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Review Article

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Applications of Green Composites in Immobilization of Radioactive Wastes and Others- A Review

Saleh HM* and Bayoumi TA

Abstract

Environmental compatibility of green composites has become an important characteristic as the need to reduce environmental hazards is increasing worldwide.

Mechanical development, which replaces the non-sustainably based materials by biomass-based ones would be the assignment forced on researchers and designers of the twenty-first century, since it is foreseen that fossil assets will vanish sooner rather than later. It is likewise viewed as that this advancement incorporates the change and improvement of biomass-based composites

Keywords

Green composites; Immobilization; Radioactive Wastes; Biomass; Polymer; Cement

Introduction

Composite materials are materials composed of two or more components of significant different chemical or physical characteristics which combined to each other producing another material with new properties defer from the original constituents. The individual components remain separate and distinct within the finished structure.

1. Characteristic engineered composite materials include:
2. Composite materials such as cement and concrete.
3. Reinforced polymer e.g. fiber reinforced plastics.
4. Metal composites
5. Ceramic composites

The obtained composite may be preferred for:

1. Improved quality to satisfy the increase needs for safe and durable structure.
2. Requirement of sustainable develop (economy of raw materials, energy and space, limitation on emission of gases and wastes.

Many industries are responsible for the depletion of large amounts of non-renewable resources. These activities generate not only millions of tons of various wastes but also, millions of tons CO_2 gas emission. Therefore, research about find materials based on renewable resources, like vegetable fibers are needed. Among this is the cement composite industry.

Bio composite is a combination of cement or polymer matrix with organic fibers obtained from agro and forest resources either as a fiber crop or residues.

Green composites are a particular class of bio composites, where a bio-based polymer lattice is strengthened by common strands, and they speak to a developing range in polymer science. New patterns in the choice of regular filaments, that is, from squander instead of from profitable yields were accounted for

Classification of natural fibers

In this context the green composites can be also defined as composites that are designed with the lowest environmental 'footprint' possible (Figure 1)

Most of resins and fibers used in the green composites are biodegradable, when they dumped, decomposed by the action of microorganisms. They are converted into the form of H_2O and CO_2. These H_2O and CO_2 are absorbed into the plant systems.

The two main components of the green composites include:

1. Biodegradable resin
2. Natural fibers

E.g. environment-friendly green composites were fabricated from a starch-based, dispersion-type biodegradable resin and cellulose fibers [1].

According to other published text, the "green composite" is a term that indicates that the composite as a whole (both matrix and reinforcement) originates from renewable resources [2].

Selection of a suitable fiber, among those available in nature, as reinforcement for a given polymer is guided by the values of composite stiffness and strength required.

Besides intrinsic properties of each component (fibers and matrix), the mechanical properties of the composite are expected to depend on fiber aspect ratio, volume fraction and orientation, and on adhesion at the fiber-matrix interface [3].

Overall, the use of fibers from waste (either agricultural by-products or avian feathers) as reinforcement in biocomposites offers a low cost and environmentally friendly solution to waste disposal and a possibility for farmers or poultry producers to gain a profit from waste.

Cement-composites: The use of vegetable fibers as reinforcement in cement based material is an add/newly developed topic. Application of plant-based natural fibers into cement concrete had also been reported by several Indian institutes [4,5].

Various plant fibers are used for production of cement, biocomposites e.g. cellulose fiber, lignocellulose waste, sugar can bagasse, wheat and eucalyptus, coconut fiber or shell, waste tea leaves or processed waste tea [6-15].

***Corresponding author:** Hosam M Saleh, Radioisotope Department, Nuclear Research Center, Atomic Energy Authority, Dokki, Giza, Egypt, Tel: +2 01005191018; E-mail: hosamsaleh70@yahoo.com

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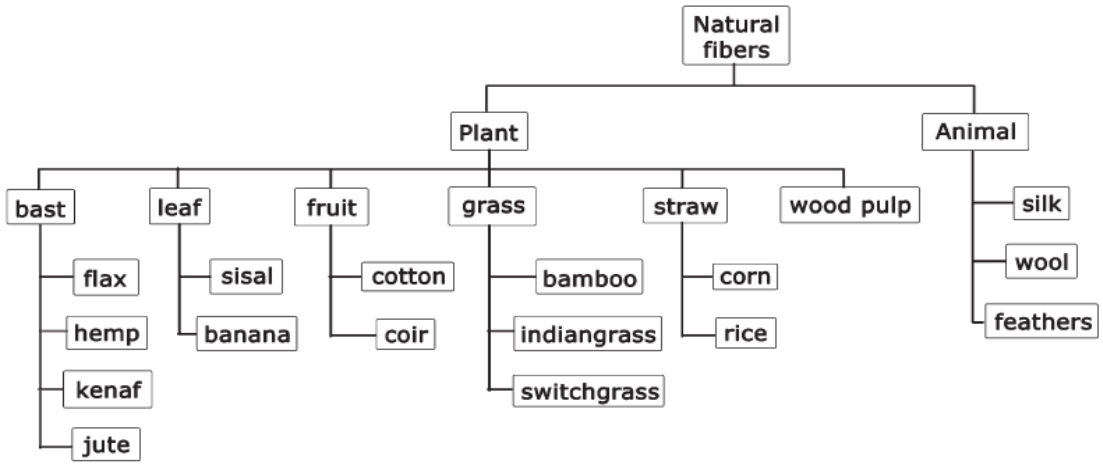

