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Exploration of Local cellulosic-fiber; its Modification and Potential use by the Industry

N. Mokaloba[1], V. P. Kommula[2]

[1]*Botswana Institute for Technology Research and Innovation, Building Material Science Division; Gaborone, Botswana*

[2]*University of Botswana, Faculty of Engineering & Technology; Department of Mechanical Engineering; Gaborone, Botswana*

Corresponding author: N. Mokaloba; E-mail: Nmokaloba@bitri.co.bw.

ABSTRACT

Demand for newer, stronger, stiffer, yet lighter-weight and environmental friendly (biodegradable) materials in the fields such as automobile for non-structural applications are ever increasing. The principal reasons for using natural (cellulosic) fibers is they possess several attractive properties such their economic feasibility, enhanced sustainability, good specific mechanical properties, and desirable aspect ratio for good performance after melt-processing. Natural fiber composite materials are now being rapidly utilized in automobile industries, and they have become the forefront of research and development activity. An interesting alternative for reinforcing soft polymeric matrices with short fibers is the use of cellulose fibers which show remarkable reinforcing effects in thermoplastics such as polypropylene. The current study made an attempt to investigate the suitability of sisal fibers for automobile industry for non-structural and low-strength interior applications. In this work native sisal fibers were extracted and the effect of alkali treatment on their morphological, tensile, moisture absorption and thermal properties were studied. Scanning electron micrographs indicated roughening of the surface of the fiber strands due to the removal of the hemicellulose layer on alkali treatment. The maximum weight-gain for the composite prepared from treated fibers was 2.12 %, while that for the composite prepared from untreated fiber was 4.33 %. From the thermograms, the results indicate initial degradation for the treated fiber to have improved from 174 ^{o}C to 230 ^{o}C (56 ^{o}C shift) when compared to the untreated fiber. This fiber has competitive advantages when evaluated with other natural fibers. A polymer composite was processed from the chemically modified fiber, profiled against equivalent material systems in Ashby material property charts exhibited its suitability for light, low strength and low flexure material applications which can use a potential replacement of fibres being used currently.

1.0 INTRODUCTION

The understanding of cellulosic fiber modification for effective reinforcement in polymer-based composites is progressive. It can be stated that the applicability of cellulosic short-fiber composites is steadily forging ahead with realization of tailored engineering materials. The major driving force of natural fibre market is the need for energy conservation. This development has made the application of natural fibre in automotive industry more prominent with a projection of attaining progress in the next decade. This is evident from global research efforts on the analysis, processing and properties of natural-fibre reinforced composite related to application in the automobile industry. The most important types of reinforcing fibers are flax, sisal, hemp, and coir [1]. Natural fibers have many advantages compared to synthetic fibers. Advantages of natural fibers include relatively high stiffness which is a desirable mechanical property in composites; and relatively low cost, a desirable economic value. They are obtained from abundant and renewable resources at low cost, which ensures a continuous fiber supply and a significant material cost saving to the plastics industry.

Natural fibers, despite their low strength, could lead to composites with high specific properties because of their low densities [2]. The sisal fibre reinforced composites have been useful found in several fields such as automotive and the built-environment sectors. The most important markets being light-weight components for automobile interiors, decking and consumer goods. The most commonly used plastics in natural fiber composites are polypropylene (PP), polyethylene (PE) and polyvinyl chloride (PVC). Bio-based polymers such as Bio-PE and polylactide (PLA) are also used [3]. Nowadays, replacing synthetic fibers with natural fibers in the automotive industry can yield economic, environmental and social benefits. This area of research continues to be of interest to engineers and professionals as natural fiber composites turning out to be an alternative solution to the ever depleting non-renewable sources. It has been found that these natural fiber composites possess better electrical resistance, good mechanical properties, good thermal and acoustic insulating properties, as well as higher resistance to fracture [4].

1.1 Fibers in industrial applications

In terms of world production, sisal occupies the 6th place among fibre plants, representing 2% of the world's production of plant fibres, and it accounts for about 70% of the world's hard fibres. The world's top 5 producers of sisal from 2005 to 2013, average quantities in kilotonnes were Brazil: 231, Tanzania: 27, Kenya: 24, Mexico: 18, Madagascar: 17 [5]. From these figures, the combined Southern- and Eastern Africa (South Africa, Tanzania, Kenya, Madagascar) output provide substantial reserves for local industrialization to support the world's lucrative automobile and aviation sectors. Some of the reasons for investment consideration in Africa include (i) the ideal climatic and weather conditions for the growing and cultivation of sisal, where there is rainfall above average and good temperature supported by suitable soil, (ii) high level research facilities which provides industry research activities in areas such as improved varieties, farming practices and processing techniques, (iii) supply of relatively affordable labour, and (iv) regulatory framework from a single source of investment advice on the developmental sectors, and (v) access to the regional market which provides investors with a large potential market for their products and duty-free exports to other countries. Figure 1 shows the utilization percentage of wood based composites used in 2002.

