Formulation and Characterization of Adhesive Produced From Polystyrene Waste Using Response Surface Optimization

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Abstract. Polystyrene is extensively used in building and construction industry, packaging and transportation of fragile equipment due its low density, high melting point, low thermal conductivity, low water absorption, etc. Polystyrene after usage is usually discarded thereby causing environmental problems. The post-usage of polystyrene has, therefore, been a subject of intense research in recent times. The aim of this work is to produce adhesive from polystyrene wastes. Polystyrene waste (PS) was collected, processed and dissolved in tackifyer and formulated with diphnyle amine and diethylene glycol dibenzoate additives to produce adhesive using 3 levels variables factors and 4 levels testing factors of design expert optimization software. The produced adhesive was further characterized for viscosity, pH, percentages solid and moisture contents for their response surfaces. The results showed that the best fit viscosity for each run was Run 1B> Run 5A > Run 5D> Run 5B>Run 4D based on the regression analysis and analysis of variance (ANOVA). The pH values obtained ranged from 4.0 to 6.3; percentage moisture content was in the order of Run 1B < 5A<4D<4B and percentage solid content was in the order of Run 1B<5A<4D. The best fitted adhesive was run 1B with 5.93 % moisture content; 5A has 7.57 % moisture content and 4D with 8.76% moisture content. The percentage solid content; Run 1B has 67.19 %, 5A has 68.16 % and 4D has 75.50 %. The produced adhesives were found within the standard range of adhesives used in production of particleboard.

Keywords: adhesive; characterization; formulation; polystyrene waste; response.

INTRODUCTION

Waste is any unavoidable material resulting from domestic, industrial or social operations that are not having any economic value and the end results is disposal. The prevailing situation of indiscriminate disposal of non-biodegradable waste materials is a great concern for sustainable ecosystem and clean environment. Nevertheless, these wastes could also contain a lot of valuable resources in the form of nitrogen, phosphorus, potassium, methane and other chemicals which might be useful [1]. The search for better life and socio-economic activities contributes to generation of these wastes from different sources, which are either classified as solid or liquid [2].

Polystyrene which is known as Styrofoam is a synthetic aromatic polymer made from monomer of styrene, which can either be solid or foamed. It is traditionally produced by alkylation of benzene reacting with ethylene to produce ethylbenzyene. Its dehydration results into formation of styrene monomer [3]. The Chemical formula is $[-CH_2 - CH - (C_6H_5) -]n$ and has molecular weight of 104.15 g [4].

Polystyrene decomposes between the temperature ranges of 350 to 450 °C [5, 6]. Take as much as 30 % of landfills worldwide [7] and has low density [3, 8]. Highly flammable, releases lots of black smoke when combusted and generally non-biodegradable [9]. Recycling is not economical [10]. Incineration requires high temperatures up to 1000 °C and plenty air, as much as 14 m^3/kg [11]. When buried remain as non-decayed materials, preventing water infiltration to the ground [12]. However; it dissolves easily in chlorinated solvents and many other aromatic

hydrocarbons [7, 13, 14]. This research is aimed at conversion of polystyrene waste to adhesive using affordable solvents and additives for stabilization. This will also be of more economic value and create clean and sustainable environment.

Adhesive is a substance when applied to substrates sticks to the surfaces that two become bonded together by wetting the surfaces to be joined [15]. It is either natural from animal bone and vegetable sources or synthetic from chemicals [16] depending on the source of formulation. Researchers have shown that adhesives are used in furniture making [17] and composite materials such particleboard for modern furniture for both industrial, domestic and office usage [12, 18]. However, 80% of adhesive used in productions of wood based panels contains urea formaldehyde [19], has been reported carcinogenic and non-friendly to the users [20, 21, 22]. Various formulations have been experimented to reduce its emission to cushion its health effect are still at the infant stage. Considering the health effects of urea formaldehyde resin in production of composite materials, this research is desired to formulate adhesive from polystyrene waste for particleboard production.

C. Xing [23] determined the effect of pH, solid and catalyst on the gel time of urea formaldehyde adhesive. The following factors determines adhesive quality; viscosity, pH, % M. C. & % TS as used in most composite materials and panel production [24, 25, 26, 27, 28].

