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Research Article

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Special Issue

A Themed Issue in Honour of Professor Clement Uche Atuanya on His retirement.

This themed issue pays tribute to Professor Clement Uche Atuanya in recognition of his illustrious career in Metallurgical and Materials Engineering as he retires from Nnamdi Azikiwe University, Awka. We celebrate his enduring legacy of dedication to advancing knowledge and his impact on academia and beyond through this collection of writings.

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Quadratic model for evaluating the hardness of hazelnut shell-polystyrene composite based on input concentration ratio of its constituent materials

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Abstract
A quadratic model was derived for evaluating the hardness of hazelnut shell–polystyrene composite (HPC) based on input concentration ratio of the constituent materials. The derived empirical model: Ꜧ = - ẞ (ɤ$_f$ /ɤ$_m$)^N + ɧ(ɤ$_f$ /ɤ$_m$) + Ꝋ validated by the core model : Ꜧ + ẞ(ɤ$_f$ /ɤ$_m$)^N ≈ Ꝋ + ɧ(ɤ$_f$ /ɤ$_m$), in that both sides of the structure are correspondingly near equal. Ꜧ, ẞ and ɤ denote the HPC hardness, input concentrations of hazelnut shell and polystyrene. The model-predicted results emphasize similar trend and spread of result points shown in previous research. Correlations between the HPC hardness and input concentration ratio of hazelnut shell and polystyrene were evaluated as 0.99998 and 0.9787 using model-predicted and experimental results. The standard error incurred on predicting the HPC hardness (relative to experimental results) is < 0.3%, for every change in the input concentration ratio of hazelnut shell and polystyrene. This translates to a model confidence level above 99%. The HPC hardness per unit input concentration ratio of hazelnut shell and polystyrene were 6.8215 and 6.9557, using experimental and model-predicted results. The maximum deviation of the model-predicted HPC hardness from experimental results was 2.36%. The derived model shows an exceptional level of functionality, and will predict the HPC hardness, within the experimental results range, on substituting into the model, values of the input concentration ratio of hazelnut shell and polystyrene, providing the boundary conditions are considered.

Keywords: Hardness; Hazelnut shell; polystyrene composite; constituents input concentration ratio

1. Introduction

The mounting global environmental and social concern, high rate of decline in petroleum resources and novel environmental policy have engendered the search for green composite materials which are environmentally friendly. Green sustainable materials with inexpensive, which are benign, and will not be lower in attributes to manmade, synthesized products; they will assist to preserving the biological equilibrium in the earth and unravel the challenge of their discarding (Ali et al., 2018; Briassoulis et al., 2021). "Green composites" are made by blending materials derived from plastic matrices and reinforcements obtained from agro wastes such as shells, husks, fibers, and kernels; it is a move in the direction of substituting artificial fibers with naturally derived fibers (Lyubushkin et al., 2023; Luthra et al., 2019).

Hardness values are conventionally read off from laboratory equipment. No existing mathematical expression or model has calculated the hardness of the highlighted composite based on the input concentration ratio of the constituents. This formed the basis for the present work to fill in the gap. Of special interest are agricultural and industrial wastes which are discarded in huge volumes and are not recycled in other sectors (Kuram 2022), whilst this proffers ecological and economic advantages (Liu et al., 2017). In the approach of a closed-loop economy, the chance of reusing agro wastes is of special significance (Tudor et al., 2020; Fan et al., 2019). They can confer composite structures the required pleasing look and modify and position some properties, as well as providing for the protection of the trees and keeping a lofty standard of physical and mechanical attributes of the produced materials, due to in certain fields in the future the appeal may surpass the availability of this resource, and its replenishing requires many years (Risse et al., 2017; García et al., 2020). Consequently, composites derived from agro wastes are increasingly becoming fashionable (Haider et al., 2018; Cherkashina et al., 2023a; An et al., 2023; Mubofu 2016), as they substitute costly materials, as well as keeping lofty quality features, and are applied in diverse areas (Lengalova et al., 2016; Feig et al., 2018) such as food packaging (Mangaraj et al., 2019; Thakur et al., 2021; Rydz and Musioł 2022), in the aerospace and automotive manufacturing to lower the mass of some interior components (Elseify et al., 2021; Kamal et al., 2017), in building industries to lower cost and to produce novel, biodegradable materials; and to some extent restore fine filler in concrete and more fields of manufacture (Cherkashina et al., 2023b; Rai et al., 2021; Ceraulo et al., 2022).

