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ACOUSTIC ABSORPTION, RHEOLOGICAL AND MECHANICAL CHARACTERISTICS OF WASTE EGG BOXES FIBERS FILLED SBR

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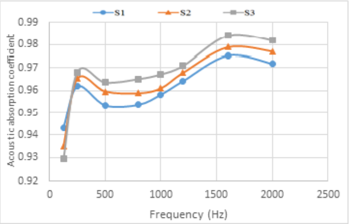
Graphical abstract



Abstract

There are a few interesting to develop a procedure for design material with high acoustic absorption with broad acoustic frequencies range and assess the potential of using waste living materials as the primary component in the production of sound absorbing materials for use in walls and ceilings. This research provides experimental investigations for design and optimization of composite sound absorbers with styrene butadiene rubber (SBR) and waste egg boxes (EB) fibers. The SBR/EB composites have been investigated for their acoustic absorption at different frequencies, their mechanical and rheological properties. Results indicated that an increasing EB composition enhanced the acoustic absorption coefficient. The density of SBR/EB composites were directly related to the presence of the EB fibers. The principal experimental evidences of nonlinear behavior of viscoelastic materials were discussed by investigating the stress-strain curve. In view of the rheological properties, the SBR/EB composites showed shear thinning behavior at various different conditions, that the apparent viscosity reduced with increasing shear rates and it was greater temperature sensitivity. Eventually, the data obtained clearly indicated that the heterogeneity and the viscosity of the materials play very important factors to provide suitable absorbers, these new materials were beneficial for using as a sound absorber and could be used as an alternative replacement for conventional product because for instance, they are cheaper, nonabrasive and may serve to reduce the noise pollution.

Keywords: Acoustic absorption, egg boxes, pseudoplastic behavior, capillary rheometer, stress-strain curve

1.0 INTRODUCTION

Nowadays the environmental pollution resulting from waste living and industrial waste materials has been concerned, the waste management is one of the priority environmental protection areas in the world [1]. In addition, it has been already pointed out that use of waste living materials and natural-organic fibers as filler permit obtaining several environmental values compared to mineral-inorganic counterparts. Actually, the environmental impact may be enhanced mainly by a decrease in the utilization of fossil-based resources [2]. In addition, if biodegradable polymers are used, fully biodegradable systems can be assumed, thus significantly reducing the troubles linked to the everyday production of firm rubber-derived waste. The primary sources of the carbon black are petrochemical, but the uneasy cost of crude oil has led to the search for fillers that are inferred from other sources. Agrarian products such as sugar cane straw, cellulose, husk rice etc. Which are low cost materials and available for use in large number anywhere, over 300 million tons of the same being produced annually [3]. However, the main mechanism in reinforcement is Filler-filler interactions, particularly at high filler contents. These attractions due to Van Der Waals forces and hydrogen bonding (physical interactions), filler volume fraction and morphology of the filler network [4].

Based on the need to quickly and accurately set and control processing conditions, ensure product acceptance and optimize product performance. Accurate rheological measurements have become essential in the characterization of ability to flow for any materials. By utilization rheological characteristics data of raw materials, process conditions may easily be determined and the final product properties and performance can be predicted. Rheology deals with materials in liquid and solid state. In general usage, rubbers are highly viscoelastic and thus their rheological behaviors are strongly shear rates dependent. These attributes underlie many uses of rubber and is emphasized in applications involving high temperature and strain rates, such as the mechanical capacitors, coatings for impact resistance and acoustic insulating and damping [5].