Adhesive pH is critical in ascertaining its longevity and handling processes [29, 30, 31]. It's determined the applicability of adhesive as curing depends of pH value. The percentage moisture (% M. C.) of adhesive determines the longevity and its adherends to substrates during application, and for non-water soluble adhesive it should be less than 10% [13, 32, 33]. Authors [34] and [19] reported that high moisture contents dilute the adhesive which could weaken its strength in adhesion to substrates. The procedure for determination of adhesive % M. C. is by using Equation 1 [32].

$$\text{Moisture content, \%} = \frac{\text{Original weight} - \text{Dry weight}}{\text{Original weight}} \times 100 \quad (1)$$

The solid content (%TS) is critical in quality parameters of adhesive, for water soluble the solid content is within the range of 55–57 % [19], while non-water soluble are greater than 65% [24, 27]. The procedure for determination of %TS is by using Equation 2 [35].

$$\text{Solid content, \%} = \frac{\text{Dry weight}}{\text{Original weight}} \times 100, \quad (2)$$

In this research, design expert 6.0.8 version software was used in the formulation of adhesive [15]. The resin was formulated using polystyrene waste and tackifyer at different ratios, and followed by 3 factor development of adhesive with additives [36]. The produced adhesive was further characterized with 4 response surfaces.

MATERIALS AND METHODS

Polystyrene waste was collected from commercial outlets in Bauchi metropolis, the tackifyer was obtained from Total Filling station, Yelwa. The reagents used: diphenyl amine (99.9 % purity), diethylene glycol dibenzoate (99.8 % purity) BDH Chemical and absolute ethanol (98 – 99 % purity) Nertherlands GPR were purchased from a local vendor. The equipment used were digital weighing balance model PGW 45021, Hot Air Oven, Rotary viscometer model TT-5, pH meter model JENWAY 3510, mechanical stirrer model Heidolph 50111 and measuring cylinders.

Polystyrene wastes were first washed and dried, then fragmented and weighed. The plasticizer was synthesized from diethylene glycol and benzoic acid, tackifyer and antioxidant were formulated based on experimental design. This was followed by formulation of resin based on 2 factorial design using mixture following the procedure as presented in Equation 3 [15, 24].

$$\begin{matrix} Polystyrene\ waste\ (PS), \text{g} + \\ + Tackifyer\ (Tkf), g = 1 \end{matrix}, \quad (3)$$

Resin design formulation is presented in Table 1.

Table 1 – Resin Design Formulation

Component, g	Low limits	Constraints	High limits	Coded factors	
				Low	High
PS	0.50	A	0.75	0	1.000
Tkf	0.25	B	0.50	0	1.000

After resin formulation, 3 factorial designs variables were developed to produce adhesive using mixture design expert and D-optimal for response surfaces. Equation 4 presents the formulation procedure for adhesive formulation [15, 36].

$$A+B+C=1, \quad (4)$$

Adhesive experimental design is presented in Table 2.

Table 2 – Design Constraints Table Adhesive formulation using D-optimal

Component	Name	Units	Actual values		Coded Values	
			Low	High	Low	High
A	Resin	g	0.65	0.79	0.000	0.993
B	Plasticizer	g	0.20	0.34	0.000	1.000
C	Antioxidant	g	0.009	0.01	0.000	0.007

The produced resin was further blended with PLZ and AOX based on the DOE ratios and stirred with the addition of additives at ambient conditions, resulting into a thin film of adhesive produced. The produced adhesives were characterized for their response surfaces results at ambient conditions.

Rotary viscometer model TT-5 was used according to the standard procedure [37]. The apparatus were set on automatic mode, with the selection of appropriate spindle for viscosity test. It was gribbed, adjusted and inserted into the adhesive up to a mark. The automatic rotary viscometer was powered on to start running as it selects the rotation in revolution per minute (RPM) at 6, 12, 30 and 60. After every rotation it displays the data for the viscosity at each RPM which was recorded as the viscosity at that point. This test was rerun three (3) times for each sample for accuracy.

The pH meter JENWAY model was used to determine the pH of the produced adhesive before and after stirring. The meter was cleansed with solvent to be free of dirt and impurities on the electrodes. It was followed by stabilization in buffer solution and immersion in the sample. The meter start reading immediately when it is immersed until it attains stability and a curve is displayed with ready showing optimal value plotted. The test was rerun three (3) times for each sample for accuracy.