Hazelnut shells are obtainable in large volumes at the gathering in agricultural crops. They are a residue, an agro waste material that can be utilized as a sustainable raw material for manufacture. Hazelnut shells are a flexible, sustainable material that can be employed for the production of very good quality products and agro-based plastics, as well as possessing a great hardness and impact resistance. Globally, in the gathering in period the hazelnut produce is up to five hundred and thirty thousand tons (García-García et al., 2015). The shells are very nearly 70% of the entire mass and are of no usefulness or importance. Because the hazelnut shells are discarded as waste, researchers are yet to unearth the complete potential of this agro-waste to date. It is employed as a warmth source, as a solid renewable fuel, as well as a crude material for the synthesis of furan-2-carbaldehyde (Barbu et al., 2020; Aliotta et al., 2024; Guerrero et al., 2019), while the sphere of its utilization can be remarkably increased. In the report (Balart et al., 2018), hazelnut shells were utilized as filler for a composite with the incorporation of poly (lactic acid). Their thermal, mechanical and physical characteristics were investigated. The results indicated that the inclusion of hazelnut shells had minimal influence on the outcome, still surged the crystallinity index, which drastically affects the volume sturdiness of the composite.

In certain work (Balart et al., 2016) included epoxidation of linseed oil to the plastic substrate and hazelnuts to lower the plasticity index that normally boosts the entire attributes of the composite with poly (lactic acid) and hazelnut shells. In their research (Aliotta et al., 2021) investigated a developed composite involving PLA3251D with the inclusion of hazelnut shells; the ensuing material presents reduced viscosity in the melt phase and a lowered degree of tensile strength. Pradhan and Satapathy (2022) evaluated the physico-mechanical properties of composites using walnut shell filler. Notwithstanding the increased content of walnut shells, density and porosity properties of the composites increased. Kufel and Kuciel (2020) reported physic-mechanical attributes of mixed composites of polypropylene with the combination of hazelnut shells/maleic anhydride/basalt fiber. The ensuing material is distinguished by enhanced physical and mechanical features, whereas reduced moisture absorption.

Ceraulo et al., (2022) studied the rheological properties of Bi® El51N0 polyester composites with the inclusion of hazelnut shells, mostly their mechanical and morphological properties, to achieve biodegradable bio-composites. At average concentrations, the stiffness and impact toughness of the material was increased, and the bio-composite is easily reusable. Despite the fact that there abound numerous researches investigating this subject, in majority of them a comparatively small amount of hazelnut shells is added owing to its undesirable similarity to other components of the polymer matrix. Polystyrene (PS) is extensively utilized in the building business in soundproofing structures and clatter-absorbing screens, in therapeutics, in service abilities, and in the meal industry. Polystyrene is recognized to be non-biodegradable (Tsochatzis et al., 2021). Thus, it is propitious to apply it for composite materials, for that; against, inertness to non-decomposition in natural environments is vital. For instance, using polystyrene, it is favorable to develop composite materials that can be employed to make construction structures for sports and garden furniture.

Benchouia et al., (2022) have produced a novel encasing material derived from date palm fibers and polystyrene. The substitution of 1/3 of the arrangement with the suggested composite manifested the potential of such implementations as thermal lagging, with a reduction in thermal conductivity nearly 50%. Onifade et al., (2020) reported the likelihood of applying strengthened bio-char from psyllium stalk fibers to strengthen a polystyrene composite. The biodegradability of the composite offers a preferable panacea to agricultural waste disposal instead of incineration. Adeniyi et al., (2022) evolved an innovation for the development of polystyrene composite with saw dust. Upon the incorporation of saw dust in the content of 30%, an enhancement in the mechanical and thermal properties of the composite was noticed. Barczewski et al., (2019) studied the effects of particle size and filler loading using hazelnut shell, walnut shell and sunflower husks agro-wastes. They found that the best mechanical properties were achieved with the addition of the hazelnut shell powder. Ogah et al., (2024) studied the development of green composites based on castor bean shell (Ricinus communis) as filler in epoxy resin polymer. Their study showed that castor bean shell is viable filler for manufacturing green composites.

