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Research paper

Design of Experiment for In-Situ Synthesis of Waste Vegetable Oils and Synthetic Based Polyurethane (PU) Foam Composites

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Abstract

This study focused on fabrication of polyurethane (PU) foams and its composites based on hydroxylated bioepoxy (B) and petroleum based synthetic epoxy (E), crosslinker and wood fillers. The fabrication of foams: B and E with different wood filler size such as flake (L) and powder (P) on different composition ratios of 5%, 10%, 15% and 20%. The fabrication techniques used in this research are open moulding technique. The physical properties such as morphological pore structure and its homogeneity of wood filler distribution in foam polymer composites were measured. The effect of different size of wood filler loading in PU foam composites were analyzed and discussed in detail such as gelling time, main pore size, interconnected pore and struts thickness. The operatory conditions of in-situ PU foams synthesis were optimized using a design of experiment (DOE). The correlations between factors (X1, X2, X3) and the responses (Y1, Y2, Y3, Y4) for B, E as well as BL, BF, EL or EF foams were further analyzed using Minitab software.

Keywords: *bioepoxy; epoxy; DOE; polyurethane foams.*

1. Introduction

Polymeric foams with high porosity are of great interest as they combine the property characteristics of polymers and porous materials [1]. Polymeric foams have been used in various applications for daily necessities, such as in energy conversation, vibration suppression, sound absorption and heat insulation [2]. Foams, such as polyurethane (PU) foams, polystyrene (PS) foams and polyethylene (PE) foams are characterized by their properties of light weight, high porosity, and good insulation performance which make them ideal materials widely used in building insulation [3].

Polyurethane (PU) foams are the most frequently employed polymeric foams due to light weight, low cost, and good processibility in industrial areas. Polymer foams include a plurality of cells in a polymer matrix. Foam can have an open, partially open, or a closed cell structure [4]. Polymer foam based epoxy is widely used in many applications. It is manufactured by adding blowing agent liquid crosslinker isocyanate to form foams. Epoxy is mixed by reaction of cyclic-carbonate-terminated polymer with triethylenetetramine and without isocyanate. Traditional linkage is used to cure elastomer from starting with polymer with hydroxyurethane linkages to foam the last extension reaction [5].

Polyols are one of the major reactants in PU foam formulations. Recent emphasis on renewable content has reactivated interest in natural oil-based polyols to minimize dependency on petrochemical sources and promote renewable green technology while taking advantage of the low cost and unique properties of vegetable oils [6].

Wood fiber is structural materials in plants and the most abundant biomass in earth. The use of wood fibers as fillers in polymeric composites show advantages as they are inexpensive, derive from renewable source, present lower density than mineral fibers, undergo little damage during process and their disposal causes minor ecological impact [7]. Fillers are defined as materials that are added to a polymer formulation to lower the compound cost or to improve properties. Appropriate loading filler give strength to polymer. The filler is normally organic or inorganic additive their having certain particle size type; flakes, L and powder, P.

Therefore, in this study, PU foam were developed based on waste vegetable oil bioepoxy, B [8-10] and petroleum based synthetic epoxy, E, crosslinker and wood filler. Foams produced from renewable resources are generally rigid because of the small chain length of the polyols, the presence of rigid groups such as aromatic rings also add to their stiffness and high crosslinking density related with high performance. Hence, this material is commonly used in thermal insulation and as structural materials [11]. Nevertheless, in spite of their porous structure, their use as sound absorbers have not received much consideration when compared to flexible foams.

2. Design of Experiment

Two different sizes from the same types of wood filler are prepared and namely as flakes, L and powder, P. The fillers were mixed by the following ratio of 5, 10, 15 and 20 wt/wt % of B and E hydroxylated polyols (monomers). The mixture was left to cure and rise at room temperature for 24 hours and namely as B5L, B5P, B10L, B10P, B15L, B15P, B20L and B20P of B foams and E5L, E5P, E10L, E10P, E15L, E15P, E20L and E20P of E foams. The foams were prepared in a cylindrical shape with a diameter 100 mm and 28 mm as shown in Fig. 1.

A design of experiment (DoE) method was used to optimize the operatory conditions of the fabricated PU foams. Three input factors X1, X2 and X3 were considered as Table 1. The designed matrix of experiments with number of levels 2, 4, 2 were given by

Minitab according to a general full factorial design is gathered and represented in Table 2.

