

A Review of the State of Research and Utilization of Biomaterials in the Manufacture of Composite Materials Todate

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Abstract

Over the last four decades composite materials, plastics and ceramics have been the dominant emerging materials. The volume and number of applications of composite materials has grown steadily, penetrating and gaining acceptance in new markets. Modern composite materials constitute a significant proportion of the engineered materials market, ranging from everyday products to sophisticated niche applications. While composites have already proven their worth as weight saving materials, the current challenge is to make them cost effective. This paper presents review on the worldwide state of research of biomaterials and their utilization in the manufacture of composite materials.

Keywords: Biomaterials, composites

INTRODUCTION

The use of natural fibres as reinforcements in polymer composites to replace synthetic fibres such as glass, is presently receiving increasing attention because of their advantages that include: cost effectiveness, low density, and, therefore, lightweight as well as their availability as renewable resources [1-3].

Owing to the poor wettability and adsorbability towards polymers resulting from hydrophilicity of plant fibres, the adhesion between the fibres and polymer matrices is generally insufficient. To improve the interfacial bonding, either surface modification of the fibres [4] or plasticization of the fibres [5] can be carried out. Pretreatment of the fibres result in chemical and structural changes not only on the surface of fibres but also in the fibre cells, which in turn influences the properties of the composites.

It is worth noting that the chemical composition and cell structure of natural fibres are quite complicated. Each fibre is

essentially a composite in which rigid cellulose microfibrils are embedded in a soft lignin and hemicellulose matrix. In addition, the microfibrils are helically wound along the axis of the fibre to form ultimate hollow cells. Uncoiling of these spirally oriented fibrils consume large amounts of energy.

Recent advances in the use of natural fibres, such as flax, jute, hemp, straw, kenaf, coir and bamboo in composites have been reviewed by several authors [6-12]. Bilba et al.[8] examined fibres from four different sources; banana trees (leaf and trunk) and coconut tree (husk and fabric) before their separate incorporation in cementitious matrices, in order to prepare insulating materials for construction. Thermal degradation of these fibres was studied between 200 and 700°C within flow of nitrogen gas. Fourier Transform Infra-Red (FTIR) spectroscopy were also observed. Similar analysis and results were reported by other researchers [9-12]

This paper discusses the worldwide state of research of biomaterials and utilization in the manufacture of composites.

2. NATURAL FIBRES

2.1 Source, and Classification of Natural Fibres

Natural fibres are renewable, bio-based materials [13]. Natural fibres are primarily classified on the basis of their origins, either plants, animals, or minerals. All plant fibres are composed mainly of cellulose, lignin and hemicelluloses, while animal fibres such as hair, silk and wool consist of proteins [14]. Mineral fibres such as asbestos, graphite and glass occur naturally or as compounds [14]. The natural plant fibres are classified according to their origin or botanical type [15]. Classification of the various natural plant fibres according to their origin is shown in Figure 1.

