Electrical Conductivity Of Carbon Pellets Prepared From Mixtures Of Pyropolymers From Oil Palm Bunches and Petroleum Green Coke

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Abstract. Green pellets (GPs), prepared at different compression pressures (cs = 6, 7.5 and 12 metric tonne) from mixtures containing self-adhesive carbon grains (sacg) from the oil palm empty fruit bunch (EFB) and different percentages (pr = 0 to 90 %) of a non self-adhesive powder of petroleum green coke (ppgc), were carbonized (800°C) and activated with CO_2 to produce carbon pellets (CPs). The measured electrical conductivity (σ) of the CP for all cs showed a curve having a minimum value at pr around 50 %, indicating that the conducting phase displays a nonlinear σ–pr relationship. A significant increase in the σ due to CO_2 activation was observed. For a sufficienctly high cs, an existence of a pr range in which the σ varies linearly with the density was also observed. These results provide some new information for modifying the electrical conductivity of carbon derived from the sacg from EFB or other types of biomass.

Keywords: Carbon pellets, Oil Palm Empty Fruit Bunches, Petroleum Green Coke, Density, Electrical Conductivity.
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INTRODUCTION

Fibers of oil palm empty fruit bunches (EFB) can be processed into self-adhesive carbon grains (sacg) or self-adhesive pyropolymers, which can then be pelletized without adding binder and carbonized into carbon pellets (CPs) [1]. This process is obviously simpler than the application of polymer binder and carbon black powder for producing CPs [2]. The sacg can be physically and chemically modified and mixed with other polymeric materials to produce GPs and CPs. Studies of the methods of production, characteristics and application of these pellets can be found in our publications [3-7].

The electrical conductivity (σ) of CP prepared from mixtures of sacg plus pre-carbonized cotton ball (pccb) and sacg plus heat treated kraft lignin (htkl) has been studied [4]. It was shown that the electrical conductivity of the CP increased linearly with increasing htkl and pccb in the sample; the former sample exhibited a sharper increase than that of the latter, because the htkl and pccb are sources of soft and hard carbon, respectively. In our present study, these htkl and pccb are replaced with powder from petroleum green coke (ppgc). In contrary to the htkl and pccb, ppgc has no adhesive property and has significantly higher carbon content. The objective of the present study is to measure the electrical conductivity of such CPs and then analyze the trend of the electrical conductivity data with respect to the

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varying ratios of sacg and ppgc in the samples, as well as with respect to the change of compression pressures (cs) during pelletization. Recent studies of the electrical conductivity of different types of carbon include, for example carbon-graphite-based monoliths [8], porous carbon composite from rice husk [9], carbonaceous powders [10], carbon nanotube composites [11] and carbon black [12,13].

EXPERIMENTAL

The sacg were prepared from EFB using the method reported previously [1]. The raw petroleum green coke obtained from Pertamina (UP II Dumai Refinery), Indonesia, was ball milled and sieved to produce powder (ppgc) that can pass through a 53 µm sieve. Mixtures of the sacg and ppgc, with the percentage composition shown in Table 1, were prepared. For all mixtures, the ball milling for mixing was carried out for 2 h and again the powder was allowed to pass through a 53 µm sieve.

The GPs for each mixing ratio were prepared by applying 6, 7.5 and 12 metric tonnes cs on ~0.75 g of powder in a 2.0 cm diameter mold. GP were carbonized at up to 800°C in a furnace filled with a 1.5 l/min flow of nitrogen gas, using a heating profile which was began with a heating rate of 1°C per min from room temperature to 340°C and holding at this temperature for 1 h, before continuing heating at 3°C per min to 800°C [1]. The activation of the CP produced was carried out in CO₂ (F l/min flow rate) at 800 °C, using a 1 hour hold time and 3°C per min rate of heating.

The average dimensions and densities of the pellets before and after carbonization, and after activation with CO₂ were measured from ten duplicate pellets for each set of samples. The electrical conductivity of the samples was measured using a four-point-probe technique, following the procedure employed previously [4].

