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Characterization of polymer–quarry dust composite for structural applications

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ABSTRACT

This study explores the utilization of quarry dust in producing polymer composite for roofing tiles application. Quarry dust was incorporated into high-density polyethylene (HDPE) and polypropylene (PP) in various weight proportions (5 %, 20 %, 40 %, 60 %, and 80 %). Experimental tests that were conducted include the melt flow index (MFI), microstructural analysis, water absorption, Shore D hardness, and Charpy impact strength. The PP+60 wt.% quarry dust had the highest MFI value of 24.5 g/10 min hence easy to process. The water absorption for 60 % and 80 wt.% composition was found to be significantly high as compared to other proportions. The Shore D hardness value of 82 attained for samples having 80 wt.% quarry dust with the highest Shore D hardness value of 82 attained for samples having 80 wt.% quarry dust. For the impact strength, only 5 wt.% composition of quarry dust exhibiting a higher impact strength of 69.6 kJ/m². This work demonstrates that the properties of waste plastics can be improved by using quarry dust as reinforcement, suiting green construction applications such as the production of roofing tiles.

Introduction

Polymers are the world's most widely used materials because they possess attractive features including their ease of manufacture, relatively low cost, and low density [1]. In our day-to-day activities, polymers are essential materials as they are used to make most of the products and appliances at home and in industry. They are applied in the production of components for automobiles, electronics, building, and construction industries. In recent decades, the production of plastics has grown, contributing to environmental pollution. Most of the plastic wastes are non-biodegradable and one of the options for the removal of huge piles of plastic wastes is recycling them to create useful materials and products. There has been extensive research on using various plastic waste materials including poly-ethylene terephthalate (PET) [2–4], polystyrene (PS) [5], and expanded polystyrene [6] as fibers, fillers, or aggregates for concrete. The utilization of waste from plastics in the production of useful products such as floor and roofing tiles is a potential solution to reducing landfilling.

The use of natural reinforcement additives, like sisal, hemp, sand, quarry dust, sawdust, etc., has attracted interest in the recent

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past. The key considerations in different manufacturing sectors, including automotive and construction, are reduced energy consumption and cost reduction. There is increased attention to the development of cheaper environmentally friendly materials with a lower negative environmental impact [7–11]. The need for the use of natural reinforcement additives as an alternative material in the production of composites has increased because many varieties of natural reinforcing additives have better physical and mechanical properties and can compete with the existing synthetic fibers already used in composite production [12].

The key factors influencing the mechanical properties of composites containing natural additives as reinforcement agents are: the selection criteria of the fiber (including its type, harvest period, treatment, method of extraction, aspect ratio, and fiber quality), fiber orientation, interfacial strength, fiber dispersion, choice of the matrix, porosity and composite production process [13,14]. Since fibers are usually more robust than their matrix, the strength and the rigidity of the composite material typically increase as the fiber content increases. This depends, however, on adequate matrix/fiber interface strength. The affinity for moisture in both matrix and fiber filler material could largely influence the strength and the rigidity of the composite material. The use of binding agents or interfacial modifiers improves this interfacial strength [15]. However, Young's modulus usually improves as the fiber content is increased but more moderately than when the optimization of the interface is carried out [16]. One of the demerits of using natural reinforcement additives is that they result in a composite material having a low Impact strength [17]. For example, the maximum Charpy impact strength reported in the literature is 115 kJ/m² for unidirectionally pultruded flax/PP composites [18].

Quarry dust is a granular naturally occurring material that is made up of finely fragmented rock and mineral particles. Based on the local rock sources and the underlying conditions, the composition of quarry dust is highly variable. The most dominant component of quarry dust is silica (SiO₂), typically in the form of quartz, a mineral that exhibits weathering resistance and substantial hardness. There are several studies and supporting data that show scientists can boost the strength and durability of a polymer composite by incorporating natural fibers and linz-donawitz slag [19], red mud [20], granite powder [21], aluminum powder [22] and blast furnace slag [23] into the polymer matrix.