Cetin et al., (2023) investigated bio-based sustainable composites from hazelnut shell and PP/SEBS blends. They found that the incorporation of SEBS into PP increased the flexibility of the composites and resulted in relatively higher strain at break. Karakus et al., (2017) studied the utilization of hazelnut shells as filler in LDPE/PP based polymer composites. They found that with increase of filler loading for PP, tensile strength, elongation at break, and flexural strength were decreased while tensile modulus and flexural modulus were increased. Impact strength of PP based polymer composites was not changed. Also, the addition of filler loading into LDPE matrix reduced tensile strength, elongation at break, and impact strength, while flexural strength, modulus and tensile modulus were increased.

Oral et al., (2023) studied the determination of mechanical and damping properties of hazelnut shell powder reinforced bio-composites by ultrasonic method. They found that a significant increase in the density, ultrasonic wave velocities, and elastic modulus values of the bio-composites was seen compared to the neat bio-based epoxy resin. Cherkashina et al., (2023b) shows that HPC hardness recorded during tests was affected by some process parameters such as the input concentrations of hazelnut shell and polystyrene. Regardless of numerous researches on the production of composites derived from polystyrene and plant fillers, there are no studies on the quadratic model for evaluating the hardness of hazelnut shell-polystyrene composite based on input concentration ratio of its constituent materials. However, no existing mathematical expression or model has calculated the hardness of the highlighted composite, based on the input concentration ratio of the constituents. This formed the basis for the present study to fill in the gap. The present study aims at deriving an empirical model that will evaluate the hardness of hazelnut shell-polystyrene composite based on input concentration ratio of the constituent materials. It is expected that the model if derived, shall predict the hardness of the composite within the experimental result range, providing the input parameters are within the boundary conditions.

2.0 Material and methods

2.1Materials

Polystyrene of grade 525 (PJSC Nizhnekamskneftekhim, Nizhnekamsk, Russia) was used as a polymer matrix. Hazelnut shells were used as a filler as shown in figure 1a. Hazelnut shell powder was obtained by grinding crude hazelnut shells in vibratory and planetary mills as shown in figure 1b. Toluene (LLC Component-Reaktiv, Moscow, Russia) was used as a solvent to modify hazelnut shells. The density (g/cm^3), melt flow index at 200^{0}C at 5 kg load g/10 min and melting point (^{0}C) of polystyrene used are 1.12, 9.0 ± 2.0 and 160-170. Moreover, the formula, density (g/cm^3), molar mass (g/mol) and boiling point (^{0}C) of toluene used are C_7H_8, 0.87, 92.14 and 110.6^{0}C (Cherkashina et al., 2023b).

(a) (b)

Figure1: (a) hazelnut shell (b) hazelnut shell powder

2.1.1Hazelnut Shell Modification

Hazelnut shells were ground in WM3 vibrating mill (LLC CONSIT Holding, Moscow, Russia) for 3 min, then in XQM-1An planetary mill (Jiangxi Victor International Mining Equipment Co., Ltd., Shichen, China) for 60 min followed by rinsing with distilled water and drying in a BINDER oven (Binder, Tuttlingen, Germany) for 60 min at 150^{0}C. Then, the ground shells were sieved through a 64 µm sieve. The filler was modified by making a polystyrene coating on its surface so as to obtain a water-repellant finish. Polystyrene, hazelnut shell powder, and toluene were blended in the ratio of 2: 8-48 wt. %: 50-90 wt. %. The composition was incubated for three days. Every 24 hours the composition was treated using ultrasound in ultrasound bath TECHMANN LABORANT L-22 Basic (ODA-Service LLC, Moscow, Russia) having 40 kHz frequency. The resultant solution was kept in an oven for 100-120 min at temperature of 80–95 ^{0}C. Thereafter, the ensuing material was ground in a planetary mill for at about 10 min and sifted using a 64µm sieve (Cherkashina et al., 2023b).

2.1.1.1Preparation of a Composite

Composites on the precise of polystyrene 10 %, 20%, 30%, 40%, and 50% concentrations of treated filler by mass were prepared for the study. Polystyrene granules were initially ground for 3 min. Then, the treated filler and ground polystyrene were blended in a planetary mill for 10 min. The ensuing homogenized blend was loaded into a mold with more heating to 165^{0}C for 60 min. Thereafter, the samples were pressed under pressure of 110 MPa with load endurance for 5 min. The process of hot pressing of the samples allows for shear deformations, which results in a uniform distribution of the filler in the melt.