On the other hand, the acoustic noise pollution created by the traffic system, electrical and mechanical apparatus at the residence and modern industry become more serious issue in our life. Urbanization and heavy growth of construction work in every neighborhood further confirming that the new technologies for noise reduction are necessary. Noise created by different machines can be controlled either by suppressing the noise generating factors or using sound insulation and sound absorbing materials, which improve the reduction of the acoustic wave's energy by different mechanism such as blocking, damping and diminution of the sound pressure. Enhancement of acoustic absorber materials, particularly at low frequencies has been introduced for building and transporting application [6]. Traditionally, noise is controlled by using expensive and non-biodegradable sound absorbing materials such as glass wool, polymer foams, fabric filler and polymer fibers, posing an additional harm to the environment. However, the mechanism of acoustic absorption in porous materials depends on the porosity. When the sound propagates in the interconnected pores of a porous material, energy is lost. This loss of energy results from the heterogeneous structure and the viscous boundary layer effects (the surface of interactions between the two phases). Due to a viscous fluid of the air, the sound energy is dissipated via friction with the pore walls and converted the sound energy into heat. Porous materials are widely used to reduce the acoustic noise levels, which are capable to absorb sound waves in wide frequency ranges [7]. A major drawback of conventional porous absorbers is their performance in the low frequency range [8]. The enhancement of sound absorption coefficient in the rubber blended might be due to the viscoelastic properties of the composites and the movement of the molecular chains [9]. This study focus on the enhancement of acoustic absorption coefficient with a broadband frequency response. Incorporation and dispersion of waste egg box fibers (EB) in order to improve sound absorbent combined with evaluating the influence of EB contents on the physical-mechanical and rheological characteristics of this novel material.

2.0 EXPERIMENTAL

2.1 Materials and Chemicals

All materials and chemicals used in this study were industrial-grade and used as supplied. The EB were supplied from a local market in Pattani province, Thailand. The egg boxes were grinded into fine powder using electrical machine and passed through the sieve with 2.00 mm mesh size to get fine powdered (fluff-egg boxes).

2.2 Compounding and Sample Preparation

Table 1 shows the formulation of the mixtures. To begin with, there are two steps to produce samples (final composites) for property evaluations, these being mastication and compounding process. In the mastication step, the gum SBR was masticated on a laboratory, two-roll mill for 2 minutes at ambient temperature. This is important to soften and reduce viscosity and increase plasticity of the fresh SBR before adding further ingredients. This was followed by mixing of the rubber compounds, this mixing occurred on an open two-roll mill which was configured to provide continuous sheets. Next, fresh SBR mixed with a specified content of ingredients (Zinc oxide, stearic acid, 6PPD, CBS and sulfur) and fillers (EB). The entire time of mixing was kept constant for all composites. Finally, the compounds were kept at 25°C for 24 hours prior to further measurements.

Table 1 Mix formulation of the egg box fibers filled SBR composites

Ingredients (phr) / **Mix code**	**S1 (GUM)**	**S2**	**S3**	**S4**
SBR	100	100	100	100
Zinc oxide	4	4	4	4
Stearic acid	2	2	2	2
CBS	1	1	1	1
6PPD	1	1	1	1
EB fiber	0	5	10	15
Sulfur	1.5	1.5	1.5	1.5

2.3 Cure Characteristics

The cure characteristics were performed with Oscillating Disc Rheometer (ODR 2000) measurement. The tests were performed in accordance to ASTM D2084. Optimum curing time (t_{90}) was determined from the Rheometer chart. The final compounded rubber mixture was sent to compression molding machine to complete the curing (rubber vulcanize) at 160˚C for the respective optimum cure time (t_{90}) obtained from Rheograph.

2.4 Mechanical Properties

The cured dumbbell-shaped rubber specimens were used to investigate the mechanical properties such as elongation at break, tensile strength and stress-strain curves. The specimens were cut from vulcanized sheets with a template and a sharp blade. The tests carried out on a Hounsfield tensile testing instrument (model H10KS). Tensile tests were performed at a constant speed of separation 50 mm/minute at ambient temperature (23 ± 2°C). The test pieces are then stretched in a tensile testing machine and the force required to stretch the samples was measured.

2.5 Density Measurement

In the present study, the Densimeter (MD-300S) was used to conduct measurement of the composites average apparent rubber density. The determination of the exact density of samples was obtained from at least three independent measurements. The test piece comprises a number of small and smooth pieces within the size 2×2×0.3 cm^3. The test piece was weighed in air and then in water using a balance reading to 1 mg.

2.6 Sound Absorption Coefficient Measurement

The sound absorption test was carried out using Standing wave apparatus, the standing wave tube (Kundt's) has been broadly performed by measuring the absorption coefficients of absorber materials. The acoustic test system comprises of an impedance tube, microphone, speaker and digital frequency analyzer. A sample was placed at one end of the pipe and a plane acoustic wave propagates parallel to the axis of the tube, for different frequencies in the range from 100 to 2000 Hz, a 100 mm diameter tube was adopted. In order to characterize the compound material acoustically, more details about this technique can be found in our previous work [5].