Although a number of studies have been conducted on the evaluation of mechanical, physical, and thermal properties of natural fiber polymer composites, little work has been done on the possibilities of using naturally found hard aggregates such as silica sand or quarry dust as reinforcement for polymer composites. This study aims to explore the possibility of developing a composite material by using HDPE/PP and quarry dust in different concentrations. The goal of this study is to investigate the possibilities of producing a composite material in various proportions of quarry dust (a byproduct of gravel stone quarries) integrated into high-density polyethylene and polypropylene. The composite is intended for the manufacture of roof tiles for green structural applications.

Experimental procedure

Materials

The quarry dust (Fig. 1) used in this research was obtained from a local quarry site in Nyeri County, Kenya where granitic stones are mined. This location was chosen because of the availability of huge hips of quarry dust that tend to cause air pollution and waste of productive land space. The selection of quarry dust as a reinforcement material in the polymer composite is typically based on its specific properties and characteristics that make it suitable for the roofing tiles application. Quarry dust is essentially a waste material produced during the extraction and processing of rocks in quarries, and it may be considered as a sustainable filler material. It is readily available in large quantities as a byproduct of the quarrying industry. Its abundance and low cost make it an attractive option for use as a reinforcement material in polymer composite. The particle size and distribution of quarry dust can be controlled, making it suitable for use in various composite applications through the achievement of various desired mechanical properties in the composite. Additionally, quarry dust particles can enhance the wear resistance of the composite, making it suitable for applications where abrasion resistance is important. The use of quarry dust as a reinforcement material aligns with sustainable and eco-friendly practices, as it repurposes a waste product that would otherwise be disposed of.

The quarry dust was used without further grinding or sieving. The particle sizes were measured using magnified images of the particles by inscribing particles in a circle and measuring the diameter of the circle. Energy-dispersive X-ray spectroscopy was used to obtain information on the elemental composition of the quarry dust.

The plastic material used in this research was virgin high-density polyethylene (HDPE) and polypropylene (PP) in the form of pellets (obtained from a polymer laboratory at TH Wildau, Germany). These virgin plastics acted as a substitute for plastic waste due to



Fig. 1. Quarry dust sample obtained from Nyeri County in Kenya.

the unavailability of the plastic waste at the time and place where the preparation and testing of the composites were conducted. However, it is reported in the literature that the difference between the properties of virgin and waste thermoplastics is insignificant if the processing parameters such as molding temperature and pressure are closely monitored although slight observed variations may be due to the use of different additives [24–26].

Preparation of polymer-quarry dust composites

To undertake the production of the roofing tile samples, a mixing machine, a compression molding machine, and a mold were used. The amount of quarry dust incorporated into the polymers varied in proportions of 5 %, 20 %, 40 %, 60 % and 80 % in weight. Achieving a well-dispersed quarry dust in a polymer composite is crucial for maximizing the material's performance and properties. Poor dispersion can lead to weak bonding, reduced mechanical properties, and uneven material characteristics. Use appropriate mixing equipment, such as high-shear mixers, twin-screw extruders, or internal mixers, which can help disperse the particles uniformly throughout the polymer matrix. The choice of equipment will depend on the specific composite and material properties. Additionally, overheating the composite during the mixing process can lead to agglomeration and poor dispersion.

The mixing of the composite constituents was undertaken using a Brabender Plasti-corder mixer (625,249,130, Germany). The mixing equipment has two counter-rotating mixing rotors and three heating walls to enhance the effective/complete melting of the polymer materials. The polymer was put in the heated chamber of the mixing machine running at 8.9 rpm after which quarry dust was added and the mixing was allowed to take place for 10 min to achieve homogenization. The quarry dust did not cause any abrasion on the mixing machine parts. The mixing temperature for HDPE was 180 °C while that of PP was 200 °C to avoid polymer degradation. After mixing, the hot composite materials were transferred to a preheated Polystat 400 S hot press for molding. The pressing machine contains two hydraulically controlled plates. Additionally, heating and cooling chambers are also provided for molding purposes. The chamber contains a 600 mm by 600 mm area of operation that can fit several sizes of molds. The hot composite mix was compression molded at 20 bar for 5 min while holding the temperature at 180 °C before cooling to 30 °C [27,28]. The formed samples were then cut into various sizes for characterization.