Figure 1: Classification of natural fibers.

In this concept, most or may be all of those composites are used as construction materials. On the other hand, large number of non-biocomposites were used and tested for incorporation of radioactive wastes. However, extending the definition of green composite for solidification/stabilization of radioactive wastes, the following laboratory scale applications can be summarized. At the end of bioremediation process for liquid waste using either fungi or plant, the generated bioproducts were incorporated into cement and the resulting products were subjected for various evaluations [16-24]. Hence, we can consider this application as the application of green composite in the field of radioactive waste.

Environmental sustainability: Few studies are available in the literature concerning the environmental advantages associated with the substitution of glass fibers with natural fibers that were reviewed in an early study by Joshi et al. [25]. An interesting result in this area concerns the cumulated energy demand that, in the case of hemp fiber production for example was shown to be only 10% of that required for production of glass fibers [26].

Industrial applications of green composites

With concern to the industrial applications, several paths have been undertaken. In short, it can be stated that the most used natural-organic filler is wood (either flour or fibers), especially as low cost filler for polyolefin [27]. Wood flour is usually obtained from sawmill waste after a simple sieving treatment; wood fibers are produced from sawmill waste by a wet thermo mechanical process. Already explored industrial applications include window and door frames, furniture, railroad sleepers, automotive panels and upholstery, gardening items, packaging, shelves and in general those applications which do not require very high mechanical resistance but, instead, low purchasing and maintenance costs [27].

Moreover, it is conceivable and advantageous to utilize reused polymers set up of virgin ones, in this manner guaranteeing enhanced cost-productivity and eco-manageability. Some examples of industrial applications can be easily found on the technical literature and on the Internet; these include, for instance, indoor furniture panels, footboards and platforms, automotive panels and upholstery, noise insulating panels, etc., mainly produced by American, German, Japanese, British and Italian firms [28]. They have started to utilize polymer composites with natural-organic fillers as materials for door panels, roof upholstery, headrests, parcel shelves, etc. Depending on the applications, it was sometimes necessary to improve the mechanical properties through fiber pre-treatment (acetylation, use of MAgPP, etc.), and the treated fibers were then used in several ways, in order obtaining mats, non-woven structures, etc.

Some authors assert also that, by means of special treatments on natural fibers, these could lead to the production of high-quality composites with mechanical properties comparable to glass fiber filled composites [28].

Green rubber composites: Green elastic composites can be customized made to suit applications with wanted properties by consolidating particulate characteristic strands into an elastic network. The rubber green composite compounding was prepared by using two roll mills and then molded by hot compression molding technique. The resulted suggested that increasing filler content tended to increased modulus and hardness but decreased Tensile strength of the composite [29].

Natural fiber used in green composites: It is surprising that common filaments, for example, kenaf, flax, jute, hemp, and sisal have pulled in restored intrigue, particularly as a fiber substitute in the car business. The upsides of common fiber over engineered are minimal effort, low thickness and adequate particular quality properties, simplicity of detachment, carbon dioxide sequestration, and biodegradability. Plastics are lighter however they are not fit for stack bearing application as a result of the absence of quality, solidness, and dimensional strength. In fiber-reinforced composites, the fiber serves reinforcements by giving strength and stiffness to the composite structure [1].

The utilization of characteristic strands for specialized composite application has been the subject of escalated explore in both Europe and USA. For the most part car segments are created from characteristic strands like flax, hemp or sisal reinforced with PP or PE. The appropriation of characteristic fiber composites in this industry is driven by value, weight sparing and showcasing contemplations instead of by specialized requests. Normal strands are presently being used economically in blend with PP in Biocomposites for car applications [30].