2.1.1.1.1Model Derivation

Table 1: Variation of HPC hardness with input concentrations of hazelnut shell and polystyrene and their ratios

(Ꜧ)	($ɤ_f$)	($ɤ_m$)	($ɤ_f$ / $ɤ_m$)
16.16	10	90	0.1111
17.31	15	85	0.1765
18.45	20	80	0.1875
19.32	25	75	0.3333
20.19	30	70	0.4286
20.07	35	65	0.5385
19.95	40	60	0.6667
18.78	45	55	0.8182
17.60	50	50	1.0000

Computational analysis of the experimental results shown in Table 1, resulted to Table 2 which indicate that;

$$\text{Ꜧ} + \text{⍰}(ɤ_f/ɤ_m)^N \approx \text{ɇ} + \text{ƀ}(ɤ_f/ɤ_m) \quad (1)$$

"Equation 1" is the core model structure

$$\text{Ꜧ} = -\text{ß}(ɤ_f/ɤ_m)^N + \text{ƀ}(ɤ_f/ɤ_m) + \text{ɇ} \quad (2)$$

"Equation 2" is the derived model

The expression in (2) can be re-written as;

$$\text{Ꜧ} = \text{ɇ} - \text{⍰}\left(\frac{ɤ_f}{100} - ɤ_f\right)^N + \text{ƀ}\left(\frac{ɤ_f}{100} - ɤ_f\right) \quad (3)$$

Recalling from (3) that

$ɤ_m = 100 - ɤ_f$ and $ɤ_f = 100 - ɤ_m$

$$Dv = \text{Ꜧ}_m \left[\frac{-\text{Ꜧ}\varepsilon \times 100}{\text{Ꜧ}\varepsilon}\right] \quad (4)$$

Where

Ꜧ_E and Ꜧ_m are flexural strengths of the composite evaluated from experiment and model-predicted results respectively. Correction factor which overcomes the deviation is calculated as the negative of equation (4)

$$C_f = -\left[\frac{\text{Ꜧ}_m - \text{Ꜧ}_\varepsilon}{\text{Ꜧ}_\varepsilon}\right] \times 100 \quad (5)$$

HPC hardness per unit input concentration ratio of hazelnut shell and polystyrene $\text{Ꜧ}^{ɤ}$ was calculated from the expression;

$$Ꜧ\text{ɣ} = Ꜧ/\text{ɣ} \quad (6)$$

Re-written as

$$Ꜧ\text{ɣ} = \Delta Ꜧ/\Delta \text{ɣ} \quad (7)$$

The expression (7), is detailed as

$$Ꜧ\text{ɣ} = Ꜧ_2 - Ꜧ_1/\text{ɣ}_2 - \text{ɣ}_1 \quad (8)$$

Hardness of the hazelnut shell-polystyrene composite (HPC) can be predicted using the empirical model expressed in (2), providing the input concentration ratio of both constituent materials are known. The variables Ꜧ, $ɤ_f$ and $ɤ_m$ are the HPC hardness and input concentrations of Hazelnut shell and polystyrene weight percent respectively. The ratio ($ɤ_f$ /$ɤ_m$) equals ɣ. The derived model is referred to as Nwoye`s Model for evaluating the hardness of Hazelnut shell-polystyrene composite or Nwoye`s HAD-HAPCOM Model. The equalizing constants; ꬴ, ẞ, N and Ƕ are 14.24, 17.0, 2.0 and 20.0 respectively. The Interaction between the variables and these constants ensures that both sides of (2) are of the same units. These constants were generated by a software (Nwoye, 2008). Table 1 reveals the relationship between HPC hardness and input concentration of hazelnut shell and polystyrene as well as their ratios. The values of the mechanical property were calculated using experimental results and conventional formula.

3.0 Results and Discussions

3.1 Boundary and Initial Conditions

Consider particles of hazelnut shell, interacting with the matrix; polystyrene. The HPC hardness is affected by the input concentrations of hazelnut shell and polystyrene. The considered range of HPC hardness, hazelnut shell, polystyrene and their ratios are 16.16-20.19, 10-50%, 50-90% and 0.1111 – 1.0% respectively.