Characterization

Melt flow index

The Melt Flow Index is used to analyze the ease of flowability and processability of a polymeric material. This was determined using a Zwick/Roell Mflow (BMF-001) machine according to ISO 1873/1 for PP-quarry dust composite at 230 °C and ISO 1872/1 for HDPE-quarry dust composite at 190 °C. A shredding machine (Retsch SM 2000) was used to prepare the samples by shredding the hot-pressed samples to obtain small particle sizes that could easily enter the heating chamber for extrusion. For each sample, 4 g was measured using a digital weighing machine so that the capacity of the heating chamber could not be exceeded. A mass of 2.16 Kg was used to produce extrusion pressure. However, the mass was increased to 10 kg for PP+80 wt.% dust since the MFR was less than 0.1 g/10 min.

Morphological analysis

It is necessary to study the size and shape of the particles and the composition of the quarry dust which can help in the analysis of the composite properties. To analyze the morphology of quarry dust, optical microscopy (Keyence Optical Microscopy VK-X1000) and SEM microscopy (model JEOL JSM-6010 LV) were used. The dust samples were prepared by forming a 45 nm thick silver layer to enhance its conductivity by applying 50 mA current at 10 bars using a vacuum sputter coater machine (LEICA EM SCD 500, Germany). These analyses were carried out at room temperature on various surfaces covered by the quarry dust powder. The Energy Dispersive X-ray spectroscopy (EDX) was carried out to give the elemental composition of the quarry dust.



Fig. 2. A composite sample of sizes 60 mm \times 60 mm \times 1 mm was produced from polymer (HDPE and PP) and quarry dust.

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Water absorption

It is necessary to undertake this test to analyze the dimensional stability of the composite, susceptibility to mold growth, and possible changes in physical and mechanical properties during service in roofing applications. The water absorption tests were carried out on 60 mm \times 60 mm \times 1 mm samples according to ASTM D 570–98 at room temperature (23 °C) [29]. One set of samples was immersed in distilled water at atmospheric pressure and another set was immersed in water at 6 bars. The mass of the samples was recorded after 1 day, 1 week, 3 weeks, 5 weeks, and 7 weeks using a weighing balance with a sensitivity of 0.0001 g. Wiping of water on the samples was carried out before being measured. Fig. 2 shows a 60 mm x 60 mm x 1 mm compression molded sample used in the water absorption test.

Hardness testing

The Shore D hardness test was conducted as per ASTM D2240 standard using a Shore D Durometer Model PCE-DX-DS. The samples measuring 200 mm \times 200 mm \times 4 mm were positioned horizontally on a metallic surface, and the indentor hand pressed into the samples for one second at various points, ensuring that the devices were perpendicular to the metallic.

Impact energy testing

The Charpy unnotched impact test was conducted according to ISO 179-1:2010(E) using Zwick PSW 4 J equipment with a hammer size of 4 Joules. The samples were prepared according to ISO 2818 to avoid creating points of weakness on the surface. The unnotched sample sizes of $200mm \times 10mm \times 4mm$ were used. The energy obtained from the readings was used in the calculation of unnotched Charpy impact strength a_{cu} (kJ/m²) using Eq. (1).

$$\mathbf{a}_{\rm CU} = \frac{E_C \times 10^3}{h \times b} \tag{1}$$

Where: $E_c =$ the stored energy (J)

b = test specimen width (mm)

h = test specimen thickness (mm)

Additionally, the homogenization and microstructural interactions between the polymer matrix and the quarry dust in the composite samples were analyzed using SEM microscopy (model JEOL JSM-6010 LV). The analysis was carried out at room temperature at various points on the fractured surfaces of the Charpy impact test samples. The analysis was conducted only on the 80 wt.% quarry dust composite because there was no change in chemical composition among the samples produced, hence not necessary to analyze all the samples. Before SEM microscopy, the specimens were coated with a 45 nm thick layer of silver using a high vacuum sputter coater machine (LEICA-EM-SCD-500, Germany) at 10–12 bars using 50 mA current to enhance their conductivity.