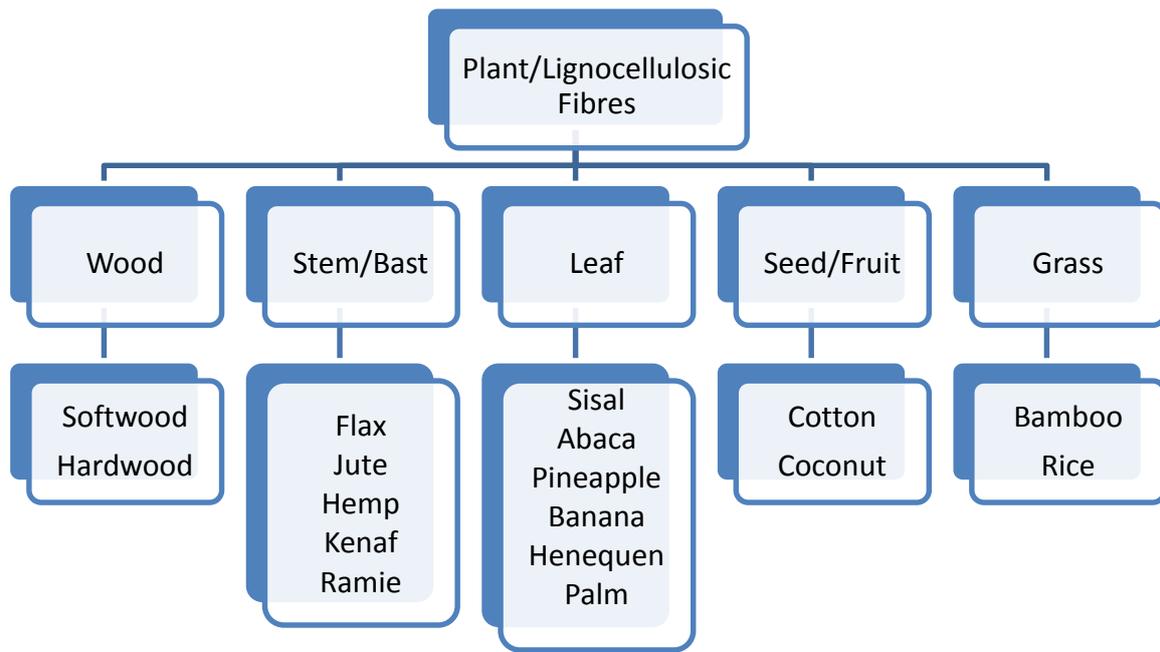


Figure 1: Classification of natural fibres [16]

Natural fibres have been used as reinforcing or filling materials for the past 3000 years. Biofibres are used for composites because of their low cost, ease of extraction, low density, enhanced energy recovery, reduced dermal & respiratory irritation, and significant biodegradability [17]. The stiffness, strength and ductility of polymers are enhanced by reinforcement with natural fibres. Furthermore, natural fibres are easily recyclable, and moreover, they do not be fracture when processed over sharp curvatures, unlike brittle fibres, such as glass fibre.

In terms of specific strength of materials, biofibres though having lower values do compare well with conventional or traditional fibres, such as glass, that are generally used for composites [18]. The sources, species and origins of natural fibres are varied as shown in Table 1.

Fibre Source	Species	Origin
Cotton	Gossypium sp.	Seed
Curaua	Ananaserectifolius	Leaf
Date Palm	Phoenix Dactylifera	Leaf
Flax	Linumusitatissimum	Stem
Hemp	Cannabis Sativa	Stem
Kapok	Ceibapentranda	Fruit
Kenaf	Hibiscus Cannabinus	Stem
Oil Palm	Elaeisguineensis	Fruit
Pineapple	Ananuscomosus	Leaf
Sisal	Agave Sisalana	Leaf
Wood	(> 10,000 Species)	Stem
Mangoes	Mangifera Indica	Fruit

Table 1. List of Important BioFibre Sources, Species & Origin

Fibre Source	Species	Origin
Abaca	Musa textilis	Leaf
Bagasse	-	Grass
Bamboo	(> 1250 species)	Grass
Banana	Musa Indica	Leaf
Broom root	Muhlenbergiamacrourea	Root
Cantala	Agave Cantala	Leaf
Caroa	Neoglaziovia Variegata	Leaf
China Jute	Abutilon Theophrasti	Stem
Coir	cocosnucifera	Fruit

2.2 Chemical Compositions and Properties of Natural Fibres

The chemical composition of natural fibres depends on the type and nature of fibre. The overall properties of each fibre are influenced by the properties of each constituent [19]. The main and prime constituent of all cell walls are sugar based polymers (cellulose, hemicellulose etc) chiefly on dry basis [20].

Natural fibres themselves are regarded as naturally occurring composites comprising mainly of helically wound cellulose microfibrils, embedded in amorphous lignin matrix. Cellulose (α -cellulose), lignin, pectins, hemicellulose, and waxes are the major components of natural fibres. The component

hemicellulose present in the natural fibres is regarded to be a compatibiliser between lignin and cellulose [21].

Table 2. Chemical compositions of some important natural fibres [22].

Natural Fibres	α -cellulose (%)	Lignin (%)	Pentosans (%)
Sisal	47 - 62	8 - 9	21 - 24
Pineapples	69.5	4.4	17.8
Wheatgrass	21.4	-	-
Bamboo	26 - 43	21 - 30	15 - 26
Rice	28 - 48	12 - 16	23 - 28
Ryegrass	33 - 50	16 - 19	27 - 30
Sugargrass	32 - 48	19 - 24	27 - 32
Wheat straw	29 - 51	16 - 21	26 - 32
Kenaf Bast	44 - 57	15 - 19	22 - 23
Jute	45 - 63	21 - 26	18 - 21
Oat straw	31 - 48	16 - 19	27 - 38
Esparto grass	33 - 48	17 - 19	27 - 32
Hemp bast	57 - 77	9 - 13	14 - 17
Barley	31 - 45	14 - 15	24 - 29

Chemical composition of some important natural fibres are illustrated in Table 2.