The percentage solid content of produced adhesive was determined using laboratory crucibles. A known quantity of the sample was weighed and oven dried at a temperature of 200oC. After 2 hours, the sample drying was discontinued and removed from the oven to cool and weighed after as dry weight.

RESULTS AND DISCUSSION

The viscosities of produced adhesives were determined and their results are presented in Figures 1–3.

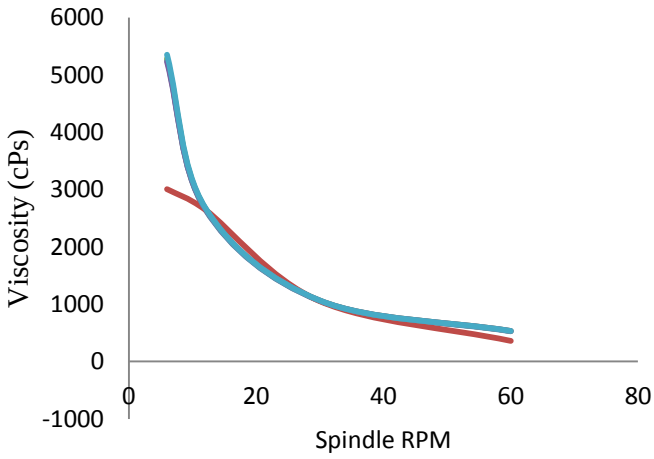

Figure 1– Run 1 Viscosities

Figure 1 present viscosity curves for run 1 of produced adhesives 1 (A, B, C, D & E) in triplicates, some curves were overlapping due to closer values of the viscosities. The values obtained are: Runs 1A, 5267 cPs; 1B, 3006 cPs; 1C, 5285 cPs; 1D, 5228 cPs & 1E, 5351 cPs. These values were found within the range of viscosity data for urea formaldehyde adhesive used in particle board [25, 26, 27]. The data was modeled to get the best fit using regression (R^2) curve and DOE response model [24]. The results showed that (R^2) are: Runs; 1A 0.7034; 1B 0.9105; 1C 0.7042; 1D 0.7077 & 1E 0.7010. Run 1B (R^2) and 0.8612. Thus; run 1B is considered significant model which can be used to navigate the design.

Figure 2 presents viscosity curve for Run 4 (A, B, C, D & E) in triplicates. The viscosity data obtained are: 4A, 5240 cPs; 4B, 5259 cPs; 4C, 5250 cPs; 4D, 4846 cPs & 4 E, 5229 cPs. Furthermore, the R^2 values are: 4A, 0.7035; 4B, 0.7039; 4C, 0.6468; 4D, 0.7265 & 4E, 0.7048 respectively.

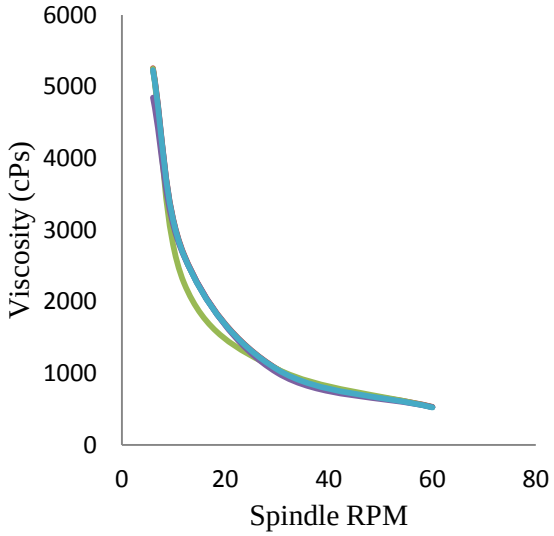

Figure 2 – Run 4 Viscosities

Viscosities obtained in this experiment falls within the range of viscosity of urea formaldehyde used as binder in panel and other composites materials production [25, 26, 27].

Figure 3 presents viscosity curve for Run 5(A, B, C, D & E) in triplicates. The viscosities data obtained are: 5A, 2431 cPs; 5B, 1925 cPs; Runs 5C, 5D & 5E data were not detected by the viscometer which implies lack of adhesive fitness. Furthermore, the R^2 values are; 5A, 0.8201; 5B, 0.7808; 5C, 5D, 0.8148 & 5E, 0.6602 respectively.

