FLY ASH/PHENOLIC RESIN COMPOSITE FOR BRAKE PAD APPLICATION: FABRICATION, MATERIALS AND THERMAL PROPERTIES

Nowadays, many parts of automotive components are made of composites. One application of composites is brake lining material in braking systems. Fly ash is waste from burning coal in the power plant industry. Fly ash was added to a polymer matrix to enhance the wear properties of the composite. The appropriate temperature and pressure for composite fabrication were chosen from the composite which has the highest hardness. The addition of 30 wt.% fly ash to the phenolic resin matrix resulted in the lowest specific abrasion of the composite. Additions of graphite, iron powder and nitrile butadiene rubber increased the specific abrasion of the fly ash/phenolic resin composite. Scanning electron microscope micrographs showed the distribution and agglomeration of the particles in the phenolic resin matrix. The addition of fly ash to the phenolic resin matrix also increased the temperature resistance of the composite. Thermogravimetric analysis shows that the starting temperatures for decomposition of the composite constituents shifted to higher temperatures as the fly ash content increases.

Keywords: composite, brake lining, fly ash, phenolic resin, graphite, wear resistance, TGA, hot pressing, thermal properties, iron powder

INTRODUCTION

Most industries, mainly power industries, use coal to boil water into steam to turn turbines. The waste of this combustion process is fly ash. This waste needs places to be deposited. It is also dangerous for the environment, especially when it is in contact with water sources. Fly ash contains silica [1], thus it has a high hardness.

Brake lining material components consist of four main classes. They are the binder, filler, reinforcement and friction modifier [2]. Another researcher classifies brake lining components into fibre reinforcement, binder, friction modifiers, solid lubricant, abrasive, and filler [3].

Fly ash can be used as reinforcement in brake lining material. Other kinds of reinforcement used are metal powders such as iron [4, 5]. The reinforcement in brake lining material increases the mechanical properties and temperature resistance of the material. The mechanical properties are the hardness and wear resistance.

A friction modifier such as an abrasive material increases the friction and wear of brake discs. The abrasive materials are alumina, silica, carbon black, zircon, quartz, silicon carbide and magnesium [1, 6, 7]. Brake lining material that contains silicon carbide has a high coefficient of friction. However, it is not stable, it causes noise and vibration on the brake pad, as well as excessive wear on the steel disk. Low fracture toughness particles such as magnesium particles tend to break into smaller particles, hence resulting in a low coefficient of friction. Zircon and quartz have wear properties between those of silicon carbide and magnesium [8].

Most of the binder material in brake lining material is phenolic resin. Phenolic resin has a high temperature resistance. It has relatively high strength among polymers and is compatible with other component materials [9-11].

Baryte is commonly used as a filler in brake lining material. Low cost filler materials are necessary in order to decrease the amount of the other expensive components in the composition. An addition of baryte up to 20 wt.% in brake lining increases the coefficient of friction and decreases the wear rate of brake lining. A further addition of baryte up to 40% to the composition decreased the coefficient of friction [12]. A filler is also added to brake lining to improve its manufacturability. Fillers are divided into organic fillers such as cashew dust and rubber, and inorganic fillers such as barium sulphate, mica, vermiculite and calcium carbonate. Organic fillers can reduce brake noise due to their viscoelastic properties. Inorganic fillers also reduce noise and have a relatively high melting point [13, 14].

Solid lubricant is very important as one of main components in brake pad material. The increase in temperature during braking decreases the coefficient of
friction and raises the wear rate. Solid lubricants used for commercial friction material are graphite, metal sulphides and SBS. The lubricant forms a film to protect the surface from excessive wear, decreases the temperature and reduces the vibration. More than one kind of lubricant is added as one of the brake pad components to attain effective lubrication during braking. Solid lubricants are affected by temperature, pressure, speed, environmental condition, etc. [15]. Graphite is also used as a friction modifier [5], as well as a heat and sound absorber. Based on a study on graphite shape, long graphite tended to reduce the noise and heat better than flake graphite [16].

A study on zircon particles compared coarse and fine zircon particles as a friction modifier [12]. The coarse zircon particles showed excellent friction stability with less lining wear. However, the coarse particles caused excessive disk wear. On the other hand, the fine zircon particles in the brake lining produced transient friction films, which caused poor friction stability and excessive lining wear. Other researchers investigated the effect of the particle size and found that abrasive particles remove wear debris in the adhesive wear mode when the particle size was smaller than the critical size [9, 16]. Rubber increased the frictional force and improved brake fade and recovery. The viscoelastic property of rubber contributed to the performance of brake lining material [17].