Hemicellulose and pentosans are responsible for thermal degradation, moisture absorption, and biodegradation of natural fibres as they show least resistance. Lignin on the other hand is thermally stable and is largely responsible for ultra violet light degradation [23,24,25].

Fibre width and length are important parameters used for comparing a diverse variety of natural fibres. The strength of fibres is an important factor in selecting a specific natural fibre for specific applications. The morphology and the anatomy of aquatic plant fibres are quite different from those of terrestrial plant fibres [26]. The necessary strength and stiffness of the fibres are provided by hydrogen bonds. Fibre variability, crystallinity, strength, dimensions, defects, and structure are the important factors governing the properties of different natural fibres [27,28].

Table 3 shows the average diameters of frequently used natural fibres. All fibres are tinny in diameter or width [29].

The major drawback of natural fibres is their high affinity to absorb water which, makes them incompatible with non-polar polymer matrices [30]. Most resins are usually hydrophobic in nature and absorb little moisture. Table 3 shows the equilibrium moisture content of different natural fibres at relative humidity (RH) of 65% and at 21 °C [31].

Table 3 : Average Diameter and Equivalent Moisture Content (EMC) of Natural Fibres [31]

Fibres	Diameter (μ m)	EMC %
Oil Palm Fronds	19.7	-
Oil Palm EFB	19.1 - 25.0	-
Banana	12 - 30	15
Sisal	7 - 200	11
Pineapple Leaf	20 - 80	13
Ramie	18 - 80	9
Coconut Husks	100 - 450	10
Bagasse	10 - 34	-
Jute	5 - 200	12
Hemp	10 - 500	9
Flax	5 - 600	7
Aloe	-	12
Wood	-	12
Abaca	-	9.5

Being natural in origin, the lignocellulosic fibres have several different stages of production, moreover, within each stage there are many factors that determine the quality of fibre, as shown in Table 4.

Table 4: Factors affecting Fibre Quality at Various Stages of Natural Fibre Production.

Stage	Factors Affecting Fibre Quality
Plant Growth	- Species of Plant - Fibre Location in Plant - Local Climate
Harvesting Stage	- Fibre ripeness, which effects cell wall thickness & coarseness of fibres - Adherence between fibres and surrounding structure
Fibre Extraction Stage	- Process used for decortication - Type of method used for retting

2.3 Polymer/Matrices

Matrices may be either metallic, ceramic or polymeric in nature. There are three types of polymers namely; thermoplastics, thermosettings, and rubbers. Thermoplastic polymers are often solvent soluble, and melt easily. Rubbers are slightly cross linked, which prevents their molecular chains from sliding when stretched, while thermosetting polymers are heavily cross linked and insoluble in solvents (32). Thermosetting matrices include phenolic resins; epoxy resins, unsaturated polyesters (UP) and amino resins.

3.0 MOISTURE ABSORPTION IN NATURAL FIBRES AND ITS EFFECT ON THE MECHANICAL PROPERTIES OF NATURAL FIBRE REINFORCED POLYMER COMPOSITES

The major limitation of employing natural fibres in composites is that the fibres are hydrophilic (attract water), while polymeric matrix materials are usually hydrophobic (repel water) (39). The uptake of water by hygroscopic substance such as cellulose and hemicellulose is a hydration process involving accessible hydroxyl groups (40). High levels of absorption of moisture by natural fibres lead to poor wettability by hydrophilic polymeric matrices and, therefore, insufficient interfacial adhesion with the polymer matrices. This limits the reinforcing effect of the fibres on the matrices (41). In order to expand the use of natural fibres, as successful reinforcement in polymer composites, their surfaces need to be modified to reduce the absorption of moisture. This enhances the adhesion of the fibres to the polymeric matrices, and since the level of interfacial adhesion between fibres and matrices determines the mechanical properties of fibre reinforced composites, this renders natural fibre reinforced polymer composites to have improved mechanical properties (42).