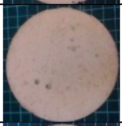
B5L		E5L	
B5P		E5P	
B10L		E10L	
B10P		E10P	
B15L		E15L	
B15P		E15P	
B20L		E20L	
B20P		E20P	

Fig. 1: B5L, B5P, B10L, B10P, B15L, B15P, B20L and B20P are bio-epoxy (B) foams and E5L, E5P, E10L, E10P, E15L, E15P, E20L and E20P are synthetic epoxy (E) foams.

Table 1: Input Factors

Factors	Name	Level
X1	Monomer Type	E-epoxy, B-bioepoxy
X2	Filler Ratio (%)	5,10,15,20
X3	Filler Type	L-Flakes, P-Powder

Table 2: Design Matrix of Experiment

Run	X1	X2	X3
1	1	1	1
2	1	1	2
3	1	2	1
4	1	2	2
5	1	3	1
6	1	3	2
7	1	4	1
8	1	4	2
9	2	1	1
10	2	1	2
11	2	2	1
12	2	2	2
13	2	3	1
14	2	3	2
15	2	4	1
16	2	4	2

Table 3 shows the design matrix of the three variables, X1, X2 and X3 in real values with samples name.

Table 3: Design Matrix of Experiment

X1	X2	X3	Name
B	5	L	B5L
B	5	P	B5P
B	10	L	B10L
B	10	P	B10P
B	15	L	B15L
B	15	P	B15P
B	20	L	B20L
B	20	P	B20P
E	5	L	E5L
E	5	P	E5P
E	10	L	E10L
E	10	P	E10P
E	15	L	E15L
E	15	P	E15P
E	20	L	E20L
E	20	P	E20P

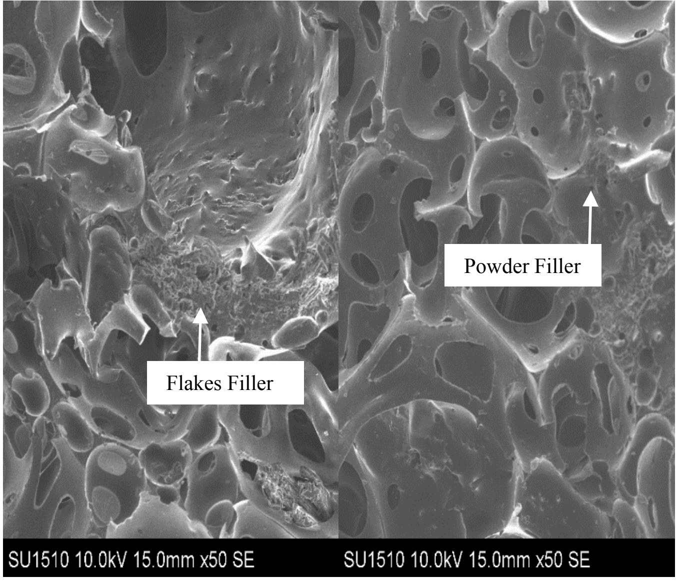
3. Results and Discussion

3.1. Scanning Electron Microscope of Morphological Structure for PUs and its Composites

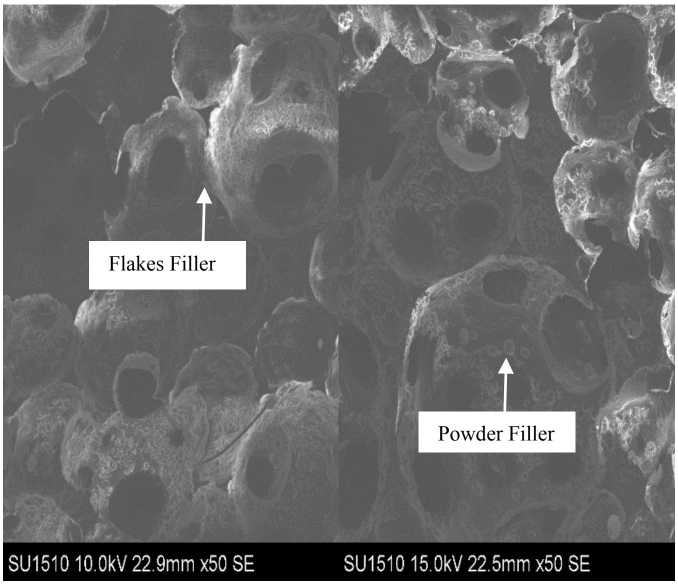
The samples of B and E, foam fabricated from the experiment is as shown in Fig. 1. The gelling time, GT of foam formation was observed. The PU foams with flakes, L filler was found takes longer time compared to PU foam added with powder, P filler. Wood filler loading was limit to 20% wt % ratio of matrix; due to slow reaction between polyol and crosslinker with higher percentage flakes filler loading to form homogeneous porous structure both polyols, both flakes and powder type of filler.