Composites are used in a wide range of applications and contribute to the growth of various industries. The composite industry value chain spans a full range of activates, from the preparation of raw materials from natural fibers and binder production to the manufacture of end products. Biocomposite products need to be further developed, as a long-term strategy to develop the tremendous wealth of natural fibers that are currently under-utilized.

Classification of biocomposites

In this section, we try to clarify where biocomposites are positioned among the whole composite materials. In the previous section, the importance of green composite studies was described, and related to the biocomposite-engineering field. The combination of natural fibers and biomass-derived biodegradable resin is common to both biocomposites and green composites. What is the difference between biocomposites and green composites?

The green composites are not necessarily a subset of biocomposites, but consist of the intersection of biocomposites and a disjoint part.

From such a point of view, we can classify biocomposites and green composites, as shown in Figure 2.

According to the International Atomic Energy Agency (IAEA), radioactive waste can be defined as "material that contains or is contaminated with radionuclides at concentrations or radioactivity levels greater than clearance levels established by the appropriate authority and for which no use is foreseen" [31]. The generated radioactive wastes are varied in form, activity and type of contamination as they are in type of generating activity. It might be strong, fluid or vaporous. Inside these gatherings are an assortment of waste sorts, for example, junk, spent radioactive sources, pumps, funnels, particle trade pitches, slop, and spent atomic fuel. Action levels go from to a great degree abnormal states related with spent fuel and deposits from fuel reprocessing to low levels related with squander from radioisotope applications in research centers, doctor's facilities, and so on. Similarly expansive is the range of half-existences of the radionuclides contained in the radioactive waste. Which radionuclides are available will rely upon the creating procedure; they may incorporate uranium and other normally happening, transuranic and additionally particular man-made radionuclides [32].

Due to its good characteristics such as early compressive strength, durability, fire resistance, immobilizing hazardous ingredients, the applications of green cement as a matrix and natural material as a filling component in special fields such as solidification/immobilization of heavy metals and building repairing have been investigated.

The British standards institutions defines cement as a hydraulic binder containing finely grinded inorganic materials, which form with water a paste that sets and hardens by means of hydration reactions [33].

Plant fiber-mortar composite: A proposed mortar-degraded spinney waste composite was developed and based on Ordinary Portland Cement (OPC) and sand that hydrated with slurry originated from the wet oxidative degradation of fibre wastes collected after the spinning industrialization of the raw cotton. Cured cylindrical mortar-composite blocks, at weight ratios cement: slurry 0.7 and cement: sand 1:3, were prepared and left to hard for 28 days. At the end of curing time the mass and compression of the control sample were recorded prior to testing. Sets of the prepared solid blocks, each of sis were immersed in various leachant solutions in well tight closed containers under static conditions. They were subjected to three chemical agents, namely: 0.1 N sodium hydroxide, 9% sodium chloride and 0.1N sulphuric acid, for different immersion times (7, 30, 84 and 360 days).

The long-term durability of those materials under the imposed environmental conditions is a key factor controlling whether the products have been confidently proven.

ACI committee has classified chemical attack into: acidic attack, carbonation, alkali attack, chloride leaching, and/or sulphate attack [34]. It can be accepted as general rule that acids are more deleterious to cementitous products. The deterioration in these products may be attributed to the reaction of the attacking ions' species (e.g. Cl^-, NO_3^-, SO_4^{2-}, SO_3^{2-}, HCO_3^-/CO_3^{2-}, OH^-,....) with the free lime, $Ca(OH)_2$, formed during the cement hydration, and in the long run it ultimately leaches out in the form of soluble calcium salt leaving behind holes in the products. These holes spread widely and weaken the whole structure of the products [35].

Mortar-spinney waste fiber composite under immersion in sulphuric acid: Sulphuric acid, like other mineral acids is very dangerous for cement-based materials. The deleterious action of sulphuric acid can be attributed to the formation of two voluminous reaction products namely, calcium sulphate dehydrate (gypsum) and calcium sulphoaluminate (ettringite). The severity of the sulphuric acid attack, in addition to a dissociation ability of the acid, is significantly dependent on the solubility of calcium salt formed.

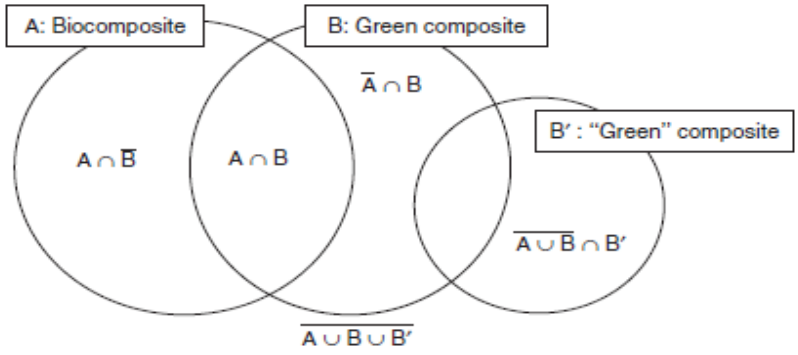

Figure 2: Classification of Biocomposites and green composites.