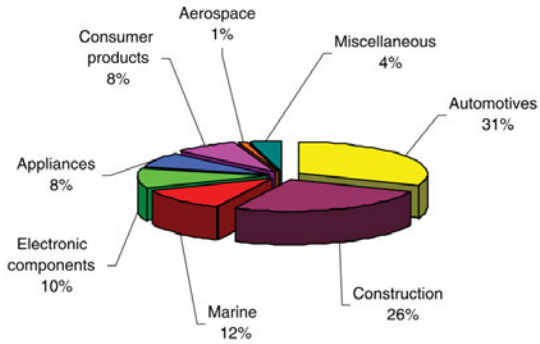


Figure 1: % utilization of fiber based composite in various industrial sectors [8].

Flax/sisal fiber mat embedded in epoxy resin was used as an improvement in door panels of Mercedes-Benz E-class [6]. About 20 % weight reduction has been achieved for door panels by introducing fiber reinforced composites [7]. Automobile companies like Audi, Opel, Daimler-Chrysler, Fiat, Ford, Mercedes Benz, Peugeot, Renault, Volvo, VW, Volkswagen and BMW are currently exploring new opportunities to use natural fibers in their products [8]. Figure 2 shows the amounts (in kg) of plant fibers used in automotive sector for different applications. Additionally, up to 5 kg could be used in other parts of a vehicle interior [9].

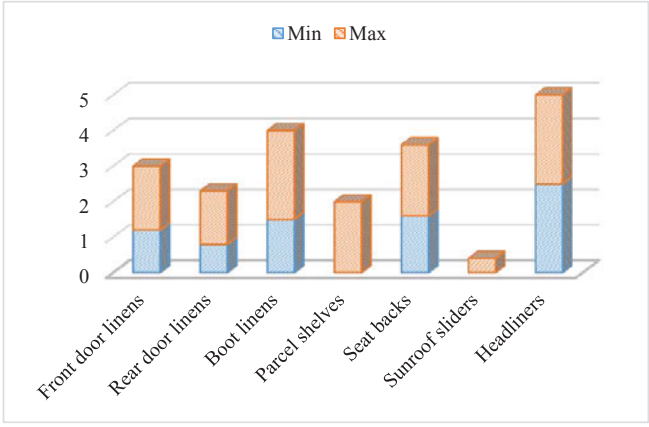


Figure 2. The amount of plant fibers used in automotive industry (in kg) [10].

Thermal conductivity is very important property when it comes to automotive interior applications. Hollow tubular structure of natural fiber enhances the insulation against heat and noise. Figure 3 shows the comparison of the thermal conductivity of various natural fibers [10]. From Figure 3, it is evident that sisal has lower thermal conductivity when compare to date palm and hemp, with little difference (0.04 W/mK), when compared with coir. This data reflects the competitiveness of sisal fiber in automotive application.

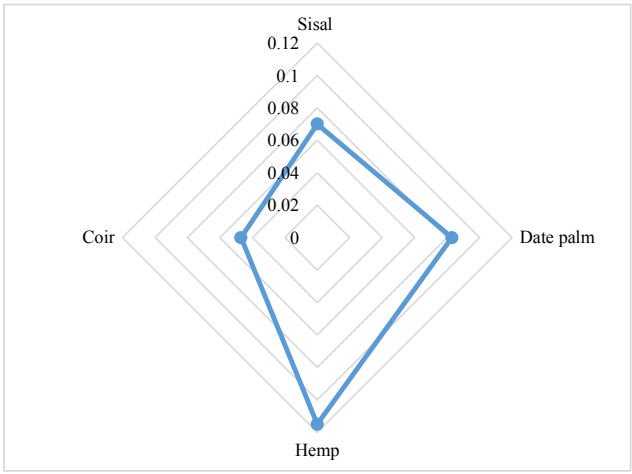


Figure 3. Thermal conductivity of various natural fibers (W/m K) [11, 13].