2.7 Rheological Measurements

The rheological properties of EB-fiber reinforced SBR composites were carried out using a capillary rheometer (model RG20-Gottfert). The capillary rheometer using Die of (l/D = 32/2) ratio 16 and angle of entry 90˚ were used in all measurements, the temperature range from 100 to 130˚C and the shear rate from 0.1 to 1000.0 s^{-1}. A capillary tube of radius (r) and length (l) is connected to the bottom of a reservoir (Die). Pressure drop (ΔP) (The difference of the pressure (ΔP) between inlet and outlet of the tube) and flow rate through the capillary are used to determine the viscosity.

The SBR/EB composite was pressed with a piston-cylinder system through a capillary tube. In this study no end corrections were applied to the stress data. Experimental data were obtained from the capillary rheometer using the following equations:

At the wall the shear stress is [10]:

$$\tau_w = \frac{r\Delta P}{2l} \tag{1}$$

For a non-Newtonian fluid, as a rule, the shear rate is defined as the apparent shear rate at the wall and given by [11]:

$$\dot{\gamma}_a = \frac{4Q}{\pi r^3} \tag{2}$$

Where Q is the volumetric flow rate and $\dot{\gamma}_a$ is the apparent or the non-Newtonian shear rate. The shear rate and shear stress are now recognized. Therefore, the apparent viscosity can be calculated using:

$$\eta_a(T,\dot{\gamma}) = \frac{\tau_w}{\dot{\gamma}_a} \tag{3}$$

Ostwald de Weale had already stated model for non-Newtonian materials so-called power law model, is written as [10]:

$$\eta_a(T,\dot{\gamma}) = k(\dot{\gamma}_a)^n \tag{4}$$

This is a two-parameter model, where k is referred to as consistency index and n the power law index (pseudo-plasticity index).

3.0 RESULTS AND DISCUSSION

3.1 Cure Characteristics

Table 2 shows the cure characteristics parameters of SBR/EB composites at various contents of EB. It can be seen that both optimum cure (t_{90}) and scorch time (t_{s1}) independent of the EB loading. In addition, different tendencies have been observed for the curing characteristics of the filled vulcanized rubber, this could be attributed to various factors, for instance, metal oxide content, particle size, surface area, moisture content and surface reactivity. Admittedly, a faster cure rate is obtained from the fillers with high moisture content, high metal oxide content and low surface area [12]. Thus, it was suggested that the observed changes of scorch time and cure time might possibly result from the acidity and the chemical composition of EB fibers. Given that curing chemical reactions take place in a base medium, the addition of any materials that rise the acidity of compound may cause an initiation time of chemical reactions (before vulcanization initiates) to change. Besides EB itself obtained from a local market, without any chemical treatment so the appearance of contaminants may have an effect on the vulcanization reaction.

Table 2 Cure characteristics and physical-mechanical properties of the SBR/EB composites

Mix. Code Cure Characteristics	S1	S2	S3
M_H (dNm)	23.78	26.75	30.89
M_L (dNm)	4.12	5.01	5.75
t_{s1} (min)	5:45	6:11	4:11
t_{90} (min)	15:57	17:16	12:17
Physical-Mechanical Properties			
Average apparent density ($g.cm^{-3}$)	0.802	0.832	0.843
Tensile strength (MPa)	1.717	1.872	1.906
Elongation at break %	385	380	263.75

It is noticeable that both maximum torque (M_L) and minimum torque (M_H) slightly increase with elevated EB contents. As greater concentrations of EB were used, more crosslinks are subsequently exhibited, raising the crosslink density that is crucial to ensure the success of the EB fibers as a reinforcing filler by enhancing the physical crosslinks. In other words, this can be attributed to the fact that the addition of a reinforcing filler increases the viscosity and maximum torque, so the effect is the same as an increase in the crosslink density[13]. The EB restricted the movement between the molecular chains is caused by additional crosslinks formed either by the filler surface (filler-filler interaction) or in the rubber matrix (filler-rubber interaction) brought about the catalytic effect of the filler on the vulcanization reaction.