The fabrication of brake lining material includes mixing, compacting and curing. The fabrication parameters are the mixing duration and speed, compacting temperature and pressure, curing duration and temperature. The composite was compressed under the pressure of 15 MPa for 15 min at the temperature of 150°C. Then, the product was post-cured at 180°C for 5 h under atmospheric pressure [18]. Another composite was compressed at 5 MPa for 10 min under the temperature of 250°C. Then, the product was post-cured in an oven at 140°C for 140 min and 180°C for 200 min [2].

Composites applied as brake lining material mainly undergo high shear stress during braking, which result in abrasion of the composite because of frictional force. An abrasion test was performed to investigate the wear resistance of brake lining material. The thermal properties of brake lining materials are also very important. Three different operating temperature ranges for brake lining materials were distinguished. The operating temperatures of 100–150°C, 200–250°C and 300–350°C occurred during braking at low speed, more severe braking and emergency stop, respectively [19]. The pressure during braking was 1.2–10 MPa. Tayeb proposed four different non-commercial brake pad materials and compared those brake pad properties under dry and wet braking environments. Under dry continuous braking, the coefficient of friction slightly increased with pressure and speed. In contrast, under wet sliding conditions the coefficient of friction decreased [16].

From the literature survey mention before, few investigations have been conducted on the effect of the fabrication parameters, i.e. pressure and temperature, on the hardness of the composite. Moreover, few papers concerned investigations on the effect of each addition of material to the composite components. In this paper, the optimal temperature and pressure in the hot pressing of a fly ash based composite were determined by taking into account the impact of those parameters on the hardness. The effect of the material components on the specific abrasion of the composites was investigated. The thermal properties of the composite with lowest specific abrasion were characterised using TGA. The morphology of the composite was observed in SEM micrographs.

EXPERIMENTAL PROCEDURE

The fly ash originally came from coal waste in the Tarahan power plant, Lampung province. The fly ash coming from fluidized bed combustion (FBC) of coal contained 68.15% SiO₂ [1]. The matrix of the composite was phenolic resin powder with an average size of 250 μm. Baryte was used as a filler in the composite. The other materials added to the composite were commercially available graphite, iron powder and NBR.

The preparation of the composite was as follows: first, the ingredients of the composite were weighed and mixed using a commercial mixer. Then, the mixture was put in a steel mould and pressed at the pressure of 60 MPa at the temperature of 250°C for 40 minutes. Finally, the sample was cured in a furnace at 150°C for 4 hours.

The wear resistance test carried out according to ASTM G99. ASTM G99 is a standard method for wear testing with a pin-on-disk apparatus. The load for the wear test was 3.16 kg, the sliding distance was 100 m and the sliding velocity was 1.97 m/s. Five specimens were used for the abrasion tests. The morphology of the composite surfaces after the wear test were observed using SEM. The TGA test was carried out using an Exstar SII TG/DTA 7300. In the TGA test, the heating temperature of 10 mg specimens was in the range of 25–1000°C. The heating rate was 10°C/min.

RESULTS AND DISCUSSION

Composite preparation: pressure and temperature

The hardness test was carried out on samples that were fabricated using different temperatures and pressures. The Rockwell hardness of the composites is shown in Figure 1. In the temperature range of 200–250°C the hardness of the composite increased with temperature as shown in Figure 1a. In the range of compression pressure of 40–80 MPa, the hardness of the composite significantly increases up to 60 MPa as shown in Figure 1b. At the temperature of 80°C, the hardness only increased slightly. Hence, for economic reasons, the pressure of 60 MPa and temperature of
250°C were selected as the temperature and pressure for hot pressing of the composite. At the high temperature and pressure, the particles diffused into the matrix and promoted an even distribution of the particles in the phenolic resin matrix.

Reinforcement materials

The morphology of each individual material was observed using SEM. SEM micrographs of the fly ash graphite and phenolic resin particles are shown in Figure 2. Figure 2a shows the paste form of the phenolic resin. The fly ash particles have an irregular form as shown in Figure 2b. This shape of fly ash particle is typical of fly ash from the fluidized bed combustion of coal [20]. Figure 2c shows large particles of spherical graphite. A substantial difference in the particle size between the fly ash and graphite might decrease the compactibility of those particles.

