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Transforming Granite Wastes into High Performance Hybrid Polymer Composites for Environmental Sustainability and its Comparative Optimization using TOPSIS Technique

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Abstract

This research work aims to examine the opportunity of reexploiting granite wastes to get inventive vinyl ester based hybrid polymer composites. The foremost advantage of recycling the granite wastes is low cost and eco-friendly production. Granite wastes generated by stone processing units are generally dumped in open lands, which creates severe environmental complications causing soil, air and water pollution. Hence, the inculcation of such kind of industry wastes into the polymer fabrication process can be a serious solution for sustainable waste utilization. The examined mechanical properties of the fabricated granite dust filled hybrid composites are compared with Titania particulate filled E-glass fiber reinforced polymer composites. Conventional hand layup technique is prominently exploited for the fabrication of composite specimens. The granite dust/titania weight fraction is varied from 5-15 wt. % while keeping bi-directional E-glass fiber weight fraction as constant i.e., 30 wt. %. Density and percentage void content has been determined and also mechanical characterization such as hardness, tensile strength, flexural strength, impact strength and inter laminar shear strength of the produced polymer composite material has been carried out. As far as incorporation of granite dust is concerned, there has been a significant improvement in the physical as well as mechanical characteristics of the fabricated composite material. Also, to determine the best proxy situation from a finite lot of decision proxies in terms of various oppugning criteria, TOPSIS decision making approach is used.

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Keywords: Granite dust, Sustainability, Titania, Hand layup, Mechanical characterization, TOPSIS.

I. INTRODUCTION

The development of particle-filled polymer composites reinforced with fibers is the need for structural components that are prone to fracture and cracks by utilization of stone waste such as granite dust. Granite dust would be a solution not only to control health issue but also pollution control such as soil, air with reduction on the dependency on depicted natural resources [1]. Therefore, reusing the waste is considered to be the most important solution in the modern world. For composites, the materials are mixed to reduce shortages and increase characteristic performance of the materials. The overall economy of these composites are found to be very low when industrial waste is added to the matrix as reinforcement [2]. Since, traditional fillers such as metals and ceramic powders possess high cost, it is important to replace traditional composite ceramics with other cheap and abundant wastes such as granite, silica [3] and boron [4] that are available in abundant form without embracing quality. Granite dust is an industrial waste material that can be used as a filler in a polymer matrix [5].

Granite dust is a combination of different oxides with better mechanical and wear properties [6]. Vikash et.al. [7] in their work concluded that with the addition of granite particles, the aluminum alloy composites has improvised hardness and desired strength. Awad et.al. [8,9] studied the influence of particle sizes and % content of marble/granite filler on the physico-mechanical properties and observed that the addition of M/G fillers in the HDPE composites outstripped its thermal and mechanical properties. Similarly, the use of Granite dust [10] along with baggage ash [11] and copper scrap [12] for concrete modification is quite common among the researchers. Also, the influence of granite dust from the fold and thrust belt [13] on the physicomechanical properties of mortar is studied and inferred that incorporation of granite powder in mortar leads to an increase in the diffusion properties of fluids due to significant improvement in porosity [14].

In series to this, hybrid polymer composites is prominently a varied kind of composite material consisting of a polymer matrix and two types of reinforcements: fibers and particles. The fiber reinforcement provides rigidity and longitudinal strength, and the particles provides tensile strength and impact resistance. One such work included graphite as a filler and E-glass fibre as a reinforcing material to fabricate hybrid VE based composites [15]. A review has also been performed on the mechanical properties of the polymer composites in terms of hybridization [16] where it was inferred that it possess better strength-to-weight ratio than most of the conventional alloys [17]. Similarly, studies on the mechanical properties of the hybrid polymer composites [18,19] with a dammar based hybrid matrix [20], based on natural fibers [21] such as animal-plant fiber hybrid composites [22] has also been performed.