3.2 Density

The average apparent density was illustrated in Table 2. It clearly indicates that the density of SBR/EB composites falls back on an existing of EB, which it was increased entirely by rising contents of EB. In addition, it has been known that the density of the rubber composites decreases as the reduction of filler contents, indicating that the density of rubbers are lower than those of most kinds of fillers [14]. Again, this occurs due to improvement in the physical crosslinks; by increasing EB content the filler-filler and filler-rubber interactions dominated resulted in an increase the active chain entanglement, accordingly decreased the free volume, these are the reasons why the density rises by elevating the EB content.

3.3 Mechanical Properties

The influence of introducing EB filler into SBR compounds on the mechanical properties was shown in Table 2. Ultimate tensile strength and ultimate elongation result from pulling the dumbbell specimen to rupture (failure). This is a destructive test, which relates to the intrinsic strength of the rubber compound. It is worthy pointed out that the tensile strength significantly increase while the elongation at break decreases progressively as a function of the EB content. That owing to the stiffening of the polymer chains and hence resistance to stretch when strain is applied. Meanwhile, elongation at break shrink with elevated EB contents was due to the stretch resistance of EB fibers in the coiling of rubber chains.

3.4 Stress-Strain Curve

The stress-strain behavior of the SBR/EB composites at various contents of EB were shown in Figures 1. These figures represent a typical stress-strain curve of the elastomeric materials (rubber-like behavior), SBR/EB curves initially starts with linear tendency, followed by extensive elongation at almost constant stress, and this is called the rubber flow region and is clearly a region of nonlinear viscoelastic. As anticipated, at low EB loading, unequal distribution of strain causes the breakage of bonds between SBR and fine EB fibers. As EB loading increased the stress distributes evenly and EB give semi-reinforcing effect, this magnificent perspective view concurs well with the findings of Mamoor and his coworker [15]. The improvement of the tensile strength may be due to the EB fibers broken into small pieces during the mixing process, resulting in particle aggregation and increased filler-filler interaction. From the physical standpoint, the purpose of introducing a fiber into a rubber matrix is to transfer the load from the weak material to stronger one, this load transfer occurs over the length of EB filaments and protect against damage [16]. Sperling has already demonstrated the mechanical properties of the fiber reinforced composites (FRC), who explained that the role of the fiber is to support all main loads and limit deformations [17]. Thus, fibers increase the strength, stiffness and toughness and decreases corrosion, elongation at break, creep and fatigue, similar explanations being also found in the previous literatures [18, 19].

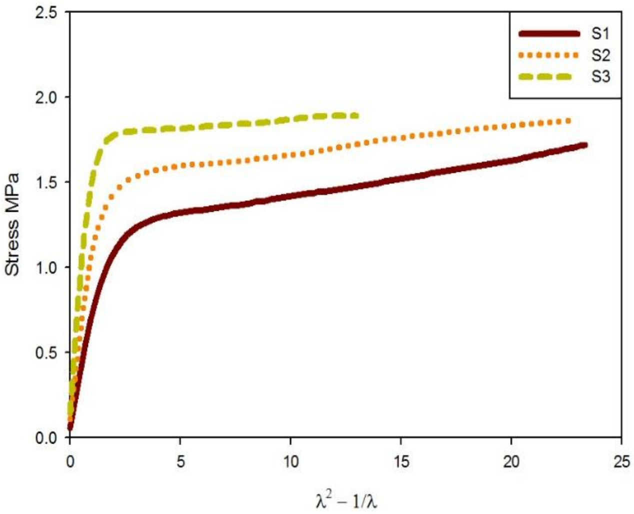


Figure 1 Stress-strain curve of the SBR filled with 5, 10 and 15 phr of EB, respectively

3.5 Sound Absorption Characteristics

The ability of the SBR/EB composites to act as sound-absorbing material was depicted in Figure 2. SBR filled with three different contents of EB 5, 10 and 15 phr (S1, S2, and S3 respectively). It clearly indicates that the acoustic absorption coefficients (α) increased steadily and apparently a peak occurred at 250 Hz (first resonance frequency). By definition, the frequency which provides the highest acoustic absorption peak is denoted as a resonance frequency [20]. It can be seen that a further gain in the sound absorption coefficient was achieved at a frequency of 1600 Hz for all tested samples (second resonance frequency). This phenomenon likely causes by improving the viscoelasticity and viscous boundary layer effect of the materials by increasing EB contents. There is also another possibility that the surfaces of great fine EB fibers might also contain micro-pores.

