



## **Effect of [C<sub>2</sub>mim][OTf] Ionic Liquid on Tensile Properties of HDPE/KCF Biocomposites: A Primary Research**

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### **Authors' contributions**

*This primary research was carried out in collaboration between both authors. Author AAS managed the literature searches, designed the research, managed the analyses of the research and wrote the protocol. Author SAAZMD wrote the first draft of the manuscript. Both authors read and approved the final manuscript.*

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### **ABSTRACT**

In this primary research, 1-ethyl-3-methylimidazolium trifluoromethanesulfonate ionic liquid ([C<sub>2</sub>mim][OTf]) was added to the high-density polyethylene/kenaf core fiber (HDPE/KCF) biocomposites by using an internal mixer at 150°C, followed by compression molding at the same temperature. The [C<sub>2</sub>mim][OTf] contents were varied from 0 to 10wt.%. The tensile properties of the prepared biocomposites have been determined by using a universal testing machine. The tensile results of the HDPE/KCF biocomposites indicated that the tensile strength and tensile modulus have decreased with the addition of [C<sub>2</sub>mim][OTf]. However, the presence of [C<sub>2</sub>mim][OTf] has significantly increased the tensile extension at break of the biocomposites. This research concluded that the addition of [C<sub>2</sub>mim][OTf] at the lower content (≈ 10wt.%) could act as a plasticizer for improving the flexibility of HDPE/KCF biocomposites.

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## 1. INTRODUCTION

Salts that experienced a melting temperature below 100°C were referred to as ionic liquids [1]. They could be synthesized with different chemical reactions [2,3]. Ionic liquids usually were used as a solvent for the isolation of chemical compounds [4]. Moreover, they also have been utilized as a medium for the conversion of byproduct to valuable chemical [5]. Ionic liquids could replace volatile organic compounds because they have very low vapor pressure [6]. In addition, ionic liquids were also applied in the preparation of advanced polymer blends and composites [7]. They have the capability to dissolve biopolymers for the fabrication of biopolymer blends [8]. Recently, ionic liquids have been used as a compatibilizer for synthetic polymer/biopolymer blends [9]. They were also utilized as a coupling agent for biopolymer/mineral filler composites [10]. Additionally, ionic liquid has been mixed with a surfactant to improve the compatibilization between the synthetic polymer and biopolymer [11]. Furthermore, they were employed as a plasticizer for improving the physicochemical properties of biopolymer films as well [12].

Biocomposites are composite materials produced by using natural fibers [13]. They have been prepared by means of various types of natural fibers such as cellulose [14], rice husk [15], bamboo [16], oil palm empty fruit bunch [17], kenaf [18], etc. Biocomposites can potentially be used in diverse applications including automotive, construction, aviation, and industrial prototype such as 3D printed products as well [19]. Even though natural fibers could decrease the manufacturing cost of the biocomposite products, however, they have poor compatibility with synthetic polymer matrices, which can potentially deteriorate their mechanical and thermal properties. Hence, surface treatment of the natural fiber has been carried out to increase the compatibility of the biocomposites [20]. On the other hand, the utilization of secondary filler, for example, mineral particles in the biocomposites system also could improve their mechanical and thermal properties [21].

In this primary research, a hydrophilic room temperature ionic liquid has been utilized as an additive in the polyethylene/kenaf biocomposites. Besides, the use of this ionic liquid in biocomposites system is beneficial as it has low

melting temperature, which is easy to handle, and also has high thermal stability, making it durable at a high temperature of biocomposite processing. The main purpose of this research is to determine the effect of 1-ethyl-3-methylimidazolium trifluoromethanesulfonate ionic liquid on the tensile properties of high-density polyethylene/kenaf core fiber biocomposites. The biocomposites with improved properties may potentially be used in the manufacturing of various products.

## 2. MATERIALS AND METHODS

### 2.1 Materials

The polymer matrix used is a high-density polyethylene, HDPE, purchased from Polyethylene (M) Sdn. Bhd., Malaysia. The natural fiber of the biocomposites is kenaf core fiber, KCF (420  $\mu\text{m}$ ), acquired from the National Kenaf and Tobacco Board (NKTB), Malaysia. The ionic liquid used is a 1-ethyl-3-methylimidazolium trifluoromethanesulfonate,  $[\text{C}_2\text{mim}][\text{OTf}]$  ( $\geq 98\%$ ), which is procured from Sigma-Aldrich (M) Sdn. Bhd., Malaysia. All materials were consumed as attained without further alteration.