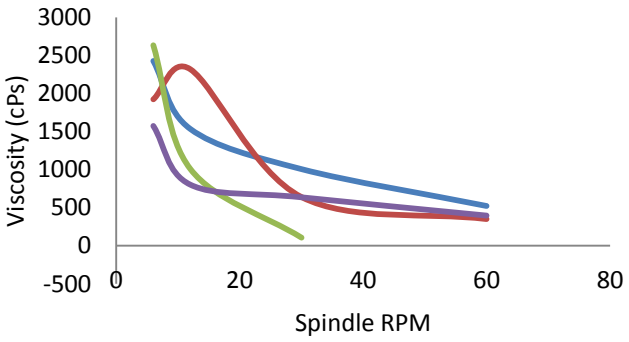

Figure 3 – Run 5 Viscosities

Even though these viscosities values falls within the range of viscosity of urea formaldehyde used as binder in panel and other composites materials production [25, 26, 27, 37], the RS model fails to recommend this model for navigation. Thus; the overall viscosities result analysis revealed that Run 1B model values shows significant model that could be used to navigate the design having R^2 value of 0.9105.

Figures 4–6 presents the pH values obtained from adhesives developed. The response surfaces were determined before and after stirring to study the effect of stirring on acidity content.

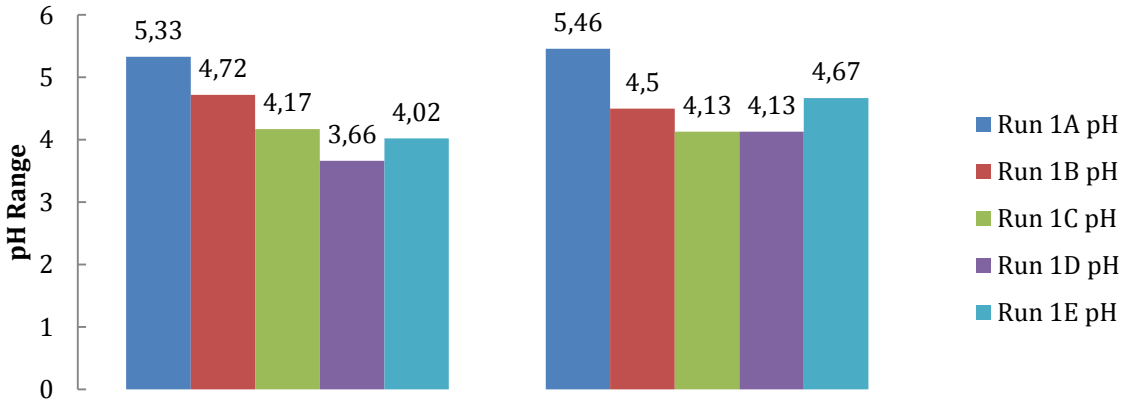

Figure 4 – Run 1 pH before and after Stirring

Figure 4 presents results of pH data obtained from experimental results before and after stirring. The pH before stirring was lower than the pH after stirring; this differential could be due to the effect of homogenization of the sample after stirring with phase disappearing after stirring. It also suggests the presence of plasticizer ratio in the produced adhesive. The values obtained were within the reported values of urea formaldehyde resin used in particleboard production [27]. However; this acidity was as result of dibenzoate used in plasticizer and is a weak acid with less effect on the cure rate of the produced adhesive.

Figure 5 presents the pH values of experimental data results for runs 4 before and after stirring.