Along with composite processing and mechanical assessment, optimization of composite criteria is also very important as far as the minimization of experimental setup and identification of parameters that most influence the characterization of the fabricated composites is concerned [23]. In this order, a multicriteria decision making approach, such as TOPSIS, has been performed by various researchers in their study. This MCDM method is found to be very crucial tool for the decision makers to choose an alternative in discrete problems [24]. Especially, TOPSIS has become much easier for the users with the help of computers. Various articles shows the use of decision making TOPSIS model for finding the optimal reinforcements in CaCO3 and Al2O3 composites [25] and natural fiber composites [26].

In this study, vinyl ester resin was used as the matrix material, granite dust/titania particulate was used as filler, and E-glass fiber was used as a reinforcement to obtain titania/granite dust filled glass fiber reinforced polymer composites. Then characterization of the physico-mechanical behavior of the obtained polymer composite material has been prominently performed using TOPSIS technique.

II. EXPERIMENTAL DETAILS

A. Materials and methodology

In the present study, vinyl ester is used for the preparation of polymer composite filled with granite dust/titania particles and reinforced with E-glass fiber through the conventional hand layup method. Vinyl ester as a matrix material imparts magnificent mechanical properties to the composite material. As a reinforcement, E-glass fiber is used which is predominantly a mixture of different oxides (55%SiO₂, 18%CaO, 11%Al₂O₃, 6%B2O3, 5%MgO, and 5% other). Further, titania as a filler material, is an oxide of titanium [27] and granite dust is a combination of different oxides that poses excellent mechanical and wear resistant properties [28]. The notations and chemical compositions of the fabricated vinyl ester composites is shown in table 1.

The composition of vinyl ester is varied in correspondence of the granite dust/titania fillers with three varied weight fractions (5 wt.%, 10 wt.%, 15 wt%) while keeping E-glass fiber weight fraction constant i.e., 30 wt.%. The vinyl ester resin is mixed with



its corresponding hardener in 10:1 weight ratio. Silicon mold releasing spray has been applied to remove the composites from a wooden mold of dimension 340*340*40 mm³ after curing. Before the removal of the composites from the wooden mold, the load of 40 kg is applied for about 24 hours during the curing of the casted composites whereas after the removal of composites from the mold, casted samples are post cured for another 24 hours. The desired size of the specimens for mechanical/physical tests are cut using the diamond cutter.

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Notations	Compositions (by weight)
EG-0	Glass fibre (30%) + Vinyl ester (70%)
EGG-5	Glass fibre (30%) + Vinyl ester (65%) + Granite (5%)
EGG-10	Glass fibre (30%) + Vinyl ester (60%) + Granite (10%)
EGG-15	Glass fibre (30%) + Vinyl ester (55%) + Granite (15%)
EGT-5	Glass fibre (30%) + Vinyl ester (65%) + Titania (5%)
EGT-10	Glass fibre (30%) +Vinyl ester (60%) + Titania (10%)
EGT-15	Glass fibre (30%) + Vinyl ester (55%) + Titania (15%)

B. Physical Characterization

To examine the composite quality, density and void content has to be investigated for physical assessment of the fabricated specimens. For precision assessment, theoretical density and experimental density are obtained separately using weight proportion method and Archimedes principle respectively. The void content is then calculated using a comparative study between the theoretical and experimental densities. For determining the experimental density using the Archimedes principle, several specimens were taken from the obtained composite material and first weighed in air and then weighed in water using electronic weighing machine. The formula for determining the experimental density is:

$$\rho_c = \frac{Wair}{(Wair - Wwater)} * \rho_{water}$$
(1)
Where, ρ_c = density of obtained composite
 ρ_{water} = density of water (gm/cc)
wair = weight of the specimen in air (gm)

wwater = weight of the specimen in water (gm)

Void content of the composites is calculated by densities based on theoretical calculation and experimental investigation using equation:

 $V_{f} = \frac{\rho_{t} - \rho_{e}}{\rho_{t}} * 100 \qquad (2)$ Where, V_{f} = volume fraction of void

 ρ_t = Density of the composite (Theoretical) ρ_e = Density of the composite (Experimental)

C. Mechanical Characterization

In mechanical assessment of the fabricated composites, micro hardness test of the specimen is carried out using 1/16" indenter (steel ball) through the Rockwell hardness testing machine and a preliminary load of 50 kgf. The tensile test has been conducted as per ASTM D3039-3062 using the digital tensometer apparatus. The flexural strength and modulus test has also been carried out for the fabricated granite dust/titania filled vinyl ester based polymers. Inter laminar shear strength of composites based on vinyl ester resin (brittle matrix), is determined using a beam shear test (BST). ASTM D7291 standard has been considered to carry



out inter laminar shear strength test of the obtained vinyl ester based polymer composites on the Materials testing machine. The low speed impact test is also conducted for measuring the damage resistance using the impact tester as per ASTM D256.

D. TOPSIS optimization

TOPSIS (Technique for Order Preference by Similarity to the Ideal Solution) works on the principle of selection of an alternative which should be "closest in terms of distance to the positive ideal solution" and "should be farthest in terms of distance to the negative ideal solution" [29]. This decision making approach has a very straight forwarded, logical and accessible concept which has the inherited function of the relative effectiveness measurement of each option in an understandable mathematical form.

Assume Z = (Zij) be a matrix consisting of "p" proxies and "q" criteria, such that we have $(Zij)_{p^*q}$ matrix. This matrix is known as the decision matrix (D). And also let $\Delta = [\Delta 1, \Delta 2... \Delta n]$ be a weight vector, where Zij, $\Delta j \in R$ and $\Delta 1 + \Delta 2 + + \Delta n = 1$

Criteria can be beneficial i.e., higher is better or Non-beneficial i.e., smaller is better.

At this point, the various dimensional attribute are converted to dimensionless attributes so that the criteria can be compared. Because varied criteria are basically measured in different units, there is a need to convert the results in the Z scoring matrix to a standardized scale. These values may be normalized using one of the very well-known standardized expressions. Some of the most commonly used methods for calculating the normalized value of ij n are:

Weighted normalized matrix (V_{ij}) is the result of multiplying the normalized matrix by its associated weight (Δj) calculated by entropy method.

Where, Δj is the weight assigned to *j*-th criterion, i.e., $\Delta 1 + \Delta 2 \dots + \Delta j = 1$.

For determining the positive ideal solution (A $^+$) and the negative ideal solution (A $^-$) for the normalized matrix based on weights of the criteria, the following expression holds true:

where,

$$\begin{split} V_{j}^{+} &= \begin{cases} \left(\min_{i} V_{ij} \right), \ \text{ if } j \text{ is beneficial criteria} \\ \left(\max_{i} V_{i}^{+} \right), \ \text{ if } j \text{ is Non-beneficial criteria} \end{cases} \ \text{ and} \\ V_{j}^{-} &= \begin{cases} \left(\min_{i} V_{ij} \right), \ \text{ if } j \text{ is beneficial criteria} \\ \left(\max_{i} V_{i}^{+} \right), \ \text{ if } j \text{ is Non-beneficial criteria} \end{cases} \ \text{ for } j = 1, 2 ... N \end{split}$$

Now, by using these positive ideal and negative ideal solutions, separation measures are to be determined.

For this, the distances measured using Euclid geometry of each of the proxies are calculated in terms of the positive ideal and the negative ideal solution:

Finally, the overall relative closeness of the proxies is calculated and then they are arranged in subside relative to their relative closeness and assigned rankings. The overall relative closeness (Pi^*) of the proxies is calculated as:

III. RESULTS AND DISCUSSION

A. Physical Assessment

In the present study, theoretical and experimental densities of the granite dust/titania filled E-glass fiber reinforced vinyl ester based polymer composites is evaluated and the corresponding weight percentage of void content has been examined and represented in the Table 2.