Eradication of the uptake of moisture by the reinforcing fibres in fibre reinforced polymer composites is a big challenge (43, 44). However, it can be minimised through surface treatment and also by ensuring a good encapsulation of the fibres by the matrix (45).

Chemical treatments of natural fibres (alkali treatment, silane treatment and acetylation treatment) and the use of compatibilisers have been suggested as a means to create strong interfacial bonds between the natural fibres and polymeric matrices in order to improve the mechanical properties and the resistance to moisture absorption of natural fibre reinforced composites (46 - 50).

3.1 Chemical Treatments of Natural Fibres and their effects on Mechanical Properties

Alkali treatment with sodium hydroxide (NaOH) also known as mercerisation is widely used for the treatment of natural fibres (51). Mercerisation is a process of chemical modification that changes the chemical behaviour of natural fibres (52). Alkali sensitive hydrogen bonds which usually exist in the fibre celluloses are broken down by this process and new reactive hydrogen bonds formed between the cellulose molecular chains. As a result of this; the hydrophilic hydroxyl groups are partially removed and the moisture resistance property of the fibres improved (53,54). The treatment also leads to a reduction of hemicellulose, pectin, lignin, wax and oils on the surface of fibres which give rise to a less uniform fibre surfaces that provide better interlocking with matrices and leads to improved interfacial stress transfer of the fibres (55). The treatment further leads to a reduction of the fibre diameter and, therefore, an increase in the aspect ratio of the fibres and hence an increase in the volume fraction reinforcing fibres (55,56). When using this treatment care should be taken not to exceed the optimum conditions since

this will lead excessive delignification of plant fibres that are damaged and weaker (57,58,59). Xue et al. (60) reported an optimum limit of 4-5% NaOH, in mercerisation, to obtain the best tensile strength of treated plant fibre reinforced composites.

Silane is mostly used as a coupling agent for the treatment of the fibre surfaces. This method involves three stages, namely, hydrolysis, condensation and formation of bonds(60). In the presence of water, hydrolysable alkoxy groups usually lead to the formation of silanols. The silanols then react with the hydroxyl groups of the fibres, forming stable covalent bonds that are chemisorbed onto the matrix surfaces (61). Therefore, the hydrocarbon chains arising the application of silane, restrain water absorption swelling of fibres by creating a cross-linked network of covalent bonds between the matrix and fibres. The method also results in the improvement of the fibre-matrix adhesion (62).

In acetylation treatment method, acetyl groups are grafted onto the cellulose structure in natural fibres with or without an acid catalyst. This method leads to improvement in the dimensional stability of composites (63).

During benzylation pre-treatment, fibres are usually soaked in benzoyl chloride for 15 minutes. Ethanol is then used to remove the benzoyl chloride that has adhered to the surfaces of the fibres. The fibres are then usually washed in water and dried in oven (64).

Interfacial adhesion between matrices and such treated fibres can be improved further by peroxide treatment. In this method, peroxide induced polyethylene is grafted onto the surfaces of fibres and the peroxide initiated free radicals then react with hydroxyl groups of fibres and the matrix. This treatment also reduces the moisture absorption tendency of the fibres and improves their thermal stability (65).

Islam et al. (66) carried out research using industrial hemp and Polylactic acid (PLA) to produce a composite material. The composite material was produced through compression moulding process. The fibres were treated with an alkali to improve the interfacial adhesion with PLA. They found that the interfacial shear strength (IFSS) improved with this alkali treatment of the hemp fibres. The tensile strength, Young's modulus, flexural strength and flexural modulus were all seen to improve upon this treatment of hemp fibres fibres. Other researchers who used alkali treatment on different natural fibres concurred with the findings of Islam et al.(66) that the tensile and flexural strength increase upon the treatment of the fibres with an alkali (67-72).

The researchers argued that mercerisation (alkali treatment) resulted in an overall improvement of the properties of natural fibre reinforced composites. The researchers, further, reported that mercerisation led to a modification of the texture of fibre surfaces and an increase in the amount of cellulose on the fibre surfaces. They noted that this leads to an increase in the number of the reactive sites on the surfaces of the fibres. They reported that alkali treatment enhanced the strength of natural fibres.

Giuseppe et al. (73), however, cautioned that the use of too much alkali treatment could lead to a decrease in both the

tensile and the flexural strengths due to the degradation and delignification of fibres. The researchers strongly recommended silane treatment as an alternative due to the strong covalent bond that results between fibres and matrices when silane is grafted on the fibres.