Table 2: Variation of Ꜧ + ẞ($ɤ_f$ /$ɤ_m$)^N with ꬴ + Ƕ($ɤ_f$ /$ɤ_m$)

Ꜧ + β($ɤ_f$ /$ɤ_m$)^N	**ꬴ + Ƕ($ɤ_f$ /$ɤ_m$)**	**Differential**
16.3691	16.4620	0.0929
17.8404	17.7700	-0.0704
19.5125	19.1400	-0.3725
21.2087	20.9060	-0.3027
23.3129	22.8120	-0.5009
25.0000	22.8120	0.0100
27.5065	27.5740	0.0675
30.1615	30.6040	0.4425
34.6000	34.2400	-0.3600

3.2 Model Validity

The validity of the derived empirical model in (2) is basically rooted in the core model structure expressed in (1), in that both sides of the model structure are correspondingly almost equal. Table 2 is the numerical verification of the model structure, having been generated through evaluation of experimental results in Table 1. The evaluated differentials between the corresponding sides of the structure components as shown in Table 2, amply highlights the functionality of the derived model. The derived model was also validated by comparing the predicted results with the experimental, through graphical, statistical and deviational analysis.

3.2.1 Graphical Analysis

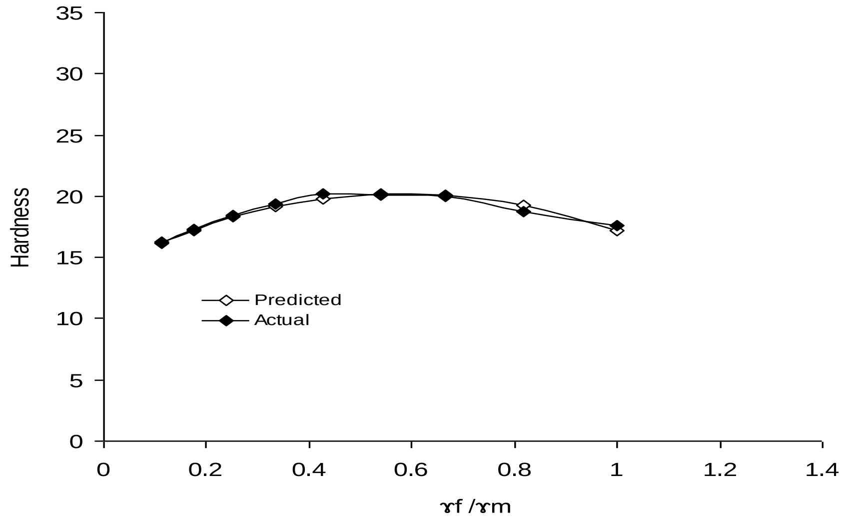

Figure 2: Comparison between HPC hardnesses (relative to input concentration ratio of hazelnut shell and polystyrene) and as evaluated from actual results and derived model.

Figure 2 shows highly aligned and fitted curves of HPC hardness, relative to the input concentration ratio of hazelnut shell and polystyrene, representing experimental and model-predicted results. These curves show very similar trend, spread of result points and almost equal corresponding point values. Furthermore, the curves are all quadratic in nature and will prompt positive or negative slopes, depending on result points considered in the evaluation.

3.2.2 Statistical Analysis

Figure 3 shows the coefficient of determination between HPC hardness and input concentration ratio of hazelnut shell and polystyrene as evaluated from actual results and derived model.

