composites had lower density, improved strength, ductility, and hardness. The better mechanical properties were ascribed to the ability of periwinkle shell ash particles to refine the grains of Al6063. It was concluded that the composites could find use in the areas where lighter weight and higher strength were required.

Table 2. Chemical Composition of periwinkle shell ash obtained at 800°C[52]

Constituent	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	TiO ₂	Na ₂ O	P ₂ O ₅	MnO ₃	K ₂ O
wt%	26.26	8.79	4.82	55.53	0.4	0.18	0.20	0.25	0.05	0.05	0.07

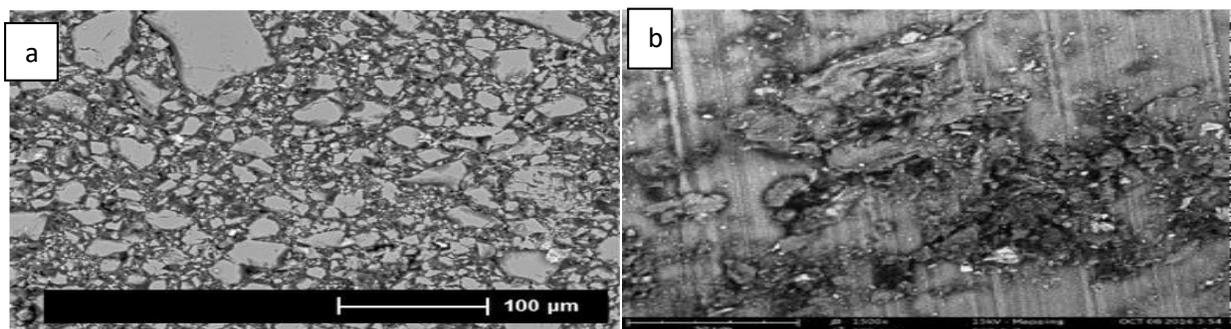


Figure 2: SEM micrograph of (a) unreinforced Al (b) Al/25%PSA. Source [55].

3.3 Mussel Shells

In 2016 alone, an estimated 1827.27 metric tonnes of green mussels were produced worldwide, and it has been increasing yearly. About 33% of mussels are made up of shells, which are usually discarded. With that, they also contribute sizably to waste generation. [57,58]

Surendra et al. [59] have fabricated composites using Al6061 reinforced with mussel shell particulate with the primary aim of studying the dry sliding wear of the composites. 2, 4, 6, 8, and 10 weight percent (wt%) of mussel shell particulate was used to reinforce the aluminium matrix. The microstructural study revealed that the particles distributed fairly in the matrix, with 6 wt% having the maximum particle filling capacity. Also, mussel shell particulates progressively increased the overall hardness and wear resistance of the composites up to 6 wt%, demonstrating the best properties. The increase in hardness was attributed to differences in the thermal properties of the reinforcement and matrix phase. This mismatch led to an increase in dislocation densities during solidification.

3.4 Oyster Shells

The corrosion behaviour of Aluminium alloy 6063-Seashell (oyster shell) composites in acidic and alkaline environments was studied by Abdulkareem et al. [60]. The composites were fabricated by varying particulate oyster reinforcements in steps of 1.5 wt% from 1.5 - 7.5 wt % and particle size of 100, 150, and 200 μm in the aluminium 6063 alloy matrix double stir casting method. The static immersion weight loss technique was used to determine the corrosion rate with the weights measured at 72 hours intervals for 18 days in line with the ASTM G31-12 standard. The acidic and alkaline media used were Sulphuric acid (H_2SO_4) and Sodium hydroxide (NaOH), respectively. The results indicated that oyster reinforcement relatively improved the corrosion resistance of the composites, with the sample containing 6.0 wt% and 100 μm particle size. The result shows the best corrosion property in both acidic and alkaline environments. However, all the composites exhibited better corrosion characteristics in the alkaline medium than the acidic medium. The reason for this occurrence was not stated.

Table 3. Chemical Composition of Oyster Shell Ash Obtained at 800°C [52]

Constituent	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	TiO ₂	Na ₂ O	P ₂ O ₅	MnO ₃	K ₂ O
wt%	13.41	4.95	3.80	57.95	0.19	0.12	0.02	0.22	0.01	0.01	0.01

4. Fabrication Methods for Aluminium Matrix Composites

Various methods have been used to manufacture AMCs. These methods are broadly classified based on the temperature of the metallic matrix while processing [61]. They are solid-state methods and liquid state methods [30,62]. In liquid state routes, the particulates are distributed mechanically over the liquid metal before casting and solidification. These routes are particularly cost-effective [63]. Liquid state routes include stir casting, compocasting, squeeze casting, spray forming, In-Situ Synthesis, and liquid metal infiltration [61].

For liquid state routes of manufacturing AMCs, the stir casting method has been the most investigated. This is because of its simplicity, being commercially viable, and flexibility [64,65]. Nonetheless, it is not without its downsides, including problems of homogenous distribution of reinforcing particles, wettability, porosity, clustering of particles, segregation, interfacial reactions, and the formation of detrimental secondary phases. Methods to contain these challenges have been reported [30]. To solve the wettability between reinforcing particles and aluminium matrix, reinforcement coatings and wetting agents such as K_2TiF_6 are used [66,67]. Clustering and

particle segregation can be avoided by optimizing mixing parameters, including rotation of stirrer, stirring speed, and blade angle to stirrer axis [68]. Formation of secondary phases and interfacial reactions, which are of high detriment to the performance of the composites during the development process. It can be prevented by selecting reinforcing materials that do not undergo interfacial reactions, such as boron carbide and alumina [69]. Employing cold deformation and hot isostatic pressing, and porosity can be substantially reduced in cast AMCs [70,71].

Solid route fabrication includes powder metallurgy (PM), high-energy ball milling, diffusion bonding, and ultrasonic probe assisted method [62]. The fabrication of MMCs via PM has been widely reported. It involves mixing reinforcements uniformly with metal alloy, and the powders are blended. The product, after solidification in a vacuum, is sintered under high temperatures [62]. Many researchers have adopted PM to eliminate the defects encountered in MMCs prepared through conventional methods like stir casting, squeeze casting, and other liquid molten metal techniques. Through this, the mechanical properties of the composites have been increased. It is also one of the much-improved ways to prepare composites and nanocomposites [72].

5. Conclusions and Recommendations

This study has reviewed various works on mollusc shells reinforced metal matrix composites, and the following conclusions have been drawn:

- (i) Mollusk shells available in sizeable quantity, are usually considered wastes with little or no economic value.
- (ii) Particulate mollusc shells at calcination temperatures produced chemicals hard phases oxides such as Fe_2O_3 , CaO , Al_2O_3 , and SiO_2 , which improved the composites'

mechanical properties and made them attain stability at elevated temperatures. This has made them highly promising for the production of AMC's at reduced costs.

- (iii) Mollusc shells can distribute uniformly in the aluminium matrix, with the ability to refine matrix materials from coarse grains to fine grains.

It is recommended that further works should be done to determine in details:

- (i) The corrosion behaviours of the mollusc shells reinforced AMCs, especially under different conditions since there were conflicting corrosion results from the reviewed articles.
- (ii) Further studies also should be focused on how to optimize the production process to ascertain the optimum processing parameters for mollusc shells reinforced AMCs

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