Fig. 2 illustrates the morphology of the filler-matrix distribution and bonding interaction of (a) EP, (b) EL, (c) BP, and (d) BL with interconnected pore cells. The differences of the interconnected pore cell sizes diameter can be related to the weight percentage composition of the filler used in the polymer matrix. As the percentage of the filler increased, the pore size diameter will be decreased.

It is also best to shows that the wood filler was distributed and bonded homogenously for smaller sizes of wood filler as compared to larger filler. This is because of the higher surface area of filler to interact with polymer matrix. Furthermore, the viscosity of the mixture increases as the percentage loading of the fillers increases which contradict to the formation of the pores. Therefore, evidently, the reduction in pore size with further increments of the filler percentage loading. The fillers turn as defects in the matrix cells structures as well as it might attribute to the agglomeration of fillers in the matrix at high percentage loading of filler [12].



(a) (b)



(c) (d)

Fig. 2: Microscopy foams structure of (a) synthetic epoxy with flakes type filler, EL, (b) synthetic epoxy with powder type filler, EP, (c) bioepoxy with flakes type filler, BL and (d) bioepoxy with powder type filler, BP

3.2. Design of Experimental Analysis for Input Factors and Responses for PUs and its Composites

The effect of different size fiber filler with different percentages loading in polymer foam production were analyzed for design of experimental, DoE. The identified factors are to evaluate responses as namely as pore size, interconnected pore and struts thickness. The results of general full factorial design were studied and interpreted using MINITAB. The equation for the design is shown in (1)

$$\begin{aligned} Y = {} & a_1 X1_1 X2_1 X3_1 + a_2 X1_1 X2_2 X3_1 + a_3 X1_1 X2_3 X3_1 + a_4 X1_1 X2_4 X3_1 \\ & + a_5 X1_1 X2_1 X3_2 + a_6 X1_1 X2_2 X3_2 + a_7 X1_1 X2_3 X3_2 + a_8 X1_1 X2_4 X3_2 \\ & + a_9 X1_2 X2_1 X3_1 + a_{10} X1_2 X2_2 X3_1 + a_{11} X1_2 X2_3 X3_1 + a_{12} X1_2 X2_4 X3_1 \\ & + a_{13} X1_2 X2_1 X3_2 + a_{14} X1_2 X2_2 X3_2 + a_{15} X1_2 X2_3 X3_2 + a_{16} X1_2 X2_4 X3_2 \end{aligned} \tag{1}$$

Table 4 shows the samples name and experimental responses; Y: Y1, Y2, Y3 and Y4.

Response Y1: Gelling Time, GT ([h]:m:ss.ss)
Response Y2: Main Pore Size,MP, (mean,µm)
Response Y3: Interconnected Pore Size, IP (mean,µm)
Response Y4: Strut Thickness, ST (mean,µm)

Table 4: Samples Name with Responses

Samples Name	Y1	Y2	Y3	Y4
B5L	0:02:14.029	488.60	154.80	100.00
B5P	0:01:48.014	582.60	219.00	58.00
B10L	0:02:28.018	460.00	153.00	105.60
B10P	0:01:54.023	576.00	208.00	60.00
B15L	0:03:03.110	396.60	143.00	107.00
B15P	0:02:22.380	538.60	169.00	78.20
B20L	0:03:35.020	360.40	140.00	109.00
B20P	0:02:39.050	500.00	166.40	99.09
E5L	0:00:47.027	342.61	119.36	91.98
E5P	0:01:02.029	335.76	136.24	109.21
E10L	0:00:49.015	305.26	231.42	106.75
E10P	0:01:18.014	245.91	140.40	78.75
E15L	0:01:26.000	306.88	109.26	107.34
E15P	0:01:31.049	304.66	160.04	53.86
E20L	0:01:28.049	289.31	87.31	163.82
E20P	0:01:29.050	294.30	134.73	103.54

The correlations between factors (X1, X2 and X3) and the responses (Y1, Y2, Y3 and Y4) for B and E foams and its composites are shown in (2) and (3). R^2 is a statistical calculation that measures the degree of interrelation and dependence between two variables. The factors X1, monomer with R^2 = 46.7% for bioepoxy, B R^2 = 51.4% for synthetic epoxy, E resulted significant different for PU formation response of gelling time (GT) and main pore size (MP) compare to response of interconnected pore (IP and strut thickness (ST).