The incorporation of granite dust particles resulted into the increment in density of the composite, thus, increases the percentage of void content. The similar effect is also shown by the titania filled glass fiber reinforced polymer composites with the incorporation of filler content. The void fraction of composites are evaluated from the difference between the expected and the observed density of each sample. This table clearly depicts that with the incorporation of filler particulates, increasing amount of voids are noticed. This may be because of the inner-fiber spacing been decreased. It is clear that a good composite should have a small void fraction. However, the presence of void is inevitable, especially when fabricated through hand lay-up manual technique.

Since void fraction also depends on type of filler used, granite filled polymer composites show lesser percent of void content than that of titania filled composites.

B. Influence of Particulate on Hardness

The hardness of the polymer composite effectively show an increment trend as the weight fraction of the granite dust and TiO2 particulate increases, as shown in fig. 1. The composites (EGT-5, EGT-10 and EGT-15) with titania particulate content is found to exhibit Rockwell hardness in the strict range of 47 HRB for EGT-5 to 71 HRB for EGT-15 while the composites (EGG-5, EGG-10 and EGG-15) with granite dust content exhibits Rockwell Hardness in the range of 43 HRB for EGG-5 to 61 HRB for EGG-15.

The average hardness of granite and TiO2 powder is usually higher like any other type of oxide ceramic and their incorporation increases the hardness of the composite. The minimum hardness is examined for unfilled composite, i.e., EG-0 and maximum hardness is examined for 15 wt.% TiO₂ particulate filled composite material.





Notations	Compositions (by weight)	Calculated density (g/cc)	Formulated Density (g/cc)	Voids content (%)
EG-0	Glass fibre (30%) + Vinyl ester (70%)	2.12	2.23	4.93
EGG-5	Glass fibre (30%) + Vinyl ester (65%) + Granite (5%)	2.23	2.35	5.11
EGG-10	Glass fibre (30%) + Vinyl ester (60%) + Granite (10%)	2.32	2.48	6.45
EGG-15	Glass fibre (30%) + Vinyl ester (55%) + Granite (15%)	2.45	2.62	6.49
EGT-5	Glass fibre (30%) + Vinyl ester (65%) + Titania (5%)	2.35	2.52	6.74
EGT-10	Glass fibre (30%) + Vinyl ester (60%) + Titania (10%)	2.48	2.68	7.46
EGT-15	Glass fibre (30%) + Vinyl ester (55%) + Titania (15%)	2.52	2.76	8.70

Table 2: Theoretical/Experimental densities and % void content with notations



Figure 1 Hardness Variation with filler content

C. Influence of Particulate on Tensile and Flexural Strength

From fig. 2, it is clearly depicted that tensile strength of the hybrid vinyl ester based polymer composites shows a declining trend as far as the incorporation of both titania and granite dust filler is concerned. Such an unexpected behavior of the Bi-directional fiber reinforced composites may be inferred to one of the possible explanation that surface defect and voids have preferably strong impact on the strength of the fabricated composites. The declining trend of the tensile modulus with the incorporation of granite dust/titania particulate in the polymer composites is depicted in the fig. 3. The decrement in the tensile modulus with addition of filler content is may be due of poor adhesiveness of the filler with the matrix material and void formation at the filler – matrix interface.

Fig. 4 prominently indicates that flexural strength of the composites is a function of filler content, i.e., with the addition of the TiO2/granite dust , the flexural strength of the fabricated material composite increases. The formation of micro cracks during the formation of the composites through hand lay-up technique, results in the stress release over a large area provided by the flexural loading, leads to increment in the flexural strength.



The flexural modulus is found to be showing increasing trend with granite dust/titania addition in the vinyl ester produced composites (fig. 5).