Herrera et al. (74) used silane treatment on short henequen fibres. The treated fibres were then used to produce a composite material with a polyethylene matrix. The researcher confirmed that the silane coupling agent was efficiently held on the fibre surface through the presence of a Silane-Oxygen-Silane bond on the lignocellulosic surface of the henequen fibres. The researchers found that the tensile and the flexural strengths of henequen/polyethylene composites increased upon the treatment of the fibres with silane. The researchers further explained that the silane molecules are grafted onto the henequen fibres and, therefore, create stable ties that increases the interfacial bonding.

Bledzeki et al. (75) investigated the influence of acetylation on flax fibre reinforced polypropylene composites. The researcher reported that the tensile and flexural strength increased with increasing degree of acetylation upto 18% and then decreased upon further addition of more acetylene.

Morphological studies have also revealed that benzylation and peroxide treatment of fibres give rise to improved fibre surface properties and lead to higher tensile and flexural strength than for composites with untreated fibres (76).

4.0 BIOCOMPOSITES

The term biocomposites is often used to refer to natural fibre reinforced polymeric composites. Composite materials are advanced and adaptable engineering materials. The combination of a plastic polymeric matrix and reinforcing natural fibres produces composites, possessing a combination of the properties of each constituent component. Natural fibre reinforced composites are eco-friendly as opposed to conventional composites and have unique advantages including lower density (77), better matrix-fibre compatibility, and recyclability (78).

The performance of the natural fibre-polymer composites are influenced by several factors, such as microfibrillar angle, defects, cellular structure, physical properties, chemical composition, cell dimensions and mechanical properties of the fibres, as well as, the interaction of fibres with the matrix.

The most important factors in the development of natural fibre reinforced composites are : surface adhesion, thermal stability, and dispersion of the fibres in the case of thermoplastic composites (79). The polarity of the natural fibre produces difficulties of incompatibility with many polymers. Hydrophilic or polar characters of natural fibres produces polymer composites with weak interfaces. Several chemical modifications or pretreatment of surface are normally made to improve and enhance the adhesion or interfacial bonding between polymers and natural fibres. Almost all known techniques for production can be used to manufacture natural fibre-reinforced polymer composites.

Table 5 displays list of techniques used for manufacturing natural fibre reinforced composites involving either thermoset or thermoplastic matrices.

Table 5: Techniques for Composite Preparation with Polymer matrix

Processing of Natural Fibre-Thermosetting Composites	Processing of Natural Fibre-Thermoplastic Composites
Hand lay up & Spraying	Extrusion
Compression Molding	Injection Molding
Resin Transfer Molding	Thermoforming
Injection Molding	Cold Pressing
Compression Molding	Filament Winding
Vacuum Assisted Resin Transfer Molding	Rotational molding

A significant and effective method of formulating biocomposites of desired and superior properties include proficient chemical modification of fibres and choice of efficient fabrication techniques (80).

4.1 Applications of Natural Fibre Composites

Natural fibre composites are cost effective materials for the following applications:

- Storage devices: post-boxes, grain storage silos, biogas containers.
- Furniture: Chairs, tables, shower cabinets and bath units.
- Electrical devices: Electrical appliances and pipes.
- Everyday applications: Lampshades, suitcases and helmets.
- Transportation: automobile and railway coach interiors, and boat hulls as well as interiors.

4.2 Factors affecting the Properties of Bio-composites

Natural fibres consist of different chemical constituents that exhibit different reactions to the environment and matrices during processing, and which need to be addressed during fabrication.

4.2.1 The Factor of the Structure of Plant Fibres

Cellulose structure is composed of crystalline and amorphous regions. Strong intra-molecular hydrogen bonds with large molecules are formed by the crystallite cellulose. Compactness of the crystalline region creates blocks of cellulose that make it difficult for the penetration of chemicals to occur. However, as in the case of dyes and resins, chemical are easily absorbed by the amorphous regions. In addition to the foregoing, plant fibres are hydrophilic in character due to

the presence of hydroxyl groups in their structures. In order to reduce the hydrophilic/hydrophobic mismatch of fibres and matrices through for instance, neutering of hydrophilic hydroxyl groups and removal of surface impurities, natural fibres need to be modified chemically [51 – 76].