Results and discussion

Determination of the melt flow index (MFI)

Fig. 3 shows the melt flow index of polymer-quarry dust composite samples with varying levels of quarry dust. The MFI test is important since it gives more information about the processability of a material. There are different MFI values obtained for both the PP and HDPE-based composites and no trend is seen as the amount of quarry dust is varied. The absence of a specific trend in the MFI test results for a composite material can be attributed to several factors that affect the flow behavior of the material among them are dispersion, orientation and agglomeration of the reinforcement. MFI measures the flowability of a polymer melt and is often used as an indicator of a material's processability. The quality of dispersion of the reinforcement within the polymer matrix plays a critical role in MFI. Incomplete or non-uniform dispersion can lead to localized variations in flow behavior, resulting in unpredictable MFI values. The orientation of fillers within the composite and the potential formation of reinforcement agglomerates can affect the MFI. If reinforcements are preferentially oriented or agglomerated, it can lead to variations in flow behavior in different directions, leading to



Fig. 3. Melt Flow Index of polymer-quarry dust composite samples with varying quarry dust composition.

mixed or unpredictable MFI results. For PP containing 60 % weight of quarry dust, a high MFI value of 24.5 g/10 min was observed and the mass flow rate through the chamber opening was too high. This implied that the quarry dust was well dispersed in the polymer matrix for the PP-60 % quarry dust composite. Small MFI values were observed in the rest of the samples and this was due to the presence of agglomerates within the composite material and/or the increase in viscosity of the material. Generally, low MFI values were observed in HDPE matrix composite samples as compared to those observed for the PP matrix composite samples. This implies that PP matrix had minimum formation of agglomerates and there was good dispersion of quarry dust in the matrix as compared to the HDPE matrix hence ease of processing.

Morphological analysis of quarry dust

Optical microscopy, SEM imaging, and energy-dispersive X-ray spectroscopy (EDX) were used to characterize quarry dust samples. Fig. 4 shows optical images of quarry dust. The various colors present in the image indicate the possible presence of different mineral compositions in the quarry dust. Fig. 5 shows SEM images of the quarry dust in which it is observed that the quarry dust contains particles with sharp edges. The size of the particles ranged from 1 to 50 μ m. EDX confirmed that the quarry dust is siliceous in nature due to the high amount of silicon and oxygen in the samples at 21 % and 62 % respectively as shown in Table 1.

The water absorption of polymer-quarry dust composites

Figs. 6 and 7 show graphs of water absorption for HDPE and PP composite samples respectively exposed to atmospheric pressure. With an increase in the concentration of quarry dust, the water absorption increases significantly for 60 % and 80 wt.% composition of quarry dust. The high water absorption at these weight concentrations (60 % and 80 %) is due to a weak adhesion between the interface of the polymeric matrix and the quarry dust and high porosity due to the increased amount of quarry dust as reported in the literature [30]. As per the general observation of the trends/shapes of the curves, the mass of the absorbed water increases with the square root of time of exposure of the samples to water, obeying Fick's Law, until saturation is achieved [31].

Figs. 8 and 9 show the graphs for water absorption for HDPE and PP composite samples respectively exposed to a pressure of 6 bars. The water absorption of composite exposed to a pressure of 6 bars is higher as compared to the samples exposed to atmospheric pressure. The high-pressure forces water into the composite increasing its water absorption capacity. Therefore, these findings suggest that the composite may not be suitable for high-pressure applications. Similar results have been reported for several composites exposed under hydraulic pressure to test their behavior and long-term service under such conditions [32]. Water absorption in composites is undesirable since it increases the chances of mechanical failures due to hydrolysis and the build-up of internal stresses in the composite. Unlike composites reinforced with organic fibers such as wood/bamboo which are organic, the plastic-quarry dust composite is not affected by mold formation and growth which usually increases water absorption [33,34].