When the sound propagates in the interconnected pores of a porous material, sound energy is lost. These losses of the sound energy were also attributed to the heterogeneous structure of a composite and the viscous boundary layer effects [21]. The sound energy is dissipated via friction with the interfacial edges (areas of adhesion between the EB fibers and SBR matrix) and then the energy of the sound is transformed into heat energy [22]. It was interesting to observe that S3 (with 15 phr of EB loaded) sample reveals higher acoustic absorption coefficient value than that of others, which is able to absorb 98.4 % (α = 0.984) of the sound at 1600 Hz. In addition, the results obtained in Figure 2 clearly indicates that the acoustic absorption properties of all SBR/EB composites related to the percentage contents of filler, and being improved significantly by increasing the content of EB. Nevertheless, it is also noticeable that the higher density of the composition material the higher the sound energy absorbed and, therefore, the higher the acoustic absorption coefficient. Our results agree well with the previous works [23, 24].

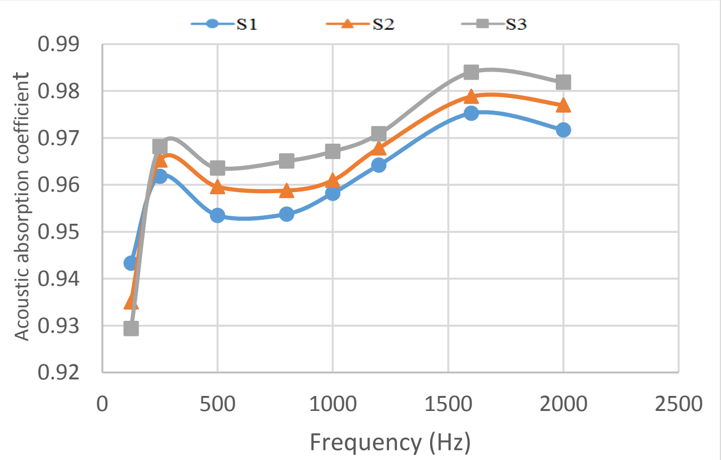


Figure 2 Acoustic absorption coefficient of SBR/EB composites with various EB loadings as a function of frequency

3.6 Rheological Properties of the SBR/EB Composites

3.6.1 *Influence of Temperature*

Substantial temperature changes take place in several stages during rubber processing, so that the influence of temperature on viscosity gains great importance to investigate the process-ability of the new rubber composites. Herein, Figure 3 demonstrates the apparent viscosity of the mixtures versus different constant shear rate at various temperatures. The alteration of the apparent viscosity in wide ranges of temperature at a given shear rate is found to be varied for different mixtures.

It is clear from Figure 3 that the apparent viscosity decreases with an increasing of temperature for the SBR/EB composites. Kumar and co-workers [25] have been already indicated similar behavior for short sisal fiber filled SBR composites. They verified that the reduction of viscosity with an increasing temperature resulting from two reasons. The first one is the acceleration of the molecular movement because of the availability to consist greater volume (free volume increased) and this leads to reduce the crosslink density within the network structures (the points of entanglements diminished). The second reason has been stated earlier due to the weak intermolecular interactions; rubber molecular chains seem to be disintegrated at high temperature ranges. In addition, it is expected that at higher temperatures a greater degree of thermally induced Brownian motion of rubber segments lead to a reduction in apparent viscosity.

It can also be seen that the apparent viscosity reduced with an increasing of the shear rates, which is ascribed to the orientation of rubber molecule chains and the EB fibers in the direction of shear flow. The viscosity reduction is caused by the reduced number of chain entanglements as the chains orient along the lines of flow. At low shear rates, EB fibers are totally disoriented while at higher shear rates are highly oriented and aligned. Moreover, the probability of filler-filler collision is higher for misaligned EB fibers at low shear rates. This collisions increase with EB content and as a result, apparent viscosity increases.