### 2.2 Preparation of Biocomposites

The biocomposites were prepared through melt mixing by using a Brabender internal mixer equipped with a real-time processing recorder. The mixing process was carried out at a temperature of 150°C, and the rotor speed was fixed at 60 rpm [21]. HDPE (24 g) was added into the Brabender mixing chamber at the beginning for 3 min, followed by KCF (16 g) for 6 min, and  $[\text{C}_2\text{mim}][\text{OTf}]$  for 3 min. Then, the mixing process was continued for another 3 min, where the plateau torque was reached [11]. The duration of the whole process was 15 min. The contents of the  $[\text{C}_2\text{mim}][\text{OTf}]$  were varied from 2 to 10wt.% of the total biocomposites weight. The biocomposite containing only HDPE and KCF has also been prepared for comparison purpose. The mixed biocomposites obtained from the internal mixer were converted into a 1mm sheet via the compression molding technique by using a hydraulic hot press machine [13,22]. The molding procedures involved the process of preheating at 150°C for 7 min, compression at the same temperature for 2 min, and eventually set it for water cooling at 20°C for 5 min [14].

### 2.3 Tensile Test of Biocomposites

The biocomposite sheets of 1mm thickness were cut into a dumbbell shape using a die cutter (type V). The tensile strength, tensile modulus, and tensile extension properties were ascertained, according to the ASTM D638-10 at room temperature by using a universal testing machine (Instron, model 5567) equipped with a 30 kN load cell. The crosshead speed was  $5\text{mm min}^{-1}$  with a 40mm gauge length [18]. The resultant biocomposites were conditioned in an oven at a temperature of  $70^\circ\text{C}$  for at least 24 hours prior to tensile test [20]. 24 samples from each composition were tested to determine the mean values, and the standard deviations were reported to show the error range [23]. The tensile test results were analyzed using the statistical software program (Microsoft® Excel of Microsoft 365). Single-factor analysis of variance (ANOVA) was employed to discover a statistically significant difference of the tensile extension mean values with a 95% confidence level between the different HDPE/KCF biocomposite samples.

### 3. RESULTS AND DISCUSSION

Figs. 1 and 2 indicate the tensile strength and tensile modulus of HDPE/KCF biocomposites with different contents of  $[\text{C}_2\text{mim}][\text{OTf}]$ . As can be seen in the tensile strength result, the sample without  $[\text{C}_2\text{mim}][\text{OTf}]$  has higher tensile strength than the other samples, which has exhibited high stiffness feature [10]. This proved that the presence of  $[\text{C}_2\text{mim}][\text{OTf}]$  had affected the tensile strength of the biocomposites.

Furthermore, a similar trend was also observed for the tensile modulus of the biocomposites added with  $[\text{C}_2\text{mim}][\text{OTf}]$ , which has not significantly increased as well. The decreases in the tensile strength and tensile modulus were expected due to the enhancement in the sliding of the KCF in the HDPE matrix. This is due to the high mobility and high polarity of the ionic liquid that could interact with KCF solely causing the biocomposites to become softer and weaker [11]. In addition, it was found that as the content of  $[\text{C}_2\text{mim}][\text{OTf}]$  increased, the tensile strength and tensile modulus of the biocomposite samples are drastically decreased down to 48% and 52%, respectively. This is absolutely the typical behavior of flexible material [12]. Therefore, from these results, the presence of  $[\text{C}_2\text{mim}][\text{OTf}]$  in the biocomposites system could decrease their stiffness characteristic.