RESULTS AND DISCUSSION

Basic Carbonization Data

The data for the weight (m), thickness (t) and diameter (d) of the pellets before and after carbonization, and after activation with CO_2 are listed in Table 1. Analysis of these data shows that after carbonization the samples underwent 13 % to of 57% and 9 % to 53 % weight loss and dimensional shrinkage, respectively. These are due to the release the non-carbon content and rearrangement of carbon atoms during carbonization. A small difference between the percentages of weight loss and dimensional shrinkage has been observed elsewhere [3-7] and it is a major contributing factor toward a small difference of the sample densities before and after carbonization.

Variation of Density

Figure 1 clearly shows that the density of GP compressed with 12 metric tonne is larger than that of the samples at 6 and 7.5 metric tone, because higher cs would obviously decrease the thickness of the pellets and hence produce a more compacted GP. Therefore, the density of GP will progressively increase with increasing cs. The increase of the GP density as pr varies from 0 to 50 % is possibly associated with the fact that softer particles made up of sacg require the addition of harder particles like ppgc to improve the compaction of the mixture, resulting in a higher density. This requirement is optimally achieved when the mixture of sacg and ppgc has a ratio of around 1:1, as shown by the presence of a peak representing a maximum of the density of the GP, particularly for the sample at highest cs (12 metric tonne). Beyond this point, as the proportion of the sacg progressively becomes less than 50 %, the mixture begins to experience insufficient quantities of softer particles that can act

TABLE 1. Basic Data for Pellets Before and After Carbonization and CO₂ Activation. m (weight), t (thickness), d (diameter)

Samples ppgc:sacg (%:%)	Before Carbonization			After Carbonization* and CO₂ activation*		
	m (g)	t (mm)	d (mm)	m (g)	t (mm)	d (mm)
A0(0:100)	0.735	2.001	20.132	0.336ˣ/0.301ʸ	1.699ˣ/1.528ʸ	15.257ˣ/14.992ʸ
A10(10:90)	0.730	1.984	20.134	0.361ˣ/0.336ʸ	1.602ˣ/1.543ʸ	15.915ˣ/15.633ʸ
A30(30:70)	0.736	1.990	20.138	0.437ˣ/0.401ʸ	1.757ˣ/1.696ʸ	17.351ˣ/16.961ʸ
A50(50:50)	0.735	1.991	20.139	0.503ˣ/0.485ʸ	1.882ˣ/1.852ʸ	18.284ˣ/18.022ʸ
A70(70:30)	0.732	1.986	20.154	0.571ˣ/0.562ʸ	1.980ˣ/1.919ʸ	18.791ˣ/18.514ʸ
A90(90:10)	0.712	1.936	20.153	0.614ˣ/0.607ʸ	1.930ˣ/1.911ʸ	19.070ˣ/18.819ʸ
B00(0:100)	0.734	1.978	20.098	0.336ˣ/0.320ʸ	1.603ˣ/1.476ʸ	15.264ˣ/15.017ʸ
B10(10:90)	0.682	1.841	20.113	0.334ˣ/0.299ʸ	1.497ˣ/1.383ʸ	15.852ˣ/15.521ʸ