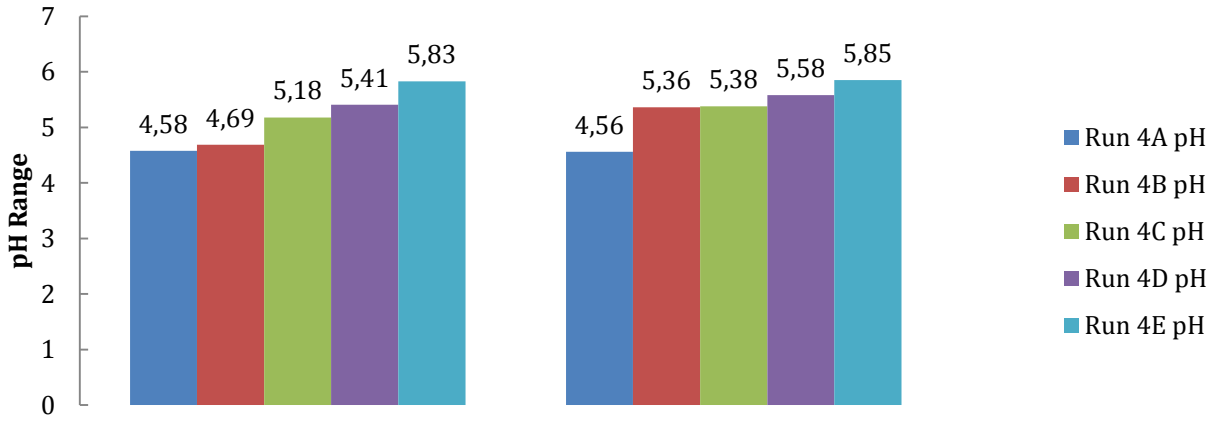

Figure 5 – Run 4 pH before and after Stirring

Except for run 4A, which shows drop in pH after stirring, the remaining had slight improvement in pH values. This difference is due to the settlement of adhesive phase layers. However, when stirred, the phases become homogenized and produced the corresponding value of the sample. Figure 6 presents the experimental results data for runs 5. The samples pH was determine before and after stirring. Run 5A & B values were < 4, while C, D & E were >4 as shown.

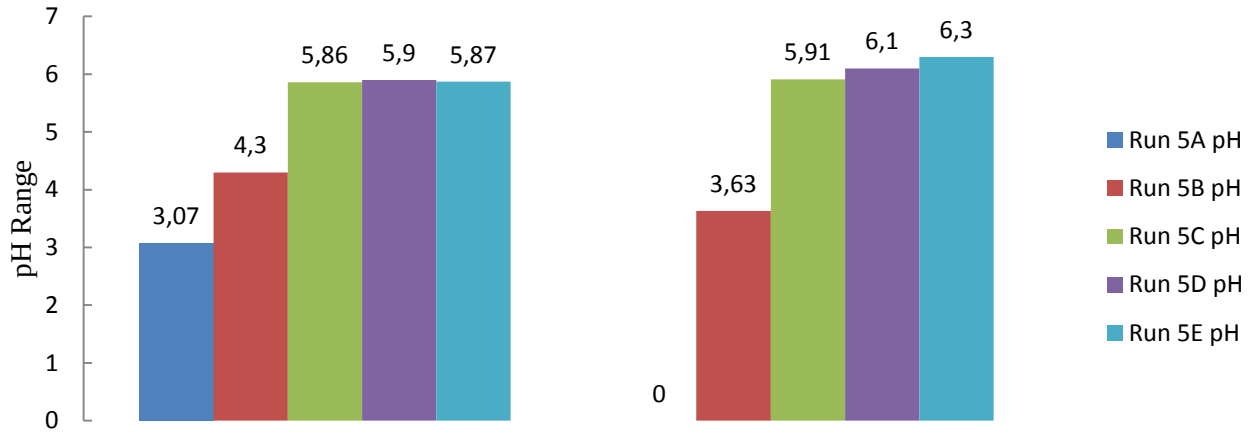

Figure 6 – Run 5 pH before and after Stirring

Run 5A & B even after stirring could not improve the pH values, these high acid values shows that the adhesive produced from this run is not suitable for application in composite materials as high acid values will weakens the strength of the substrate. Runs 5 (C–E) show favorable pH value which falls within the reported values of UF resin used for particleboard production.

The produced adhesives were characterized for % M. C. as presented in Figures 7–9.

Figure 7 presents the % M. C. of produced adhesive for Run 1 (A-E). Run 1 B has the lowest % M. C. of 5.93 % as compared to D (11.01 %), C (15.18 %), A (17.46 %) and E (40.47 %). This low moisture suggests that the model could be used as binders in formulation of composite materials production as high moisture degrade the quality of the adhesive rapidly. Figure 8.0 presents the % M. C. of Run 4 (A, B, C, D % E) of produced adhesives. Out of the 5 samples, Run 4D had 8.76 % being the least % M. C. as compared to B (10.74 %), C (11.54 %), E (13.17%) and A (13.57%).