4.2.2 The Factor of Thermal Stability of Plant Fibres

The structural constituents of the fibres (Cellulose, hemicelluloses, lignin, pectin and waxes) are sensitive to different ranges of temperatures. It has been reported that lignin starts degrading at a temperature around 200°C, while hemicelluloses and cellulose start degrading at temperatures of between 200°C and 400°C [62-76]. Thermal stability of natural fibres with reference to the structural component cellulose can be enhanced by removing certain constituents such as hemicelluloses and lignin, from the fibre by different chemical treatment.

4.2.3 The Factor of Fibre Length/ Loading, Distribution and Orientation

The properties of fibre reinforced polymer composites depend on the fibre length, percentage of fibre volume/ loading, distribution and orientation into the matrix. When load is applied to the matrix, stress transfer occurs by shear at both the interface along the fibre length and at the ends of the fibre. The extent of the load transmitted to the fibre is a function of critical fibre length, direction and orientation of fibres relative to each other and the compatibility between fibre-matrix interfaces. Depending upon the orientation and nature of fibres in the matrix, three types of fibre reinforcement can be obtained. Firstly, longitudinally aligned (continuous and discontinuous) fibrous composites generally have higher tensile strength and stiffness but lower compressive strength due to the fibre buckling. Secondly, in the transverse direction, the failure strength is equal to the interfacial rupture strength which is generally lower than the strength of the matrix. Finally, for randomly oriented short fibre composites, prediction of mechanical property is far more difficult, because of fibre dispersion, orientation and complexities of load distribution along the fibre matrix interface. By controlling factors such as aspect ratio, the dispersion and orientation of fibres, considerable variation in the properties of composites can be obtained [67-72].

4.2.4 The Factor of the Presence of Voids

During the introduction of fibre into the matrix, air or other volatile substances may be trapped in the material. During curing, micro-voids may form in the composite along the individual fibres due to the fibre spacing and between the laminate and in the resin rich regions, which has the adverse effect on the mechanical properties of the composites. The process of curing and cooling rate of the resin are responsible for the formation of voids [65]. A higher void content (over 20% by volume) is responsible for the low fatigue resistance, greater affinity to the diffusion of water and increased

variation in the mechanical properties of composites [66-68]. Composites with a higher fibre content usually display the formation of more voids [69].

4.2.5 The Factor of Fibre Cross-Sectional Areas and Profile

Maringa in his work showed the cross-sectional area and profile of natural sisal fibre to vary from fibre to fibre and from one location on each fibre to the other [70]. Natural fibres also lack rigidity and therefore, are not properly aligned to one another during reinforcement. This leads to a variation of the properties of natural fibre reinforced composites along the length of the reinforcing fibres.

5.0 CONCLUSIONS

- Natural fibres are gaining interest as reinforcement for polymer composites due to their potential mechanical properties, processing advantages, environmental and economic benefits.
- The performance of natural fibre reinforced composites can be affected by many factors including length, orientation, distribution, voids and temperature.
- Surface treatment is essential to reduce hydrophilicity of natural fibres, resulting in an increase in adhesion with hydrophobic matrices.
- The methods of chemical surface treatment include silane treatment, acetylation, peroxide treatment, benzylation and mercerisation.
- The methods of chemical surface treatment modify the structural constituents and surface morphology of natural fibres. Significant improvements in mechanical properties and thermal stability have been recorded from these surface treatments.
- The use of natural fibres from renewable resources for the manufacture of structural composites offers acceptable properties and provides benefits to the environment by replacing non-biodegradable synthetic fibres.

6.0 RECOMMENDATIONS

- Future research on natural fibre composites requires to explore further research on the different responses of the different constituents of natural fibres to the environment.
- There is need for more analysis on the effect of varying profiles and cross-sectional areas of natural fibres.
- There is need for more research to be done on the effects arising from variations due to plant growth, harvesting and extraction stage.

- More research is needed on the possibilities of natural fibres being used in the form of hybrid natural fibres or natural fibres with synthetic fibres.
- More research is needed on the effects of voids on the composites formed.

7.0 CONFLICT OF INTEREST

The researchers have no conflict of interest to disclose with regard to the current review.

8.0 ACKNOWLEDGEMENTS

The review was supported by the Vaal University of Technology, South Africa.

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