1.2 World fiber production and prediction of demand

The growth in the world fiber production continues to find relevance in view of weight reduction these materials portend to offer the automobile industry. In recent times, it has been affirmed that automobiles industry alone requires sisal fiber in the range of 170,000 ton/yr in parts-formation, while the replenishing approaches may not deliver 50 % of these fibers in the next 50 years. According to other researchers, the demand of natural fibers appear to have cut across continents. In China alone, the prediction for composite and lighter weight materials demand is likely to reach 113 million tons by the year 2020, while in Western Europe, the demand may reach 76 million tons most importantly in the manufacturing of automotive parts [11]. Figure 4 (a) and shows the world fiber production and the forecast for composite materials demand into the year 2020. Africa requires about 10 million tonnes of composites to be supplied from year 2010 to 2020. From this, it is evident that the automobile sector has been a prominent consumer of fiber-based composites, also considering the escalating demand forecasted for the year 2020, as indicated at Figure 4 (b).

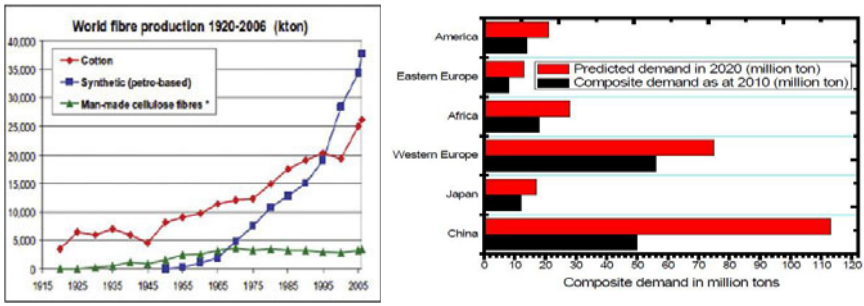


Fig. 4(a) World fiber production, (b) The forecast of composite materials by producer countries [10].

1.3 Cellulosic fibers and Composite processing challenges

Moisture sensitivity is the most serious problem when natural fibers are used to reinforce polymer composites. The quantity of absorbed moisture through the fiber structure depends on: (i) the relative humidity of the surrounding environment, (ii) the purity of the cellulose, and (iii) the degree of crystallinity. The hydroxyl groups, at least those in the amorphous regions of the fibers, have a strong affinity for water and other highly polar chemical species. This characteristic of cellulose fiber is also one of the major drawbacks for the successful use of natural fibers because of high moisture absorption and poor dimensional stability, as well as their susceptibility to rotting [2]. Sisal , being a source of lignocellulosic fiber, the composition of which includes cellulose, hemicelluloses, lignin, pectin, fat, waxes, and water soluble substances, bear hydroxyl groups from cellulose and lignin [2]. The large amount of hydroxyl group gives the hydrophilic nature to the cellulosic fiber. In order to develop cellulosic fiber matrix composites with better mechanical properties, it becomes necessary to increase the hydrophobicity of the reinforcing agent and to improve the compatibility between the matrix and cellulosic fibres. In general, all plant fibers are based on carbohydrate-based polymers, the most abundant polysaccharide of which is cellulose, which is composed of glucose units that are joined through hemmi-acetal linkages in the ratio 1:4 positions [1]. This structure makes plant fibers amenable to conferment of effective reinforcement properties via chemical process for composite applications with polymeric matrices. Several chemical treatments exist such as alkalization, acetylation, silane coupling agents, isocynates, and graft polymerization are conducted on cellulosic fibers to modify the interface and the morphological aspects in fibers [2].

With respect to processing, thermoplastics are favoured over thermosets owing to their low cost, ease of compounding and recyclable properties. Polyethylene and polypropylene are the most common thermoplastic matrix materials used. Despite the advantages from chemical treatment, the use of cellulosic fibers in thermoplastics has challenges associated with it. Reasons that contribute to unsatisfactory final properties of the composite include (i) limited thermal stability at typical melt processing temperatures of ~200 ^{o}C, which limits the type of thermoplastic that can be used with the fibers, (ii) poor dispersion characteristics in the non-polar olefinic thermoplastic melt due to strong hydrogen forces between the fibers, (iii) limited compatibility with many thermoplastic matrices due to their highly hydrophilic character resulting in poor mechanical properties of the composites produced, and (iv) high moisture absorption of the fibers which can affect the dimensional stability of the composite together with the interfacial bond strength [13]. The reinforcement caused by cellulose fibers in the thermoplastic material