Figure 2 Tensile strength Variation with filler content



Figure 3 Tensile modulus Variation with filler content



Figure 4 Flexural strength Variation with filler content



Figure 5 Flexural modulus Variation with filler

D. Influence of Particulate on Impact Strength

The determination of overall energy absorbed during the fracture of the granite dust/titania filled vinyl ester composites is carried out using a low velocity impact tester as per ASTM D256. The outcome of the impact strength test is shown in fig. 6. The graph clearly infer the fact that there is a serious positive change in the damage resistance or impact strength of the fabricated composite with the addition of filler content, i.e., TiO2 particulates as well as granite dust.

It is seen from the test graph that with the incorporation of granite content, the impact strength of the unfilled vinyl ester polymer composite improves effectively by about 10-20% as expected. High deformations loads are required for many structural composite and marine applications. Therefore, the usability of a composite material for such applications must be identified not only by its general design parameters, but also by its energy absorption capacity.



Figure 6 Impact strength Variation with filler

E. Influence of Particulate on ILSS

Laminated composites are most susceptible to stratification between the fiber/matrix interfaces to which a tangential stress is applied. Inter laminar shear stress strongly depends on the matrix prioritized mechanical properties in the construction of laminated composite materials. ILSS shows significant improvement from 14.76 MPa for EG-0 composites to 16.67 MPa and 16.49 MPa for EGT-10 and EGG-10 respectively, and then declines to 15.69 MPa and 15.37 MPa for EGT-15 and EGG-15 composite specimen respectively.



Titania particulate Granite dust

Figure 7 Variation of inter laminar shear strength with filler

The possible reason behind this behaviour can be attributed to high void content for 15 wt.% granite dust/titania powder filled vinyl ester composites. From fig. 7, it is clearly depicted that inter laminar shear strength shows variation for both granite dust/titania particulate in a very identical fashion as reportedly observed by Patnaik et. al. [30] for fly-ash and SiC filled glass reinforced polyester based composites.







F. TOPSIS Optimization Assessment

The TOPSIS method is an empirical and convenient method for selection of alternatives and identification of their respective rankings. Due to the high flexibility of the TOPSIS concept, additional extensions for better choice in different situations is now possible. In fact, TOPSIS is used to solve many theories and the real world issues.

In this work, all the fabricated granite dust filled composite materials are compared with titania particulate filled composites based on TOPSIS technique. For selection of individual alternatives and identification of their rankings, the decision matrix (Table 3), the normalized matrix (Table 4), the weighted normalized matrix (Table 5), positive and negative ideal solutions (Table 6), separation measured values (Table 7), relative closeness values and finally the rankings (Table 8) for the respective alternatives are tabulated below.

Table 3. Decision Matrix (D) Of Fabricated Composites

Composites	Hardness (HRB)	Impact stren. (kJ/m2)	Void con. (%)	Tensile stren. (MPa)	Tensile modu. (GPa)	Flexural stren. (MPa)	Flexural modu. (Gpa)	ILS stren. (Mpa)
0	30	21.51	4.93	540	8.92	330	6.99	14.76
G05	43	23.63	5.11	515	8.7	359	7.27	15.61
G10	58	30.46	6.45	490	8.52	373	7.59	16.49
G15	67	32.81	6.49	475	8.39	392	7.98	15.37
T05	47	26.34	6.74	520	8.76	363	7.31	15.81
T10	61	34.57	7.46	505	8.67	380	7.67	16.67
T15	71	34.91	8.7	483	8.44	397	8.03	15.69