This revealed that Run 4D could serve as binder in composite material such as particleboard production as it has met the required minimum % M. C. of adhesives used as reported [19, 27, 32, 33, 37].

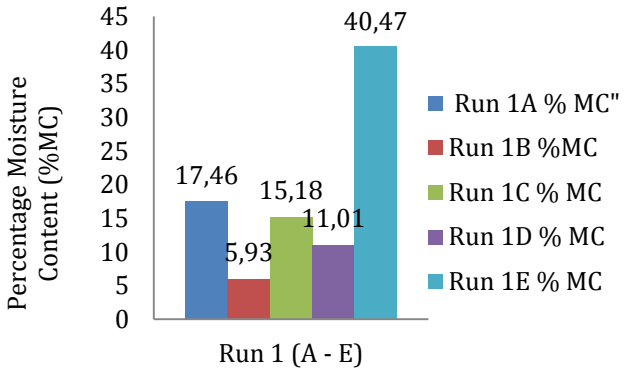

Figure 7 – Run 1 % M. C.

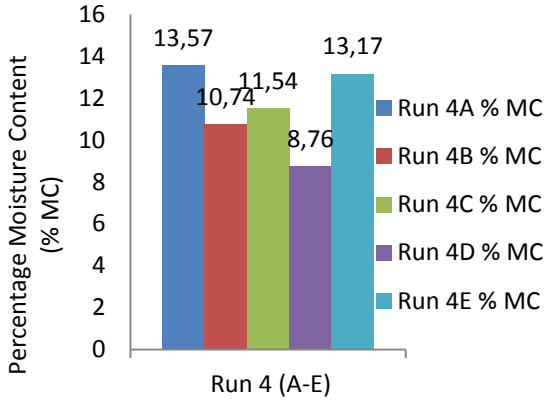

Figure 8 – Run 4 % M. C.

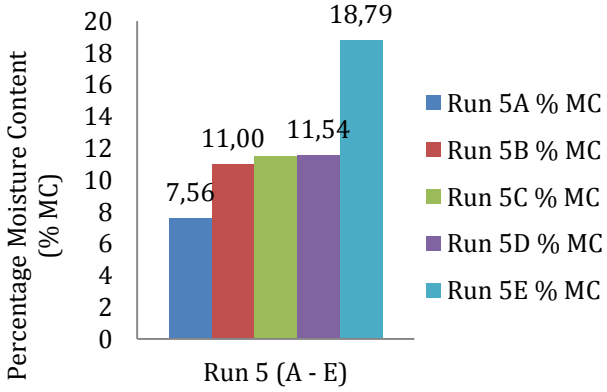

Figure 9 – Run 5 % M. C.

Figure 9 presents the % M. C. of Run 5 (A, B, C, D & E) of produced adhesives. Out of the 5 samples, Run 5A were found to be least with 7.57 % as compared to B (11 %), C (11.45 %), D (11.54 %) and E (17.85 %) respectively. This shows that adhesive with low % M. C. has better property of bonding to substrate and might not degrade rapidly. And of all the samples, Run 1B was the least and best fit based on the urea formaldehyde adhesive used in particleboard as reported is Run 1B < 5A < 4 D.

Figures 10–11 presents the %TS contents of the produced adhesive for different experimental runs.

Figure 10 presents percentage solid contents for run 1 (A, B, C, D & E). The following data were obtained from the experimental runs. A 67.01 %, B 67.19 %, C 58.36 %, D 61.39 % and E 48.41 %, these implies data, Run 1B exhibits the highest %TS of 67.19 % which depicts the solid content of urea formaldehyde used for particleboard reported by [24] which had %TS >65 % for non-water soluble adhesive. While other runs falls below 65 % is not within the expected range of adhesive solid content.