Another researcher shows that in the wear test, as the load increases the coarse zircon particle reinforced phenolic resin composite has a constant wear rate [12]. Meanwhile, phenolic resin reinforced with fine particles undergoes a change in the wear rate as the load increases. However, coarse particles cause a higher wear rate of the brake disk. This shows that the particle size affects brake pad performance, especially if the difference between the particles sizes in composite is substantial.

The addition of some materials to the composite components was carried out. The first component of the composite was kept very simple to minimise the costs. The components of the fly ash composite are listed in Table 1. It was designated as the Fa composite. It consists of three materials: phenolic resin as the binder, baryte as the filler and fly ash as the reinforcement.

The specific abrasion of the Fa composite as a function of fly ash percentage is shown in Figure 3. The optimal wear resistance was achieved when the specific abrasion has the lowest value. The Fa composite with 30 wt.% fly ash has the lowest specific abrasion of 0.81E-6 mm³/mm as compared with other Fa composites that contained 20 and 40 wt.% fly ash. The Fa composite with 40 percent fly ash has the highest specific abrasion.

Figures 4 and 5 show the failure surfaces of the Fa30 and Fa40 composites, respectively. Failure of the composite surface because of the frictional force on the Fa composite was observed. The area under the failure part is shown in Figures 4c and 5c.
TABLE 1. Composition of Fa composite

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Phenolic resin [%]</th>
<th>BaSo4 [%]</th>
<th>Fly ash [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fa20</td>
<td>70</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>Fa30</td>
<td>60</td>
<td>10</td>
<td>30</td>
</tr>
<tr>
<td>Fa40</td>
<td>50</td>
<td>10</td>
<td>40</td>
</tr>
</tbody>
</table>

Fig. 3. Specific abrasion of fly ash/phenolic composite (Fa) as a function of fly ash percentage

Fig. 4. SEM micrographs of Fa composite with 30 wt.% fly ash: a) composite surface, b) failure of composite, c) distribution of fly ash in phenolic resin matrix

Fig. 5. SEM micrographs of Fa composite with 40 wt.% fly ash: a) composite surface, b) failure of composite, c) agglomeration of fly ash particles

The surface of Fa40 shows agglomeration of the fly ash particles. The agglomeration of fly ash accelerated during mixing. A weak van der Waals force bonded the particles together. The agglomeration in the 40 wt.% fly ash particle composite was higher than that of the 30 wt.% one. It reduced the compatibility between the fly ash and the phenolic resin. The low compatibility caused low strength of the composite because the load could not be completely transferred from the phenolic resin matrix to the particles. Furthermore, the lack of compatibility between the fly ash particles and the phenolic resin matrix behaved as pores instead of reinforcement in the phenolic resin matrix.

In order to ascertain the effect of the solid lubricant on the specific abrasion of the composite, graphite was added to the Fa composite. It was designated as the FaG composite. The composition and the specific abrasion are listed in Table 2. The lowest specific abrasion of the FaG composite was 1.34E-6 mm$^3$/mm. The specific abrasion of FaG was higher than that of the Fa compos-
ite. The 10 wt.% decrease in fly ash content in the FaG composite lowers the reinforcement ability of the fly ash in the composite. Meanwhile, graphite as a solid lubricant may show its contribution in the mechanical properties of the FaG composite during the braking test [16].

<table>
<thead>
<tr>
<th>Composite name</th>
<th>Phenolic resin and BaSO$_4$</th>
<th>Fly ash</th>
<th>Graphite</th>
<th>NBR</th>
<th>Fe</th>
<th>Specific abrasion</th>
</tr>
</thead>
<tbody>
<tr>
<td>FaG</td>
<td>70</td>
<td>20</td>
<td>10</td>
<td>-</td>
<td>-</td>
<td>1.34E-06</td>
</tr>
<tr>
<td>FaGN</td>
<td>70</td>
<td>5</td>
<td>5</td>
<td>15</td>
<td>5</td>
<td>1.97E-06</td>
</tr>
<tr>
<td>FaGFe</td>
<td>70</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td>15</td>
<td>3.13E-06</td>
</tr>
</tbody>
</table>

Failure on the surface of the FaG composite was observed as shown in Figure 6a and b. The large graphite particles were not completely adhered to the phenolic resin matrix as can be seen in Figure 6c. The difference in particle size increases the homogenity between the constituents.

![Fig. 6. SEM micrographs of FaG composite: a) composite surface, b) large size of graphite particles, c) failure of composite](image)

The load, speed and grit size were found to be the influencing parameters other than the material properties. The grit size is the second parameter that affects the wear resistance of the composite [10]. Hence, less force was needed to separate the graphite particle from the phenolic resin matrix. Hence, a high specific abrasion value of the FaG composite was obtained.