B30(30:70)	0.686	1.848	20.126	$0.408^x/0.383^y$	$1.647^x/1.584^y$	$17.304^x/17.064^y$
B50(50:50)	0.681	1.819	20.136	$0.470^x/0.454^y$	$1.744^x/1.719^y$	$18.272^x/18.037^y$
B70(70:30)	0.690	1.849	20.163	$0.542^x/0.536^y$	$1.826^x/1.804^y$	$18.801^x/18.702^y$
B90(90:10)	0.682	1.827	20.181	$0.596^x/0.587^y$	$1.821^x/1.810^y$	$19.087^x/18.834^y$
C00(0:100)	0.733	1.945	20.098	$0.313^x/0.297^y$	$1.504^x/1.474^y$	$15.275^x/14.989^y$
C10(10:90)	0.725	1.922	20.102	$0.349^x/0.328^y$	$1.601^x/1.568^y$	$16.096^x/15.762^y$
C30(30:70)	0.725	1.907	20.098	$0.410^x/0.390^y$	$1.717^x/1.686^y$	$17.227^x/16.739^y$
C50(50:50)	0.726	1.889	20.099	$0.500^x/0.484^y$	$1.858^x/1.830^y$	$18.304^x/18.014^y$
C70(70:30)	0.720	1.928	20.098	$0.564^x/0.553^y$	$1.908^x/1.875^y$	$18.836^x/18.564^y$
C90(90:10)	0.672	1.791	20.128	$0.569^x/0.557^y$	$1.786^x/1.754^y$	$18.974^x/18.728^y$

as a binding agent, as well as particles that can fill the small voids when they are subjected to high cs during pelletization. Consequently, the density of the GP decreases as the proportion of ppgc increases to become the dominant component in the mixture. An explanation for the progressive change of the density of samples has also been made in terms of the interparticle voids in the samples, for example for powdered activated carbon subjected to varying cs [14].

The variation of the density of CP against pr is also shown in Figure 1. The dramatic change in the variation of the density of samples is due to the carbonization and CO_2 activation processes. The densities of the CP were smaller than that of GP but still higher than 1 g/cm³. In contrast to the variation of the density of GP, the density of CP for the samples compressed at 12 metric tonne decreased sharply until it reached a minimum at a pr value of around 50 % and then increased sharply for the pr
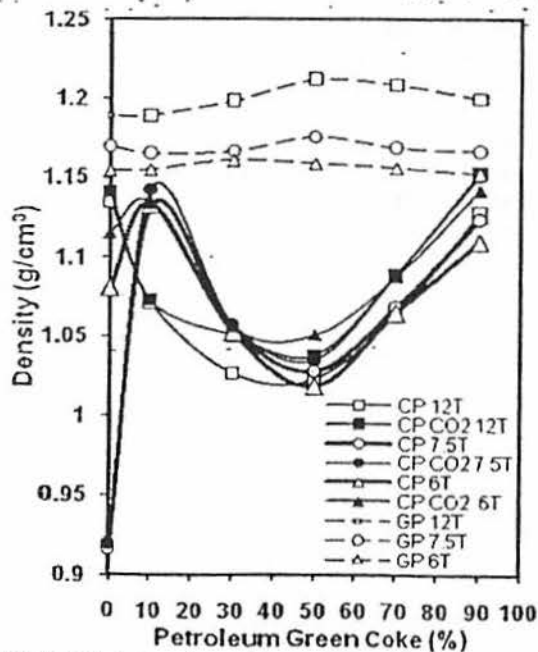
>50 %. Similar behavior is shown by the samples after CO_2 activation. This trend could be due to the carbonization that involves sacg and ppgc particles with different shrinkage rates, which obviously give an adverse effect on the densification of the samples in terms of creating more pore and less dense solid phase in the CP. In the context of volume change due to material shrinkage during carbonization it is logical that the GP having around 1:1 ratio of sacg and ppgc will tend to give the maximum adverse effect and therefore would produce a minimum value for the CP density. A similar trend is also exhibited by the samples at 6 and 7.5 metric tonne, but it excludes the data points at $pr = 0.0$, which indicates that these pressures are insufficient to optimally compress the GP.

Variation of Electrical Conductivity

The overall decrease of σ against pr for the samples after carbonization, as shown in Figure 2, is gradual and almost linear, but one can clearly see the presence of a minimum point for the σ value at the pr value around 50 %. It is also noticeable that in the region of pr values less than 50 % (i.e. higher sacg percentage), the data show a sharper decrease in σ compared to the region where pr is higher than 50 %.