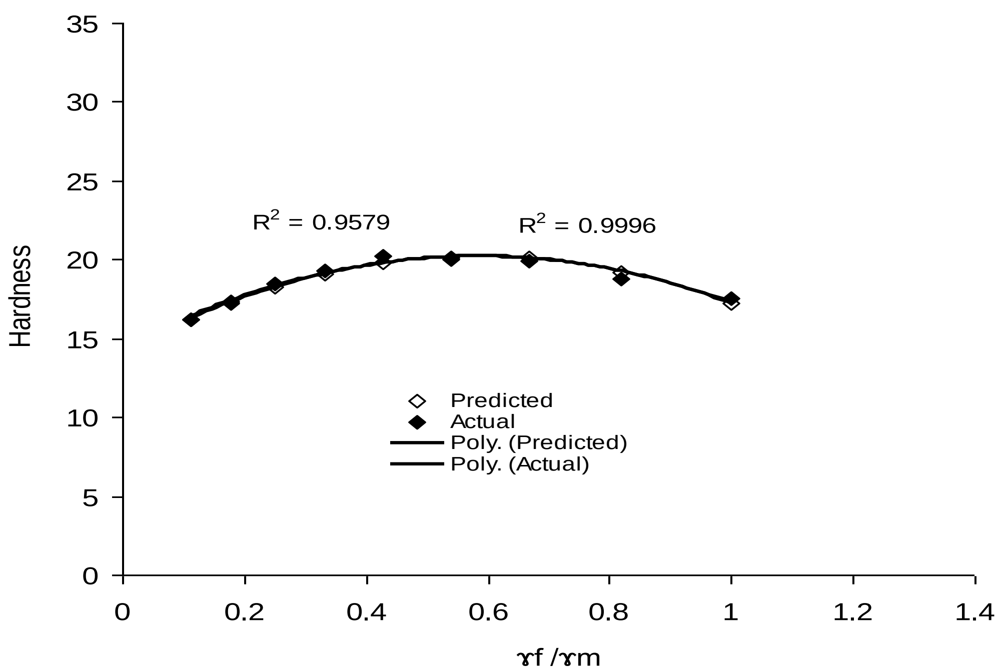

Figure 3: shows the coefficient of determination R^2, which was generated from a plot of HPC hardness against input concentration ratio of hazelnut shell and polystyrene.

The correlations evaluated from these coefficients, between the highlighted variables are 0.99998 and 0.9787 using model-predicted and experimental results respectively. The standard error incurred on predicting the HPC hardness (relative to experimental results) is < 0.3%, for every change in the input concentration ratio of hazelnut shell. This translates in a model confidence level above 99%.

3.2.3 Deviation Analysis

Table 3: Differential between experimentally determined and model-predicted HPC hardness during service

(Ƴ)	**ΔƳ = ƳM - Ƴε**
16.6	0.0929
17.31	-0.0704
18.45	-0.1725
19.32	-0.2027
20.19	-0.4009
20.07	0.1100
19.95	0.1675
18.78	0.4425
17.60	-0.3600

Table 3 reveals the corresponding differentials between model-predicted and experimentally determined HPC hardness. It is shown that some differential are negative and positive, indicating decrease and increase in the model-predicted results respectively, relative to those from the experiment. Overall analysis of the table also emphasizes the derive model functionality. It is therefore very instructive to state that the experimentally determined HPC hardness can be equated to the model-predicted values since the maximum differential between both results is less than 0.5.This is quite negligible, considering the magnitude of both hardness results.

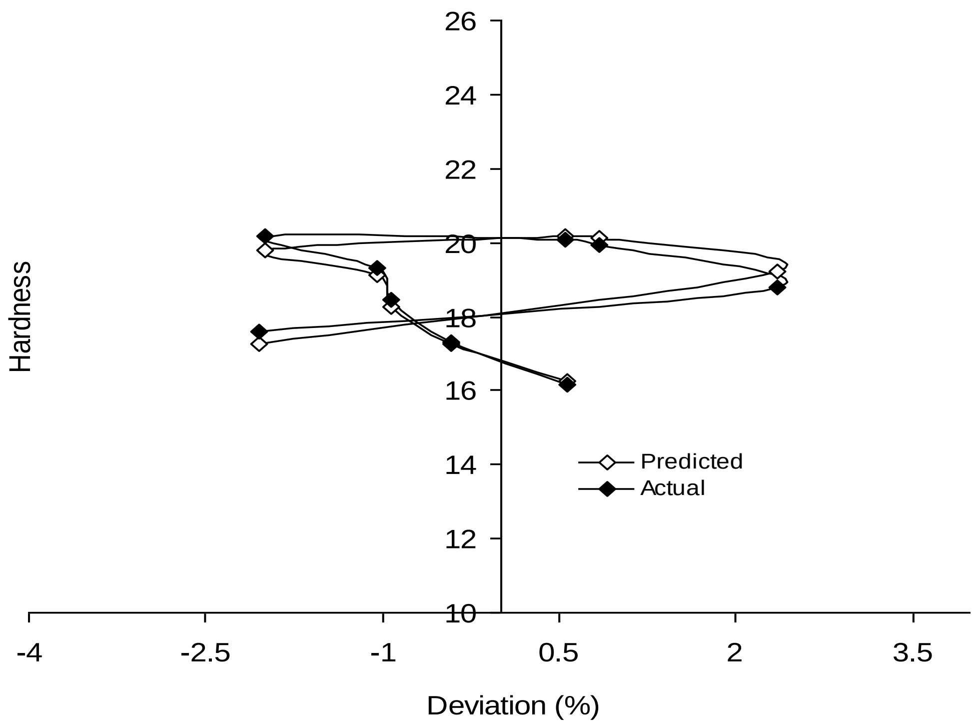

Figure 4: Variation of model-predicted HPC hardness with its corresponding deviation from experimental results.