Table 1 represents the compressive strength and mass loss values for mortar composite blocks subjected to 0.1 N sulphuric acid for increasing exposure time.

Mortar blocks immersed in sulphuric acid were suffered from the attack of sulphate ions and dissolution effect caused by hydrogen ions, therefore, the deterious reaction, can be distinguished to two processes [36]. The first, deterioration of Ca $(OH)_2$ resulted from the hydration reaction of OPC with expansive gypsum formation. The second where the formed gypsum reacts with C_3A in an aqueous environment and forms a more expansive product "ettringite". The formation of these two materials (i.e. gypsum and ettringite) is accompanied with detectable diminishes in compressive strength integrity (Table 1). It is essential to notify that, even after one year of exposure to 0.1 N sulphuric acid yet the mortar-composite still has a mechanical integrity greater than recommended. Mortar cement composites are readily capable of achieves compressive strength of 5.45 N/mm^2 at the end of 48-week exposures to 0.1N H_2SO_4. This value still exceeded the minimum strength criteria (≈3.33N/mm^2) recommended by NRC [37].

The deterioration in the compressive strength data conceded with the increase in mass loss values for mortar- composite blocks immersed in 0.1N sulphuric acid as shown in Table 1. This may explain on the basis that, sulphuric acid decomposes cementitious matrix by decalcifying portlandite (CH) and calcium silicate hydrate (C-S-H), thus contributing to strength loss and increase in mass loss [35].

It is clear from the data represented in Table 1 that there were relative small decreases in porosity percent values up to 4 weeks, and then it was actuated to increase by extending the exposure periods. These small changes in porosity values are due to the reaction of sulphuric acid with portlandite, formed during hydration of the mortar-composite, which gives rise to the formation of less soluble or almost insoluble calcium salts (e.g. $CaSO_4$) that remain in the corroded layer and slightly increase its resistance to leachant diffusion.

However, by increasing the exposure of the mortar products to sulphuric acid accompanied with an extensive formation of gypsum that not providing any protective aid to these blocks and also tends to cause their expansion. Consequently, build up high internal stress that ultimately leads to spalling and exposure of the successive fresh surface layers of the mortar blocks to the acid attacks.

Formation of insoluble gypsum layer at early time attenuated the water absorbed by the mortar- composite specimens, later on; this gypsum layer was loosely connected to the block and fell off spontaneously, Table 1. These imply that the internal microstructure became porous and absorbs more water [38].

Integrity of composite under immersion in sodium chloride solution: Apart from climatic conditions that produce degradation, especially by freeze/thaw actions, the other elements that play an important role in degradation of the cement-based mortar products are soluble salt such as chloride. Mortar-composite blocks composed of cement mixed with the slurry of partial degraded spinning waste at the end of the hardening time, these specimens were completely immersed in 9% NaCl solution for increasing periods (1, 4, 12, and 48 weeks). At the end of each interval, the blocks were subjected to compressive strength measurements. Mass losses, porosity and water absorption, were evaluated, (Table 2).

It is clear from the mechanical integrity results that, there were detectable decreases in compressive strength values with increasing the immersion time up to 48 weeks. The action of sodium chloride on the mortar-composite is thought as being mainly physical, created by its crystallization, at continuous relative humidity, filling the pores and creating inherent pressure on the pore walls [39]. By increasing the immersion time up to 48-week significant increases in the compressive strength values was detected. This may be referred to the accumulation of more Ca $(OH)_2$, formed through cement hydration by the time and its reaction with pozzolane in the blocks.

The mass-loss data, due to the NaCl solution action on the mortar blocks, showed non-significant changes up to the 12 weeks relative to the unattacked samples. Even after 48 weeks of immersion in only 4.3% mass loss percentage was reached, this considered as a low value under the extensive condition of complete immersion. (Table 2).

Undistinguished variations in the porosity and water absorption values were reported for the candidate mortar composite blocks even after one year of static dipping. According to Lubelli [40], it should be notified that, the presence of sodium chloride can act as a catalyst dissolving gypsum, which considered as a big destructive agent for mortar products keeping a free surface around for the formed calcite that precipitated through the mortar pores [40]. Based on the data obtained for the mechanical integrity and porosity measurements, it could claim that the candidate mortar-composite products can tolerate the aggressive action of sodium chloride solution.