is governed by parameters which include fiber dispersion, fiber-matrix adhesion, fiber aspect ratio, fiber orientation, and fiber volume fraction [14]. These factors need to be addressed during the processing of composites. The conventional techniques for processing thermoplastic composites comprise two or more stages of exposing natural fibres to high shear forces with consequent fiber-structure damage. One of the mixing methods used for cellulosic fibres and polymeric matrices is melt mixing, dominates the industrial sector. The best use of natural fibres occurs when processing methods reduce or eliminate fibre damage. Alternative processing of natural fibre-thermoplastic composites has become an important consideration for scientists and engineers. One such alternative is the application of environmentally benign powder impregnation processing technique where chopped fibre bundles and polypropylene powder are mechanically mixed and subjected to compression moulding to make composite panels.

2.0 EXPERIMENTAL

2.1 Fiber origin and chemical modification

Sisal fibers used in this research were harvested locally. The fibers were extracted from the leaves of the plant *Agave sisalana.* There was no information on fiber physical and mechanical properties. To ensure consistency of properties, fibers from the same plant were used. Fiber extraction involved mechanical liberation of the fiber from the inner core of the leaf, where leaves were split into longitudinal strips and pulled between metal blades. The fibers were rinsed, sun dried for 6 hourrs, and combed to remove impurities adhered to the surface. Peripheral mechanical fibers extracted from the leaf's mid-span region were used. Figure 5(a) shows the cross-section of the sisal leaf cut from the mid-span section of the leaf and the chopped fibers (figure 5(b)).

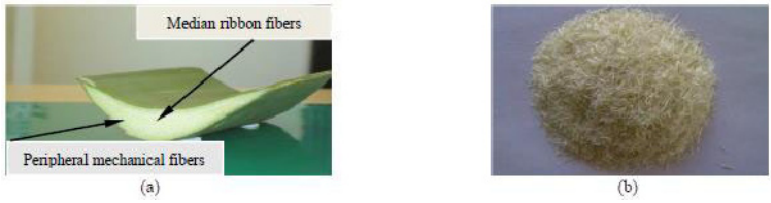


Figure 5. Sample of (a) cross-section of a fleshy, green sisal leaf showing fiber situate, (b) chopped sisal fibers.

Laboratory reagents used in preparing solutions for chemical treatment were sodium hydroxide pellets of 99 % purity and acetic acid of 99.5% purity, supplied by Merck Chemicals (PTY) Ltd, South Africa. The polypropylene used as matrix was supplied by Sasol Polymers, South Africa.

2.2 Alkali treatment protocol

The main modification done by alkaline treatment was the disruption of hydrogen bonding in the network structure, thereby increasing fiber surface roughness [15]. Treatment protocol involved soaking the fibers in sodium hydroxide (NaOH) of 6 % (w/w) for 45 min. The fibers were then thoroughly washed with distilled water and dried at room temperature for 48 hours. The fibers were also soaked in 1 % acetic acid to neutralize excess sodium hydroxide. They were then rinsed with distilled water and dried in an oven at 80 ^{o}C for 2 hours to remove free water.

2.3 Fiber surface morphology

The fiber surface morphology was undertaken to study the effects of chemical treatment on the surface structures of the fiber. The scanning electron microscope Model XL 30 was used. Excitation energies were set between 10-15 keV. Conductivity was via carbon deposition.

2.4 Thermo-gravimetric analysis (TGA)

TGA was used to study fiber weight loss as a function of temperature. This property depicted the thermal stability and fraction of volatile components in the fiber following chemical treatment. The Pyris 1 Thermo-gravimetric analyzer supplied by PerkinElmer was used. Samples weighing about 6.5 mg were analysed. The samples were held for 1 min at 30 ^{o}C and then heated from 30 ^{o}C to 600 ^{o}C at 10 ^{o}C/min in nitrogen atmosphere flowing at 20 ml/min.

2.5 Composite processing and Strength design

The composite was prepared from polypropylene and the fiber of 67 % crystallinity, 580 MPa U.T.S and 13 % enhanced thermal performance. A fiber volume fraction, V_f of 0.23, and fiber length of 5 mm were adopted. The polypropylene-sisal blend was achieved through melt-mixing at 180 ^{o}C, for 10 mins, at a rotor speed of 30 r.p.m. The composite panels were compression moulded at a pressure of 0.5 metric tons. Specimens for characterization were prepared from the panels. The strength of the composite was estimated from the Rule of Mixtures [16]:

$$\sigma_c = \sigma_m V_m + \sigma_f V_f \quad (1)$$

where σ_c represents the strength of the composite, σ_m and σ_f are stresses in the matrix and fiber correspondingly, and V_m and V_f are the matrix and fiber volume fractions correspondingly.