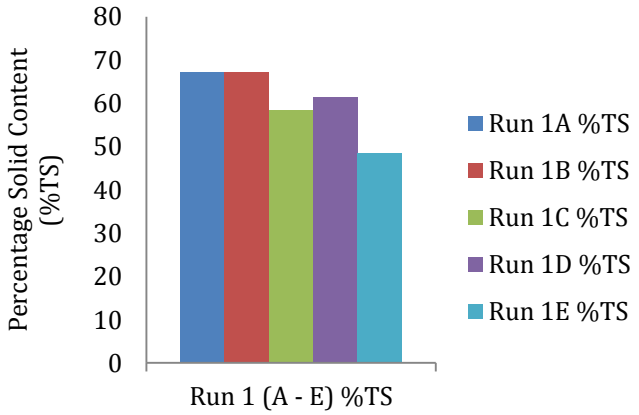

Figure 10 – Run 1 % Solid Content

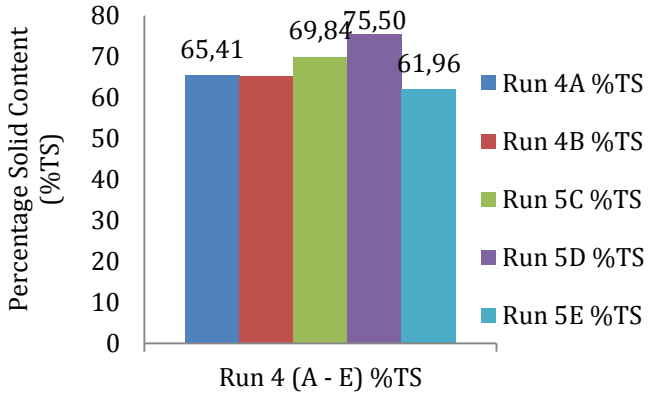

Figure 11 – Run 4 % Solid Content

Figure 11 presents the %TS for run 4 (A, B, C, D & E). The results shows that A 65.41 %, B 65.15 %, C 69.84 %, D 75.50 % and E 61.96 %. from these analysis only run 4E which has 61.96 % TS fall below the TS of adhesive reported used in parti-

cleboard production, whereas runs 4 (A –D) are above 65 %. This implies that adhesives from runs 4 (A-D) could be used for composite material production if other quality parameters are met as reported [24, 27, 28].

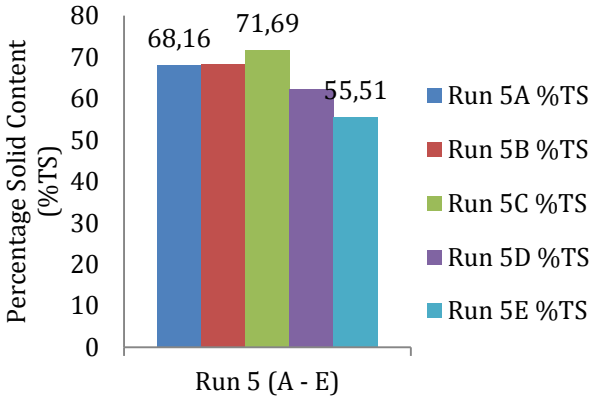

Figure 12 – Run 5 % Solid Content

Figure 12 presents the %TS for run 5 (A, B, C, D & E). The results shows that A 68.16 %, B 68.22 %, C 71.69 %, D 62.15 % and E 55.51 %. From these analysis Runs 5 (D &E) falls below 65 % and Runs 5 (A–C) are within the range >65% for non-water soluble adhesive. Thus, the produced adhesives with %TS above the urea formaldehyde adhesive used in particleboard and other panels could be used in the production of composites materials such as particleboard [24, 27, 28].

CONCLUSION

Adhesive was formulated from polystyrene and tackifyer, using additives as stabilizer. The process revealed that, polystyrene to tackifyer ratios of 0.5625 to 0.4375 was the adequate proportions for resin formulation; while additives with ratios of plasticizer (0.19) and antioxidant (0.01) blended with resins (0.80) produced the best fitted adhesive among others. Run 1B with viscosity of 3006 cPs was best fitted based on the R^2 and ANOVA from the design expert. The corresponding pH of 4.5, percentage total solid content of 67.19 % and percentage moisture content of 5.93 % were obtained. The design expert modeling suggested that; Run 1B could be used to navigate the model as it has met the criterion for adhesive used in panels. Therefore; the produced adhesive could be used for particleboard production.

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