Fig. 3 shows the tensile extension at break of the HDPE/KCF biocomposites with different contents of  $[\text{C}_2\text{mim}][\text{OTf}]$ . From the data, it is apparent that the tensile extension of the biocomposite sample without  $[\text{C}_2\text{mim}][\text{OTf}]$  is lower than that of the biocomposites containing  $[\text{C}_2\text{mim}][\text{OTf}]$ . This is due to the stiff behavior of the HDPE/KCF biocomposite, which is higher compared to the other biocomposite samples. Fortunately, after  $[\text{C}_2\text{mim}][\text{OTf}]$  was added to the biocomposites, their tensile extension increased. This is because of the creation of free volume by the ionic liquid in the biocomposites system, which can permit the slippage of the HDPE chains [12]. Moreover, it is perceived that the tensile extension result has increased directly proportional to the  $[\text{C}_2\text{mim}][\text{OTf}]$  content. From this observation, the

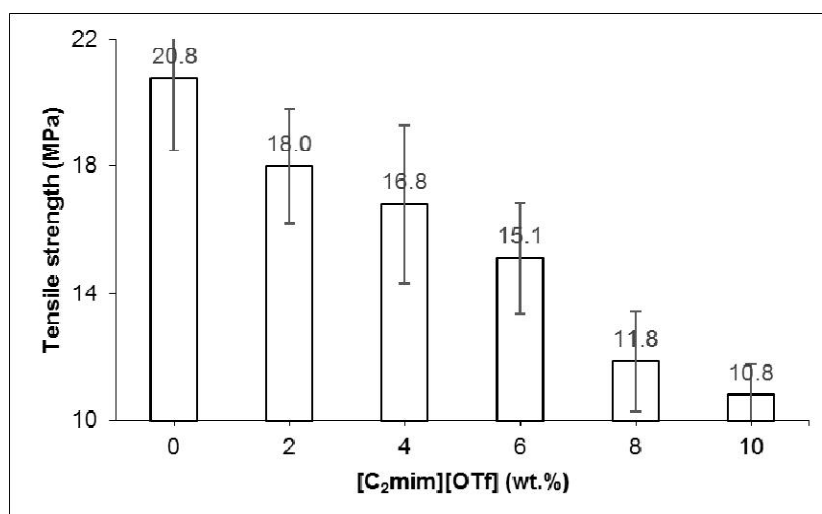


Fig. 1. Effect of  $[\text{C}_2\text{mim}][\text{OTf}]$  content on tensile strength of HDPE/KCF biocomposites

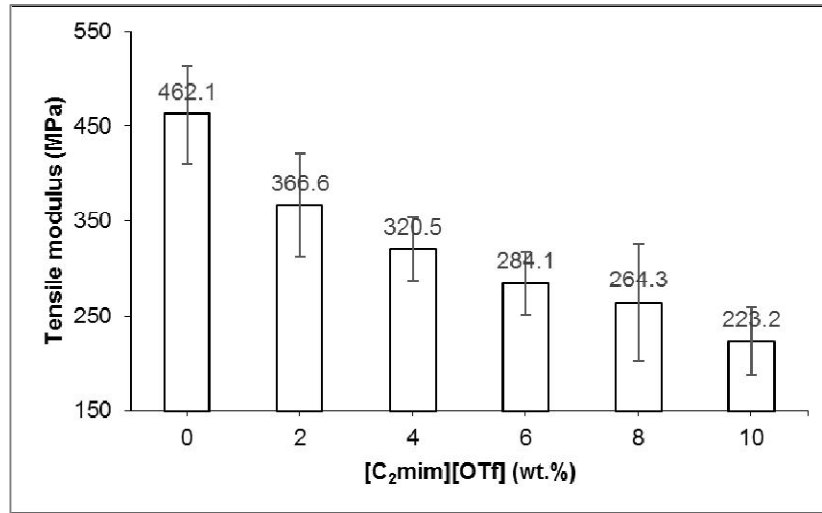


Fig. 2. Effect of [C<sub>2</sub>mim][OTf] content on tensile modulus of HDPE/KCF biocomposites