3.6.2 *Activation Energy*

The activation energy of melt flow at a constant shear rate (ΔE) may be calculated using Arrhenius-Frankel-Eyring equation [25]:

$$\eta_a(T,\dot{\gamma}) = Aexp\left({}^{\Delta \mathrm{E}}/_{RT}\right) \qquad (5)$$

$$log\ \eta_a(T,\dot{\gamma}) = logN + {}^{\Delta E}/_{2.303\ RT} \qquad (6)$$

Where: η_a is the apparent viscosity at a constant shear rate as a function of temperature, A is a constant, R is the gas constant and T is temperature in Kelvin. . $Log\eta_a$, plotted versus $1/RT$ and from the slope is ΔE/2.303. The activation energy values make attainable and alternative information on the sensitivity of the materials toward the alterability in temperatures. The higher the activation energy the most temperature sensitive the material will be [26] . The activation energies of the mixtures at various EB loadings are represented in Table 3, which is clearly indicated that the activation energy of SBR/EB composites increase with elevated EB contents. The quantity ΔE is associated with free volume and qualitatively would be expected to decrease [27]. This decrease has been explained by the extra free volume created by the thermal expansion. Conversely, with this consideration of the reduction of the free volume, it is assumed that the EB fibers severely restricts the motion of the chain segments, viscosity may be governed by consecutive jumps of segments of the chain. Thus, it was thought that the observed changes in the apparent viscosity over all SBR/EB composites is greater temperature sensitivity than that of the virgin SBR compound (SBR gum).