The effect of more material additions to the FaG composite was investigated. The additions of NBR and Fe to the composite were designated as the FaGN and FaGFe composite, respectively. The composition and specific abrasion of the FaGN and FaGFe composites are listed in Table 2. The purpose of the NBR addition was to enhance the damping properties of the composite since elastomers such as NBR exhibit viscoelastic behavior [7]. Figure 7a and b presents the SEM micrographs of the FaGN composite. The FaGN composite exhibits matrix cracking and fly ash particles on the crack path. This indicates lower compatibility between NBR and the phenolic resin matrix. The lower compatibility created stress concentration in the composite. This stress concentration causes the cracking within the matrix.

![Fig. 7. SEM micrographs of FaGN composite](image)

In Figure 8, the SEM micrographs of the FaGFe composite show the roughness of the sample surface. The difference in the particle size between the Fe particles and the others, as well as uneven distribution of the particles within the phenolic resin matrix caused the particles to debond easily from the phenolic resin matrix. The compatibility issue requires more attention when more materials are added to the composite. In the case of thermosetting composites, cracks initiate at the filler-matrix interface and propagate very easily through the matrix towards the other filler-matrix interfaces. When the network of cracks intersects, the filler particles are pulled out from the matrix and are removed in the form of wear debris. The resin also becomes removed in the form of fine wear debris. The failure mode of the resin is a brittle fracture. Thus material removal is easier for thermosets than for a thermoplastic matrix composite [10]. The anisotropy in the material microstructure affected all the mechanical properties of the brake pad from the longitudinal to the transverse direction [21].
In this study, the Fa composite shows the lowest specific abrasion among the composites being studied. Hence, if only specific abrasion was used as the wear parameter, the Fa composite would potentially be used as brake pad material over the FaG, FaGN and FaGFe composites.

**Thermal properties of fly ash/phenolic resin composite**

The TGA results of the Fa composite reinforced with 20, 30 and 40 wt.% fly ash are plotted in Figure 9. Weight loss occurred in three steps. Firstly at 100°C weight loss of the Fa composite occurred because of the evaporation of water moisture. Secondly, from 150 to 350°C, the phenolic resin was decomposed. The temperature between 150 and 300°C is the decomposition temperature of phenolic resin. Finally, from 350°C barite decomposition started.

Figure 10 shows the weight loss as a function of the fly ash addition in the composite. As the percentage of fly ash in composite increases, the weight loss curve shifts to the higher temperature. This indicates the increasing temperature resistance of the Fa composite as the addition of fly ash increases from 20 to 40 wt.%. Another researcher added carbon fiber to the brake lining material to enhance the temperature resistance [2]. However, the damping properties of the brake lining material decreased. Degradation of the brake lining was as follows: at 450°C degradation of phenolic resin occurred; at 550°C thermal degradation of aramid fiber; and 650 to 900°C correspond to the degradation of carbon fiber and graphite [2]. The superior high tempera-
ture stability was related to the high temperature resistance of the binder resin and fiber reinforcement of the resin [20].

CONCLUSIONS

Fly ash as a result of coal combustion poses an environmental problem. Fly ash until now has not been extensively managed to create a useful material. The utilisation of fly ash as a reinforcement material in brake pads can help solve the problem. A phenolic resin based composite with fly ash as reinforcement was made for a brake lining application.

The fly ash/phenolic resin composite that was hot pressed under the pressure of 60 MPa and at the temperature of 250°C has the highest hardness. The abrasion test shows that the addition of graphite decreased the wear resistance of the fly ash/phenolic resin composite. It might be caused by the substitution of fly ash with phenolic resin, which decreases the percentage of reinforcement material in the composite. Furthermore, the large difference in particle size between the graphite and other particles might contribute to the decrease in the wear resistance of the composite. The addition of NBR and iron particles to the composite decreases the wear resistance of the composite further. It was shown by the higher specific abrasion of the composite. NBR was added to enhance the damping properties of the composite. Meanwhile, iron particles were added as a reinforcement for the composite.

Based on the specific abrasion test alone, a simple composition of the composite consisting of phenolic resin, baryte and 30 wt.% fly ash has the lowest specific abrasion. The TGA results of that simple composite composition also shows that the temperature resistance of the composite increased with the fly ash addition. Future work will be on the rheological and mechanical testing of those composites since excellent properties of the composite are necessary to apply the composite as brake pad material.

Acknowledgment

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REFERENCES