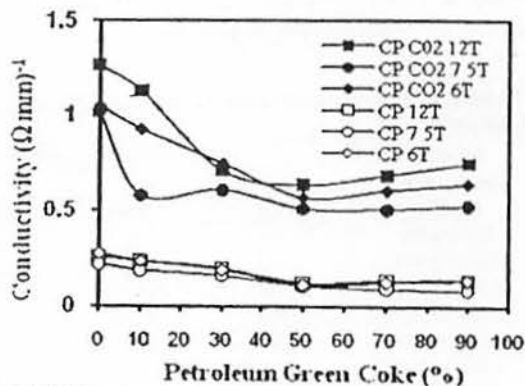FIGURE 1. Density of GP and CP versus pr in the GP.

FIGURE 2. Conductivity of CP versus pr the GP.

This behavior can result from the nature of the sacg particles which can 'experience' a larger carbonization effect, namely larger weight loss and dimensional shrinkage, compared to that of the ppgc particles. A larger carbonization effect can be observed for CP after CO_2 activation, where the σ data exhibit a sharper decrease for $pr<50$ % and sharp increase for $pr>50$ %, and therefore result in a deeper minimum for the σ value. This large effect results from the longer hold time (1 hour) for activation. Other features of the σ-pr data for the samples after carbonization and after CO_2 activation are (i) the σ values for the former samples are smaller than those for the latter and (ii) the change of σ values with respect to cs is smaller for the former samples. This could be due to insufficient hold time for carbonization at 800°C, which can prevent the complete formation of the conducting solid phase that is supposed to be occurring at this temperature, as it was reported for carbon samples from PAN porous–hollow fibers [15].

A linear correlation of the σ-ρ plot in Figure 3 for the CP from GP with pr from 90 % to 50 %, is consistent with the behavior normally found in the σ-ρ relationship for carbon samples within certain ranges of the density [9]. Non-linear and linear behavior of the σ-ρ relationship was observed for the samples of anisotropic expanded graphite-based monoliths [8], and usually it can be explained by the percolation theory and granular model which assumes that the carbon samples are made up of conducting and insulating phases [16].

Figure 3 shows the σ does not systematically increase with the values of cs from 6 to 7.5 metric tonne. However the data at 12 metric tonne has the highest σ, suggesting that the lower cs are insufficient to ensure reproducible results because the inter-particle distances in the GP were not small enough to facilitate the formation of more of the conducting phase in CP during carbonization.
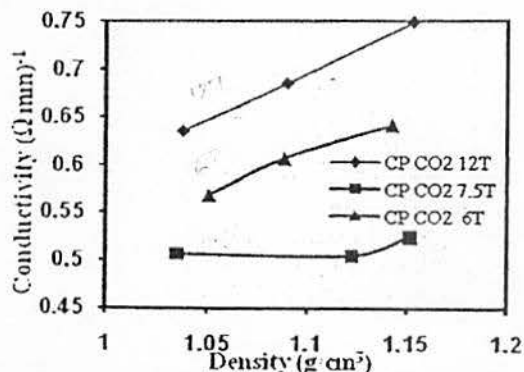

FIGURE 3. Conductivity of CP as a Function of Density.

CONCLUSIONS

The electrical conductivity (σ) of CP and CO_2-activated CP prepared from the carbonization (800°C) of GP containing powdered mixtures of sacg from EFB and ppgc, with pr from 0 to 90 % and with cs at 6, 7.5 and 12 metric tonnes, has been investigated. It can be concluded that the addition of ppgc to sacg to form GP was a significant factor that causes the CP to have values of σ, which vary nonlinearly with pr. The σ–pr curve shows a minimum value at pr~50 % and the effect of the addition of ppgc is more pronounced for higher cs. The results also show a significant increase in the σ after CO_2 activation and furthermore for a sufficiently high cs, the results show an existence of a pr range which leads to a linear variation of the σ againsts the density.

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