Increasing water absorption in a composite material can be a concern, especially for high-pressure applications where moisture ingress can weaken the material over time. To mitigate this issue and make the composite more suitable for specific applications, a number of modifications and coatings options can be considered. Applying hydrophobic coatings to the composite's surface can help repel water and reduce water absorption. These coatings can be based on various materials, such as silicone, fluoropolymers, or specialized hydrophobic treatments. Additionally, a barrier layer can be incorporated within the composite structure to prevent moisture from penetrating the material. This can be achieved by adding a layer of impermeable material, such as a thin film or sheet, between the composite layers. This will act as a barrier to water and enhance the composite's resistance to moisture. A hydrophobic additive can also be incorporated directly into the composite matrix. These additives, such as hydrophobic fillers or surface-modifying agents, can reduce the material's affinity for water. Finally, in some applications, the composite material encapsulated within a



Fig. 4. Images of quarry dust when viewed using Optical Microscopy (a) showing various colors possibly due to different mineral compositions and (b) showing various shapes and sizes of particles.



Fig. 5. Images of quarry dust when viewed using SEM with (a) showing various sizes of the quarry dust particles and (b) showing sharp-edged particles.

Table 1
EDX analysis with the distribution of elements in the quarry dust sample at the region where SEM images were taken.

Element	Ord. Z.	Net	Mass%	Norm. Mass	Atom	Abs error [%] (1 sigma)	Rel. error [%] (1 sigma)
Oxygen	8	45,842	61.75	52.44	60.43	7.35	11.91
Silicon	14	78,168	20.95	17.79	11.68	0.92	4.41
Carbon	6	2819	12.5	10.62	16.3	2.22	17.73
Aluminum	13	31,623	10.44	8.87	6.06	0.53	5.08
Iron	26	4494	3.76	3.2	1.06	0.14	3.8
Magnesium	12	8352	3.33	2.83	2.15	0.22	6.49
Potassium	19	4100	1.55	1.32	0.62	0.08	5.17
Calcium	20	3212	1.41	1.2	0.55	0.08	5.35
Sodium	11	2279	1.38	1.17	0.94	0.13	9.14
Titanium	22	1183	0.68	0.58	0.22	0.05	7.95
		SUM	117.76	100	100		



Fig. 6. Water absorption for HDPE- quarry dust composite samples exposed to atmospheric pressure.

waterproofing material or enclosure. This can be an effective way to protect the composite from moisture. For example, in the case of pressure vessels, an external layer of waterproofing material can be applied.

Shore D hardness

Fig. 10 shows the Shore D Hardness values of the polymer-quarry dust composite samples with varying amounts of quarry dust. A



Fig. 7. Water absorption for PP- quarry dust composite samples exposed to atmospheric pressure.



Fig. 8. Water absorption for HDPE- quarry dust composite samples exposed to a pressure of 6 bars.



Fig. 9. Water absorption for PP- quarry dust composite samples exposed to a pressure of 6 bars.

positive impact on the Shore D hardness is seen on both HDPE and PP polymer composites when quarry dust is used as a reinforcing agent of the polymer matrix. The Shore D hardness increases linearly with increasing quarry dust. For HDPE, the Shore D hardness improves from 66 to 82 with the addition of 80 % wt quarry dust. A similar observation is reported for PP, where the Shore D hardness value improves from 71 to 82 with the addition of 80% wt quarry dust. The linear increase in hardness is attributed to the increase in the concentration of quarry dust which is hard due to high silica content. Nevertheless, due to the high hardness value of PP compared to that of the HDPE, the hardness values of the PP composites were generally higher than those of the HDPE composites. When 80% wt composition of quarry dust is used, both HDPE and PP composites exhibit the same hardness value of 82.