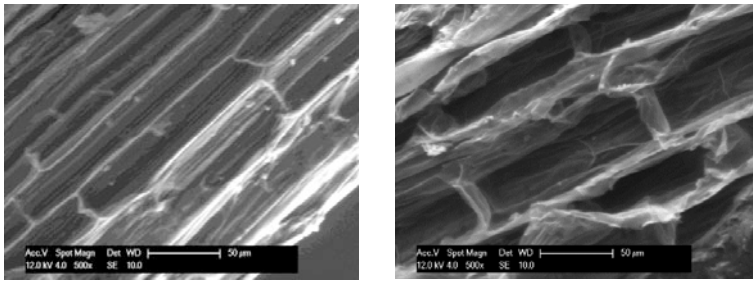
2.6 Moisture absorption characteristics of the composite

The water absorption characteristic was studied to establish dimensional stability. The specimens were cut from the composite panels and measured 50 mm wide, 70 mm long and 5 mm thick. The specimens were testing according to the ASTM D543-87 standard. In this method, the samples were initially weighed and then dipped in the water for 24 hours. Later the samples were removed from the water and dried. The samples were then weighed and the percentage of weight gain was determined.

3.0 RESULTS

3.1 Fiber surface morphology

Figures 6(a)-(b) show the surface morphology for the untreated and the alkali treated fibers respectively. When compared to the untreated fiber, the micrograph for the treated fiber exhibited less of attached particles on the fiber surface. This effect indicated disappearance of surface impurities, normally associated with waxes and fats. The micrograph also depicts exposure of the cell-wall structure, a phenomenon which promotes fiber-matrix interlocking from an increased contact surface area.



(a) (b)

Figure 6(a) Untreated fiber (b) fiber subjected to alkali treatment

3.2 Fiber structural integrity

Figure 7 shows the micrographs of a cellulose helical spiral depicting fractures (sites shown by arrows) sustained during extraction. Fractures are attributed to excessive mechanical stresses effected on the fiber. This phenomenon has potential to thwart optimal performance of the composite from the inefficient structural performance of the reinforcing agent. The transverse fractures were observed ranging from minor splits to total dissociation of adjoining coils. Susceptibility of cellulose helical spiral fracturing has also been reported for coir fibers [2].

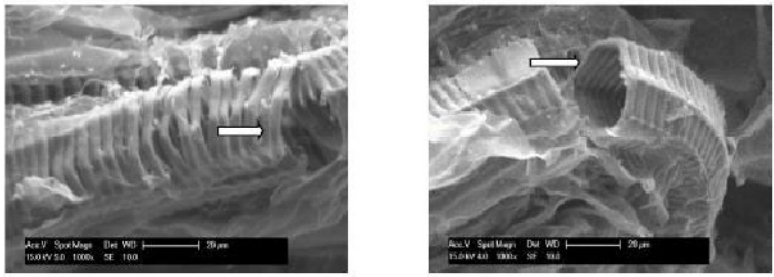


Figure 7 (a) Sisal fiber showing the fractured cellulose helical spirals and, (b) circumferential split.

3.3 Fiber thermal degradation

Figure 8 shows the TGA thermograms for untreated and the alkali treated fibers. From the thermograms, the results indicate initial degradation for the treated fiber to have improved from 174 ^{o}C to 230 ^{o}C (56 ^{o}C shift) when compared to the untreated fiber. This effect suggests suitability for the treated fiber to be processed at temperatures lower than 230 ^{o}C to preserve fiber structural integrity. A similar shift in onset of values was also observed for the inflection point (335 ^{o}C -379 ^{o}C). An inflection point is where the degradation rate is maximum [17]. This phenomenon indicates enhanced fiber thermal resistance resulting from chemical treatment.