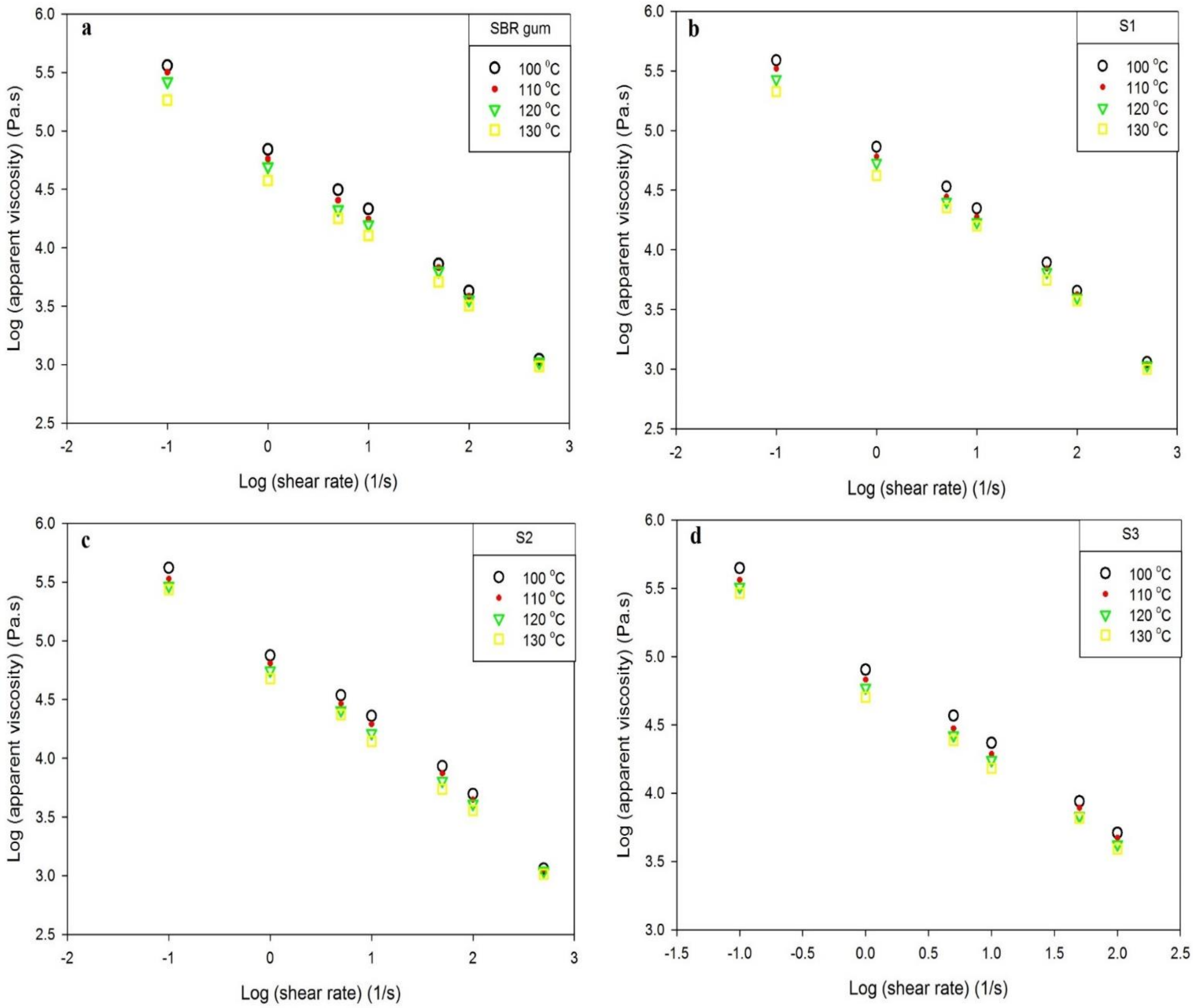


Figure 3 Variation of apparent viscosity with shear rate at different temperature and EB contents with (a) SBR gum, (b) 5 phr (S1) (c) 10 phr (S2) and (d) 15phr (S3)

Table 3 Activation energies (kcal) of SBR/EB composites at three different shear rates

Mix. No.	Activation energy at different shear rates (kcal)		
	0.1 (1/s)	10 (1/s)	100 (1/s)
SBR gum	18.40	17.78	9.72
5 phr EB (S1)	19.35	17.80	10.48
10 phr EB (S1)	20.20	20.15	11.47
15 phr EB (S3)	22.06	20.84	11.58

3.6.3 Power Law Index

The power law index indicates whether a material is Newtonian, shear thickening or shear thinning (pseudo-plastic) in nature. Table 4 clearly implies that the SBR/EB composites exhibit typical shear thinning behavior and a Newtonian plateau is fairly hard to capture. The (n) values for the various SBR composites were less than unity; For example the value of n decreased from 0.326 to 0.303 (at 100°C) and from 0.389 to 0.357 (at 130°C) with elevated EB contents from 0 to 15 phr. The alteration in n values is attributed to the various orientations and distribution of EB fibers in the composites during its flow up to leave the capillary tube.

It is evident from the Table 4 that the pseudo-plasticity index increases with rising the temperature. Higher values of n pointed out to a low non-Newtonian behavior of the composites [28]. Menon has reported that as the behavior departs more from Newtonian (with lower values of n), the shear flow becomes more plug-like with high shear rates being concentrated near the surface of the compounding machine leading to low shearing with the primary consist of the rubber composites [29].

Table 4 Pseudo-plasticity index as a function of EB loading at different temperature

Mix. No.	Pseudo-plasticity index at different temperatures			
	100°C	110°C	120°C	130°C
SBR gum	0.326	0.341	0.360	0.389
5 phr EB (S1)	0.318	0.337	0.352	0.377
10 phr EB (S2)	0.313	0.332	0.345	0.363
15 phr EB (S3)	0.303	0.327	0.336	0.357

4.0 CONCLUSION

A comfortable environment free from unwanted noises is always dreaming of every individual. This discomfort sound may lead to some injuries like hearing loss (temporary or permanent), heart problems, weakness in nerve and pain in internal tissues, and even higher blood pressure in the long term. Therefore, this study has been concentrated on the acoustical and mechanical properties of novel materials produced from the mixture of SBR and EB. Acoustic absorption properties of SBR/RR-EB composites were examined by using a standing wave tube (Kundt's tube) to measure the reflection coefficient of the composites. The absorption coefficient increases with an increasing of the EB contents. Regarding to the mechanical, physical and acoustic absorption configurations, samples with 15 per EB is of interest. It provides excellent composite absorber and suitable to be employed in various sound absorption applications, which were able to effectively absorb approximately 98 % of the sound particularly at frequencies above1200 Hz. On the other hand, cured dumbbell-shaped rubber pieces were utilized to indicate the mechanical properties of SBR/RR-EB composites. Obtained results overall composites showed that the tensile strength and apparent density are enhanced with further increasing in the EB contents while the elongation at break was reduced. The rheological properties of SBR/EB composites filled by various EB loadings have been studied. The viscosity was found to increase with increasing EB contents, the increase in viscosity is more predominant at low shear rates rather than that of higher shear rates. Moreover, like other natural fiber reinforced rubber at higher temperatures the viscosity of the composites is lower than that at lower temperatures. The Pseudo-plasticity index values for all SBR/EB composites were less unity, indicating that all of the composites are pseudo-plastic (shear thinning) in nature. Therefore, the results obtained suggests that the SBR/EB composites may able to consider for many heavy-duty sound damping applications.

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