Table 4. Normalized Matrix

Composites	Hardness (HRB)	Impact stren. (kJ/m2)	Void content (%)	Tensile stren. (MPa)	Tensile modu. (GPa)	Flexural stren. (MPa)	Flexural modu. (Gpa)	ILS stren. (Mpa)
0	0.204252	0.274666	0.279596	0.404603	0.39065	0.336044	0.34961	0.353468
G05	0.292761	0.301737	0.289804	0.385872	0.381015	0.365575	0.363615	0.373823
G10	0.394887	0.388951	0.3658	0.36714	0.373132	0.379832	0.37962	0.394897
G15	0.456162	0.418959	0.368068	0.355901	0.367439	0.39918	0.399126	0.368076
T05	0.319995	0.336342	0.382246	0.389618	0.383643	0.369648	0.365615	0.378613
T10	0.415312	0.441433	0.42308	0.378379	0.379702	0.38696	0.383621	0.399208
T15	0.483396	0.445774	0.493404	0.361895	0.369629	0.404271	0.401627	0.375739





Composites	Hardness (HRB)	Impact stren. (kJ/m2)	Void content (%)	Tensile stren. (MPa)	Tensile modu. (GPa)	Flexural stren. (MPa)	Flexural modu. (Gpa)	ILS stren. (Mpa)
0	0.097374	0.059151	0.067803	0.005138	0.001147	0.00786	0.005554	0.003709
G05	0.139569	0.064981	0.070279	0.0049	0.001119	0.00855	0.005777	0.003923
G10	0.188256	0.083763	0.088708	0.004662	0.001096	0.008884	0.006031	0.004144
G15	0.217468	0.090225	0.089258	0.00452	0.001079	0.009336	0.006341	0.003863
T05	0.152552	0.072433	0.092697	0.004948	0.001127	0.008645	0.005809	0.003973
T10	0.197993	0.095065	0.102599	0.004805	0.001115	0.00905	0.006095	0.004189
T15	0.230451	0.096	0.119653	0.004596	0.001086	0.009455	0.006381	0.003943

Table 5. Weight Normalized Matrix

Table 6. Best & Worst Solutions

Solution	Hardness (HRB)	Impact stren. (kJ/m2)	Void content (%)	Tensile stren. (MPa)	Tensile modu. (GPa)	Flexural stren. (MPa)	Flexural modu. (Gpa)	ILS stren. (Mpa)
Positive Ideal Solution(A+)	0.230451	0.096	0.067803	0.005138	0.001147	0.009455	0.006381	0.004189
Negative Ideal Solutions (A-)	0.097374	0.059151	0.119653	0.00452	0.001079	0.00786	0.005554	0.003709

Table 7. Separation Measures of Attributes

Composites	Dj+	Dj-	
0	0.138098	0.051853	
G05	0.096069	0.065215	
G10	0.048661	0.099118	
G15	0.025744	0.12773	
T05	0.085114	0.062838	
T10	0.047597	0.108199	
T15	0.051853	0.138097	

IV. CONCLUSION

Some of the conclusions were drawn through mechanical experimentation as below:-

1. The granite dust filled E-glass fiber reinforced vinyl ester based polymer composites are fabricated successfully using the hand lay-up fabrication technique and compared with the

Table 8. Relative Closeness and Composite Ranking

Composites	Pi*	Rank
0	0.272983	7^{TH}
G05	0.404348	6 TH
G10	0.670718	4 TH
G15	0.83226	1 ST
T05	0.42472	5 TH
T10	0.694491	3 RD
T15	0.727017	2 ND

experimentation results of titania particulate filled E-glass fiber reinforced hybrid polymer composites.

2. It was inferred that with the inclusion/incorporation of granite dust as well as titania particulates in the polymer composites, the mechanical properties, i.e., density and void content, hardness, flexural strength and modulus, impact strength and ILSS shows a significant improvement in the characteristics.





3. Incorporation of granite dust/titania particulate only affected tensile strength and modulus in the negative declining fashion.

4. TOPSIS decision making approach has been predominantly employed to determine the ranking of the fabricated polymer composites and it was inferred that granite dust filled hybrid polymer composite with 15% wt. is ranked 1ST and unfilled polymer composite specimen is ranked 7TH among all the composite specimens.

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