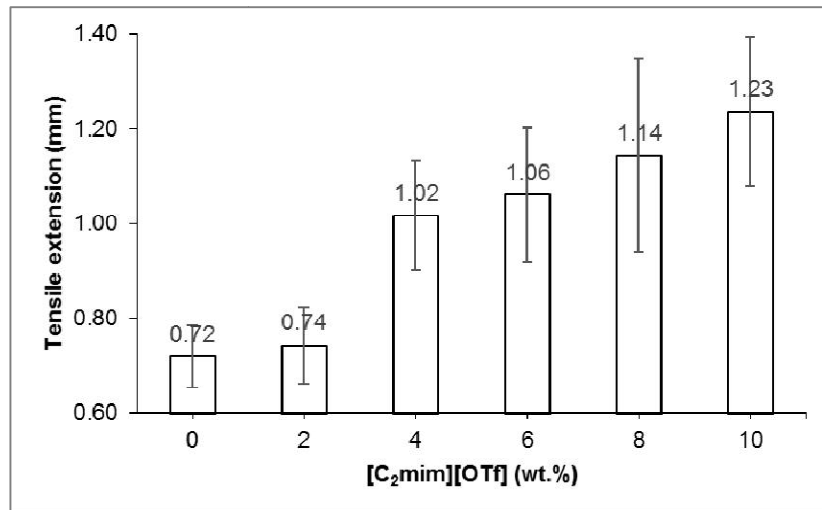


Fig. 3. Effect of [C<sub>2</sub>mim][OTf] content on tensile extension of HDPE/KCF biocomposites

improvement in the tensile extension is due to the ability of the ionic liquid to act as a plasticizer even at the lower content [7]. The significant increase of this result was shown by the sample with 10wt.% of [C<sub>2</sub>mim][OTf], which has demonstrated an increment of up to 71%. Consequently, the addition of [C<sub>2</sub>mim][OTf] has significantly improved the tensile extension of the HDPE/KCF biocomposites, which has provided more flexibility but less stiffness.

Statistical analysis was conducted by using single-factor analysis of variance (ANOVA) to discover a statistically significant difference of the tensile extension between the different HDPE/KCF biocomposite samples. Table 1

indicates the summary result, whereas, Table 2 displays the ANOVA result of the tensile extension for the HDPE/KCF biocomposites without and with containing [C<sub>2</sub>mim][OTf]. The total numbers of the biocomposite samples are six, 24 replicates were tested for each sample. The source of variation of the tensile extension has been divided into two categories, namely, between groups (BG) and within groups (WG). F-value is the ratio of the mean square of BG to the mean square of WG. The P-value is less than 0.05 in Table 2, which rejects the zero hypothesis [9]. Therefore, it can be concluded that there is a statistically significant difference between the tensile extension of the HDPE/KCF biocomposite samples at a 95% confidence level.

**Table 1. Summary result of the tensile extension of the HDPE/KCF biocomposites without and with containing [C<sub>2</sub>mim][OTf]**

Sample (wt.%)	Count	Sum	Mean	Variance
0	24	17.24311	0.718462917	0.004266928
2	24	17.83234	0.743014167	0.00654922
4	24	24.41365	1.017235417	0.013471046
6	24	25.44079	1.060032917	0.019990445
8	24	27.43018	1.142924167	0.0415597
10	24	29.63731	1.234887917	0.025164326

**Table 2. ANOVA result of the tensile extension of the HDPE/KCF biocomposites without and with containing [C<sub>2</sub>mim][OTf]**

Source of variation	SS	df	MS	F	P-value
BG	5.367481620	5	1.073496324	58.025958611	2.9527 × 10 <sup>-32</sup>
WG	2.553038265	138	0.018500277	-	-

SS = sum of square, df = degree of freedom, MS = mean square, F = F-value, Number of samples = 6, Number of observations = 144

#### 4. CONCLUSION

In this primary research, the HDPE/KCF biocomposites containing [C<sub>2</sub>mim][OTf] were prepared through melt mixing process and they were converted into a sheet. The biocomposite sheets were cut into a dumbbell shape for tensile test according to the ASTM D638-10. The tensile strength and tensile modulus of the biocomposites have decreased with the addition of [C<sub>2</sub>mim][OTf] as displayed by the tensile test results. However, the tensile extension of the biocomposites has increased in the presence of [C<sub>2</sub>mim][OTf]. The increase of the [C<sub>2</sub>mim][OTf] content also has enhanced the tensile extension of up to 71%. It can be concluded that the flexibility of the HDPE/KCF biocomposites could be improved with the addition of [C<sub>2</sub>mim][OTf] at the lower content (≈ 10wt.%), which has acted as a plasticizer.

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#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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