Referring to Fig. 3(a), the response of mean GT for X1 factor revealed that the B monomer is higher than E as well as X3 factor of filler type shows L is higher than P. This is also revealed that the increasing of filler ratio, X2 was evidently increases the GT. While in Fig. 3(b) with the increasing of filler ratio, X2 shows reduces the size of main pore of PUs foam. The X3 of filler types for mean of pore sizes for L is smaller than P. In other words, the PU foams added with flakes, L filler will have smaller main pore size as compare to powder, P filler.

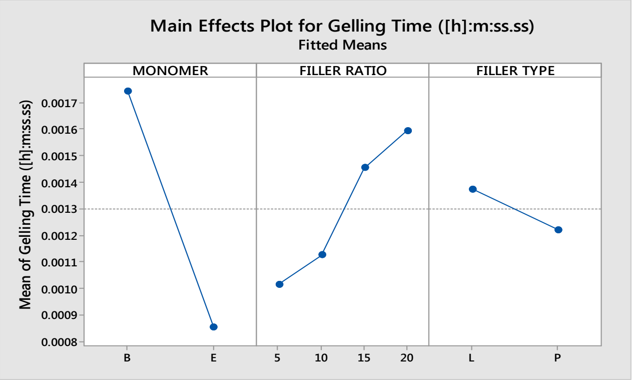
Referring to Fig. 3(c), the Y3 response of interconnected pore size, IP (mean,µm), with X1 factor shows that the B monomer have a bigger size than E monomer. The factor, X2 of filler ratio shows inconsistent effect to the size of interconnected pore, IP. This also revealed that the smaller size of filler, powder, P will increase the size of interconnected pore, IP. While in Fig. 3(d), the response of mean strut thickness, ST ((mean,µm), for X1 factor discovered that the synthetic epoxy, E has higher strut thickness than bioepoxy, E. The X2, filler ratio factor revealed that the increase of filler ratio beyond 15 % appears to increase in strut thickness. The X3 factor for strut thickness added with flakes, L filler is thicker than strut thickness for powder, P filler in polymer foams. In other words, the PU foam add with flakes, L filler will have higher strut thickness as compare to powder, P filler.

Bioepoxy (B) Foam, $X1_1$:

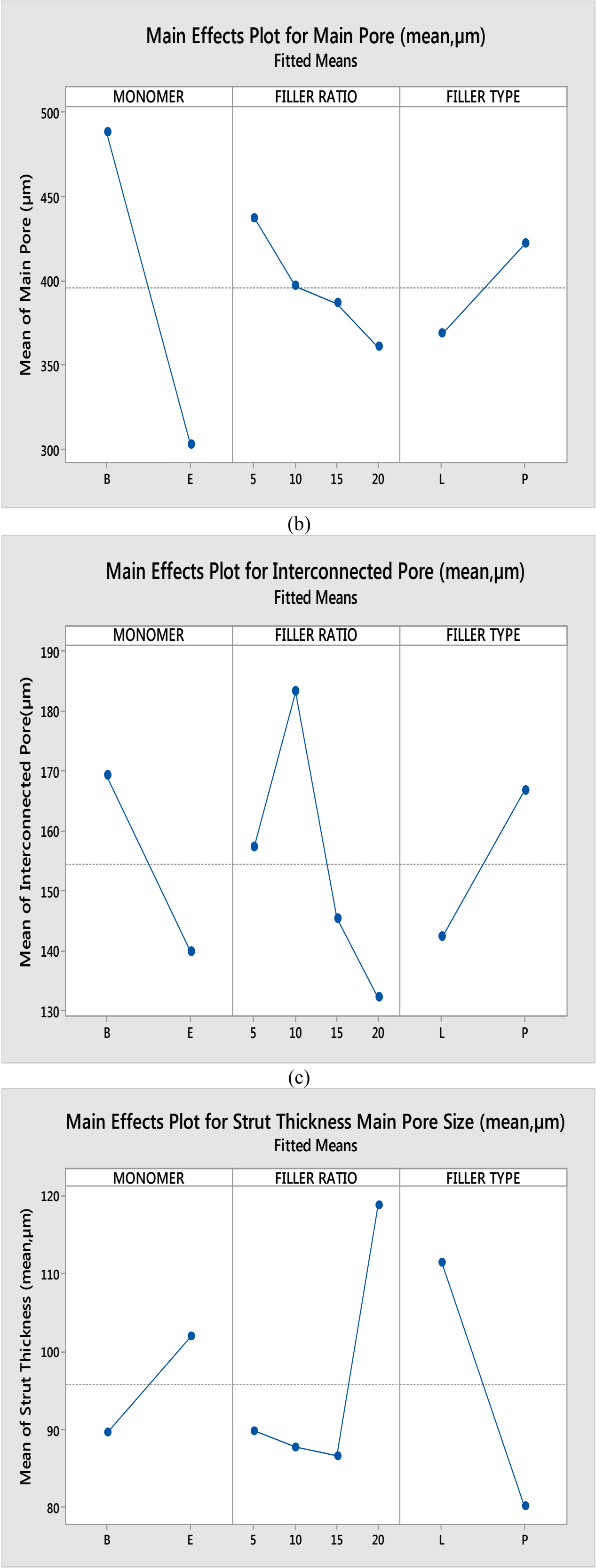
$$\begin{aligned} & Y1 = 0.05857 + 0.03105X2; R^2 = 46.7\% \\ & Y2 = 478.5 + 3.642X3_2; R^2 = 6.3\% \\ & Y2 = 556.3 - 10.16X3_1; R^2 = 45.2\% \\ & Y3 = 172.1 + 0.417X3_2; R^2 = 0.4\% \\ & Y3 = 191.9 - 3.129X3_1; R^2 = 19.2\% \\ & Y4 = 88.63 - 0.4672X3_2; R^2 = 1.0\% \\ & Y4 = 73.56 + 2.217X3_1; R^2 = 20.1\% \end{aligned} \tag{2}$$

Synthetic Epoxy (E) Foam, $X1_2$:

$$\begin{aligned} & Y1 = 0.03302 + 0.01307X2; R^2 = 51.4\% \\ & Y2 = 321.7 - 1.790X3_2; R^2 = 4.6\% \\ & Y2 = 315.9 - 0.848X3_1; R^2 = 0.9\% \\ & Y3 = 137.8 + 0.330X3_2; R^2 = 0.2\% \\ & Y3 = 147.9 - 1.467X3_1; R^2 = 3.3\% \\ & Y4 = 105.2 - 1.347X3_2; R^2 = 2.7\% \\ & Y4 = 78.96 + 3.288X3_1; R^2 = 14.9\% \end{aligned} \tag{3}$$



(a)



(b)

(c)

(d)

Fig. 3: Main effects plot for (a) Response Y1, Gelling Time, GT (b) Response, Y2, Main Pore Size, MP (c) Response, Y3, Interconnected Pore Size, IP, (d) Response, Y4, Strut Thickness, ST

4. Conclusion

Bioepoxy, B from waste vegetable oil represents an alternative renewable resource as PU foam synthesis. PU foam reinforced with synthetic epoxy, E foam with filler show perfect distribution, compared to bioepoxy, B foam with filler. The powder type wood filler was homogenously distributed in foams compared to flakes filler because of the higher surface area to interact with polymer matrix. The size of main pore for PU foam for flakes filler have contributed to smaller sizes compare to powder filler, proved that decrease in pore size with an increase in filler diameter.

Statistically, the main three factors were used such as monomer type, filler ratio and filler type to evaluate the response of gelling time, GT and the size of main pore, MP as well as interconnected pore, IP and struts thickness, ST responses. The correlation between the gives factors; X revealed a significant important of finding which were concluded as:

1. the increasing of filler ratio, X2 was evidently increases the gelling time, GT of the polymer foam reaction process.
2. the bigger the size of filler such as flakes, L will produce smaller main pore size as compare to powder, P filler in polymer foams as well as to increase the strut thickness.
3. the smaller the size of filler such as in powder, P will increase the size of interconnected pore, IP of polymer foams.

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