Integrity of composite under immersion in sodium hydroxide solution: Table 3 represents the compressive strength values, pH, porosity and mass loss percentages for the mortar composite blocks

Table 1: Compressive strength, mass loss, porosity and water absorption for mortar-composite blocks immersed in 0.1 N H_2SO_4 for increasing periods.

Exposure time, (week)	Compressive strength, (N/mm^2)	Mass loss, %	Porosity, %	Water absorption,%
Zero	23.88	-	21.28	10.65
1	18.92	2.5	21.0	10.6
4	13.4	2.55	18.71	107
12	6.4	3.3	20.4	11.7
48	5.43	5.0	21.35	13.92

Table 2: Compressive strength, mass loss, porosity and water absorption for mortar composite blocks immersed in 9 % NaCl.

Time of immersion, week	Compressive strength, N/mm^2	Mass loss, %	Porosity, %	Water absorption, %
Zero	23.88	-------	21.28	10.65
1	14.34	2.5	19.2	9.6
4	13.18	2.6	20.15	10.41
12	10.62	2.64	20.64	10.42
48	17.0	4.3	20.3	10.47

Table 3: Compressive strength, porosity, pH and mass loss for mortar-composite blocks* immersed in 0.1 N sodium hydroxide for increasing periods.

Exposure time (week)	Compressive strength, (N/mm²)	Apparent porosity, %	Mass loss, %	pH
Zero	23.88	21.28	-	12.3
1	19.0	19.9	1.21	13.0
4	10.0	19.1	2.61	13.01
12	14.5	19.4	2.34	13.04
48	19.3	20.3	2.26	13.05

*Mortar-composite composed of cement-Slurry at ratio 0.7 and cement: sand at ratio 1:3

subjected to 0.1 N NaOH for the various intervals. It is clear from the table that, the mass-loss percentage is nearly duplicated at the end of the fourth week. However, by extending the exposure time non-significant changes in the mass-loss percentages were detected. Furthermore, it is clear from the same table that there is a parallel decrease in the compressive strength values for the mortar-composite blocks up to the fourth week. By increasing the immersion periods, the compressive strength values escalated again after 12 and 48 weeks, respectively. The diminish in the mechanical durability in the first period may be attributed to the dissolution of calcium salts from the hard products while the increase in the compressive strength values with the immersion time progression can be referred to the $CaCO_3$ precipitations in the presence of NaOH solution and atmospheric carbon dioxide [41]. It should be noted that the lowest compressive strength value recorded (i.e.10 N/mm^2) is three times more than the value recommended by NRC required for subsequent transportation and final disposal process [37]. Moreover, the composite showed mechanical stability against

alkaline attack compared to the acidic one. Hence, the products can be used safely in circumstance when the alkaline conditions are expected.

Besides, it is clear from Table 3 that undistinguished variations in the apparent porosity percentage up to one year of immersion were detected. This indicates that non-vital deteriorations in the microstructure of the obtained mortar-composite were disclosed. This confirmed by the data obtained for the pH values of the leachant solution through the whole immersion period.

Bioremediation of low and intermediate level radioactive waste simulate was carried out using fungi or high plants. At the end of the treatment step the obtained spiked mushroom or high plant fibres were incorporated into cement composites. The evaluation and characterization of the obtained composites were studied systematically.

A direct encapsulation of solid cellulosic-based waste simulate in cement without any treatment and that for immobilization of the bioproduct generated from the biodegradation process revealed an improvement in physical, mechanical and chemical characterizations of the final waste form following the second route. Therefore, whole approach (i.e. bioremediation followed by cementation of the bioproduct) seems to be promising for treatment and solidification of un-neglected part of solid low and intermediate level radioactive solid wastes (i.e. solid cellulose-based waste) [42].

The capability of water hyacinth as a potential candidate plant for phytoremediation of waste streams contaminated with ^{137}Cs and ^{60}Co was studied. Phytoremediation has been perceived to be cost-effective, aesthetically pleasing and more environmentally friendly (green) with lower complicated technology compared to more sophisticated and intrusive remedial methods [43].

The submitted work provides an introduction for cost effective, natural and environmental friendly clean up technology based on the capability of the floating plants, *Veronica anagallis-aquatica (L.),* to treat waste stream simulates contaminated with ^{60}Co and/or ^{137}Cs. The proposed trial was developed to overcome the drawbacks of the well-known conventional treatment methods [44].

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Author Affiliations

Top

Radioisotope Department, Nuclear Research Center, Atomic Energy Authority, Dokki, Giza, Egypt