Charpy impact strength

Fig. 11 shows the variation of impact strength with varying amounts of quarry dust in PP and HDPE. Comparing the PP and HDPE



Fig. 10. Shore D Hardness value on polymer-quarry dust composite samples with varying amounts of quarry dust.



Fig. 11. Charpy Impact strength for the HDPE/PP-quarry dust composite samples produced at various proportions by weight of the quarry dust.

results, it is observed that (i) 5 % weight of quarry dust in the composite tends to increase the composite materials' impact strength to 69.4 kJ/m^2 for PP and 33.0 kJ/m^2 for HDPE and (ii) composites that are based on PP possess a higher value of the impact strength compared to the HDPE-quarry dust composite. They may, therefore, have a more versatile set of applications. The reduction in impact strength with a further increase in quarry dust concentration beyond 5 wt.% quarry dust is attributed to the presence of an increased number of voids in the composite [15]. Additionally, the sharp-edged particles observed in the quarry dust sample in Fig. 6 act as crack propagators, unlike spherical particles which act as crack inhibitors [35]. Hence, an increase in the amount of quarry dust deteriorates the composite's mechanical properties. The impact strength can be increased by preheating the quarry dust and by incorporating impact modifiers and interfacial modifiers in the samples produced at high quarry dust concentrations [36–38]. Impact modifiers are materials that are added to polymer composites to improve their toughness and durability, for example, low molecular weight hydroxyl-terminated natural rubber (HTNR), graphene oxide (GO), TiO₂, Acrylic, etc. [39–44].

Figs. 12 and 13 show the SEM images of fracture surfaces of the impact strength test samples prepared using HDPE and PP



Fig. 12. SEM images of fracture surfaces of impact strength tested samples of (a) pure HDPE and (b) HDPE+80 wt.% quarry dust.



Fig. 13. SEM images of fracture surfaces of the impact strength tested samples of (a) pure PP and (b) PP+80 wt.% quarry dust.

respectively. In SEM analysis, the secondary electron imaging (SEI) mode usually displays darker regions which means few secondary electrons were collected from that region because they are obscured from the detector due to the roughness of the surface. Therefore, valleys tend to appear darker and ridges tend to appear brighter. Therefore, the valleys could be the regions where the polymer matrix did not bond well to the quarry dust particles therefore creating porosity-like defects and crack-like defects. However, no agglomeration was observed which implies that homogenization was achieved.

Conclusion

To counter the negative effects to the environment caused by plastic pollution and quarry dust damping, it is necessary to come up with methods that can help utilize these materials. One of the methods is coming up with products that use the waste plastics and quarry dust as raw materials. This study mixed different proportions of PP/HDPE and quarry dust and drew the following conclusions.

- The polymer roof tile having 60 % quarry dust in the PP polymer matrix had the highest MFI value (24.5 g/10 min) hence easy to process.
- The SEM images indicated that the smaller-sized particles (about 10 µm) of quarry dust strongly adhered to the polymeric matrix. It was observed that the quarry dust particles were well distributed inside the polymers.
- Generally, water absorption was relatively high in all composites having 60 wt.% and 80 wt.% of quarry dust with samples exposed to a pressure of 6 bar having comparatively high-water absorption than those at atmospheric pressure.
- The Shore D hardness increases with an increase in the amount of quarry dust as a result of increasing hard particles in the quarry dust with the highest Shore D hardness value of 82 was attained for samples having 80 wt.% quarry dust.
- For the impact strength, only 5 wt.% composition of quarry dust in both polymer composites had a higher value of Charpy impact strength as compared to pure polymer with 69.4 kJ/m^2 for PP+5 wt.% quarry dust being the highest. The sharp-edged particles and the void defects reduced the impact strength.

Credit author statement

All the materials and information obtained from the literature have been referenced accordingly as per the IEEE referencing system. In this way, the authors have been credited for their works.

Declaration of Competing Interest

The authors declare no conflict of interest.

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