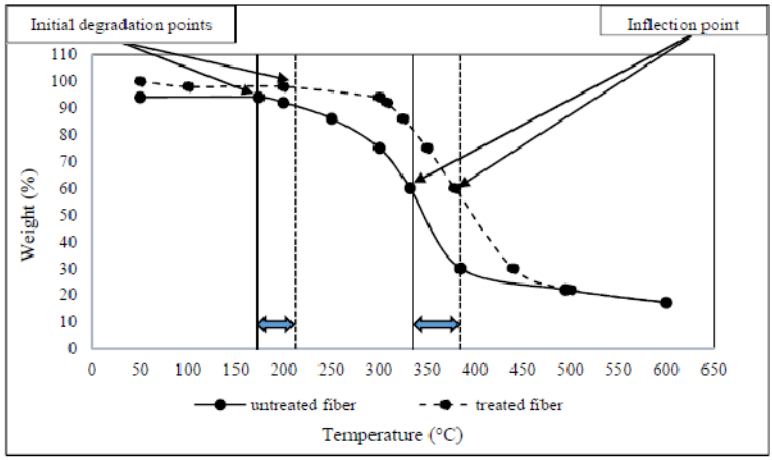


Figure 8. TGA thermograms for untreated and alkali treated fibers

3.4 Moisture absorption of the Composite

The maximum weight-gain for the composite prepared from treated fibers was 2.12 %, while that for the composite prepared from untreated fiber was 4.33 %. Moisture gain was attributed to the plasticization effect of the absorbed water molecules weakening interfacial bond strength between the fiber and matrix. Moisture absorption for the treated cellulosic-fiber composites have been reported at about 3 % [18]. A percentage gain in moisture absorption of 3.7 % for sisal/coir fiber-based composite at V_f of 0.2 has been reported by other researchers [19]. Compared to characteristics reported by other researchers, the pattern showed by the resultant composite was comparable, which suggested satisfactory functionality.

4.0 PROFILING COMPOSITE PROPERTY IN ASHBY PLOTS

The comparative evaluation of the resultant composite's performance against other materials was profiled in the Ashby materials-property chart for the performance index of Young's modulus *vs* Density. The Young's modulus and the Density for the resultant composite were 0.53 GPa and 1120 kg/m^3 correspondingly. Figure 9 shows the Ashby plot for the Young's modulus against Density. From the figure 9, it can be observed that the resultant composite (E: 0.53 GPa; ρ: 1120 kg/m^3) shown by a star symbol, lay within the envelope for engineering polymers. Other researchers have reported the Young's modulus of between 0.4 to 1.8 GPa for polymer-based cellulosic fiber composites [20]. The "cloud-like" boundary which envelopes the resultant composite represents approximate performance characteristics for some polymer-based composites. For flat plates or components loaded in bending, represented by the guideline $\frac{E^{\frac{1}{3}}}{\rho}$, the resultant composite connotes potential for light, low elastic deformation and moderate bending application.

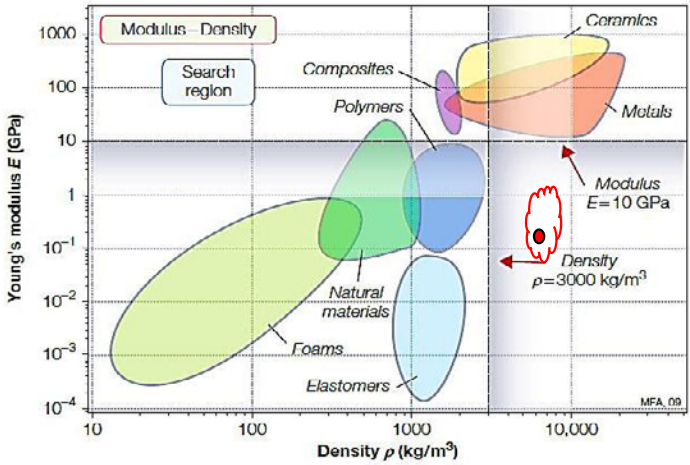


Figure 9. Young's modulus *vs*. Density Characteristics plot (Ashby chart adopted from [22])

5.0 CONCLUSIONS

This research work has investigated the functional characteristics of the local sisal fibers in appropriation for composite material application with alkali treatment. The following conclusions were drawn from this study:-

1. The local cellulosic fiber from *agave sisalana* exhibits physical (surface functionality) and mechanical (U.T.S, Stiffness) characteristics suitable for application as reinforcement agent in polymer-based composites.

2. The Ashby profile for the resultant composite denotes suitability for light, low-stiffness applications which fits the automobile interior panels/linings manufacturing domain.

3. Beneficiation exclusive to the automobile interior niche can be achieved by tapping optimally from the abundant resources of high-quality natural fibers from the Southern- and Eastern Africa trading blocs. The existing research facilities have potential to support realization of a sustainable research-to-manufacturing continuum.

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