Figure 4 shows the deviation of model-predicted HPC hardness, for each corresponding experimental results. Comparative analysis of curves from the figure indicates that maximum deviation of model-predicted HPC hardness is 2.36%. This confers on the derived model an operational confidence levels above 97%. The figure also shows that the least and highest deviations of the model-predicted HPC hardness are - 0.41 and 2.36% respectively. These correspond

to HPC hardness: 17.2396 and 19.2225 and input concentration ratio of hazelnut shell and polystyrene: 0.1765 and 0.8182 respectively. The overall model confidence level therefore places between 97 and 99%, considering evaluations from the correlations, standard error and model deviation.
The deviation Dv, of model-predicted HPC hardness from the corresponding experimental result was evaluated from the expression.

$$Dv = Ꞗ_m \left[\frac{-ꞖE \times 100}{ꞖE}\right] \quad (4)$$

Where

$Ꞗ_E$ and $Ꞗ_m$ are flexural strengths of the composite evaluated from experiment and model-predicted results respectively. Correction factor which overcomes the deviation is calculated as the negative of equation (4)

$$C_f = -\left[\frac{Ꞗ_m - Ꞗ_E}{Ꞗ_E}\right] \times 100 \quad (5)$$

HPC hardness per unit input concentration ratio of hazelnut shell and polystyrene $Ꞗ^{ɣ}$ was calculated from the expression;

$$Ꞗɣ = Ꞗ/ɣ \quad (6)$$

Re-written as

$$Ꞗɣ = \Delta Ꞗ/\Delta ɣ \quad (7)$$

The expression (7), is detailed as

$$Ꞗɣ = Ꞗ_2 - Ꞗ_1/ɣ_2 - ɣ_1 \quad (8)$$

Where
$\Delta Ꞗ$ = Change in the HPC hardness $Ꞗ_2$, $Ꞗ_1$ at two input concentration ratios of hazelnut shell and polystyrene $ɣ_2$, $ɣ_1$.
Plotting points (0.1111, 16.16) and (0.6667, 19.95) and (0.1111, 16.2529) and (0.6667, 20.1175) shown in Figure 2, designated as ($ɣ_1$, $Ꞗ_1$) and ($ɣ_2$, $Ꞗ_2$) for experimental and derived model results, and substituting them into the expression (8), gives the slopes: 6.8215 and 6.9557, as their respective HPC hardness per unit input concentration ratio of hazelnut shell and polystyrene.

4.0. Conclusion

A quadratic model was derived for evaluating the hardness of hazelnut shell –polystyrene composite (HPC) based on input concentration ratio of the constituent materials. The derived empirical model; $Ꞗ = -\ ß\ (ɤ_f/ɤ_m)^N + ƅ(ɤ_f/ɤ_m) + Ꞓ$ anchored its validity on the core model structure; $Ꞗ + ß(ɤ_f/ɤ_m)^N \approx Ꞓ + ƅ(ɤ_f/ɤ_m)$, in that both sides of the structure are correspondingly near equal. Correlations between the HPC hardness and input concentration ratio of hazelnut shell and polystyrene were 0.99998 and 0.9787 using model-predicted and experimental results respectively. The standard error incurred on predicting the HPC hardness (relative to experimental results) is $< 0.3\%$, for every change in the input concentration ratio of hazelnut shell and polystyrene which are filler and matrix respectively. This translates in a model confidence level above 99%. The HPC hardness per unit input concentration ratio of hazel shell & polystyrene were 6.8215 and 6.9557, using experimental and model-predicted results. The maximum deviation of the model-predicted HPC hardness from experimental results was 2.36%. Based on the foregoing, the derived model shows an exceptional level of functionality, and will predict the HPC hardness, within the experimental results range, on substituting into the model, values of the input concentration ratio of hazelnut shell and polystyrene, providing the boundary conditions are considered.

5.0 Recommendation

The authors recommend further studies should be carried out to derive mathematical models for the tensile and flexural strengths and moduli of hazelnut shell-polystyrene composite based on the input concentration ratio of the constituent materials.

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Nomenclature

Ꞙ= Hazelnut Shell-Polystyrene Composite Hardness
β= Input Concentration of Hazelnut Shell
ɤ= Polystyrene
Wt% = Weight Percentage (g)
^{0}C = Degree Celsius
MPa = Mega Pascal

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