Physical characteristics of baked carbon mixes employing coal tar and petroleum pitches

R. K. AGGARWAL, G. BHATIA

Division of Materials, Carbon Technology Unit, National Physical Laboratory, New Delhi 110012, India

A study of the physical characteristics of baked carbon mixes employing coal tar and petroleum pitches as the binder has been made to explore their relative suitability in the fabrication of carbon products. The study reveals that coal tar pitch is the most suitable binder and a petroleum pitch of the same softening point leads to a carbon product of much inferior characteristics. Raising the softening point of the petroleum pitch from 78 to 150° C or adding 10 parts of carbon black in the lower softening point pitch, increases the density, strength and also the electrical resistivity of the baked carbons. The addition of 10 parts of carbon black to the higher softening point petroleum pitch leads to a carbon product with further improved values of density and strength, which are comparable to those obtained with the coal tar pitch. However, the electrical resistivity also is marginally increased. It appears that a petroleum pitch of high aromaticity may fulfill the requirements expected of a good binder.

1. Introduction

Carbon products are usually fabricated from a mixture of carbonaceous filler and binder, such as coke and pitch respectively. The mixture, after being shaped by moulding or extrusion into an artifact of the desired shape and size, is fired in a non-oxidizing atmosphere to obtain a rigid carbon product. The properties of this baked product depend on the choice of the filler, binder and the processing conditions. In fact, any viscous or adhesive material which pyrolyses to give a carbon residue can serve as a binder with varying degrees of suitability. However, coal tar pitch is most commonly used because of its high thermal plasticity, high specific gravity, high coking value etc. [1-3] compared to that of the binders from other sources. A search of the literature reveals that little [4] has been done towards the use of petroleum pitch as binder in the fabrication of carbon products. Charette and Bischofberger [4] considered the compressive strength of the baked carbon as a criterion of the pitch quality. 1632

Recently, a study was made on the rheological characteristics of coal tar and petroleum pitches with and without carbon additives [5]. The viscosity of the coal tar pitch was found to be higher than that of a petroleum pitch of the same softening point. However, the addition of 10% of carbon black to the petroleum pitch increased its viscosity to a value higher than that for the coal tar pitch. In the light of these observations, an attempt was made to explore the possibility of using a petroleum pitch as a suitable binder. Ten per cent of carbon black was purposely added in the petroleum pitches to match with the quinoline insolubles that are invariably present in a coal tar pitch. The relative compatibility of coal tar and petroleum pitches has been estimated with regard to such characteristics as baked density, crushing strength and electrical resistivity of the baked carbons.

2. Experimental procedure

The filler used in the present study consisted of graded calcined petroleum coke with the compo-© 1978 Chapman and Hall Ltd. Printed in Great Britain.

TABLE I Composition of the calcined petroleum coke filler

36%
sh 24%
40%
$0.864 \mathrm{g}\mathrm{cm}^{-3}$
• 0.4% (by weight)

sition given in Table I. The uniformity in the particle size distribution of the filler was checked by its bulk density determination described elsewhere [6]. One kilogram of this filler taken five times was mixed with various proportions of pitch binders A, B, C, D and E consisting respectively of coal tar pitch (Ring and Ball softening point, $T_s =$ 78° C), petroleum pitch I ($T_s = 78^\circ$ C), petroleum pitch II ($T_s = 150^{\circ}$ C), (PPI + 10% carbon black) and (PPII + 10% carbon black). The various characteristics of these binders are given in Table II. The mixtures were roll-mixed for 20 min each and then ground cold to pass through 60 B.S. mesh. About 12 g portions from these powdered mixes were then moulded into cylindrical blocks of about 19 mm diameter under a load of 1200 kg at suitable temperatures. These blocks were measured for the green density determination and then baked in an electrical muffle furnace to a temperature of 950° C in a five-day baking cycle.

These baked carbons were finally subjected to apparent density, electrical resistivity and crushing strength measurements as described previously [7]. The data of these measurements are plotted in Figs. 1 to 4 and the results obtained have been summarized in Table III.

3. Results and discussion

Figs. 1 and 2 represent respectively the variation of the apparent densities of green and baked carbon mixes with the increase in binder content for various pitch binders. As is obvious, the green as well as baked densities of the product, in general, increase with the increasing content of the binder up to certain levels beyond which they start decreasing. The maximum green and baked apparent densities corresponding to the optimum levels of different binders are given in Table III. It is seen from this table that the apparent density of the green and baked carbon blocks is maximum for the carbon mix containing the coal tar pitch as binder compared to those containing the petroleum pitch binders.

Fig. 3 shows the variation of crushing strength of the baked carbon mixes with increasing proportions of the different binders. It is apparent that the crushing strength also varies in a similar manner as the apparent density, except for the binders B and D, where it goes on decreasing with their increasing contents. Furthermore the crushing strength of the blocks using the petroleum pitch I is less than 40% of the strength of ones using the coal tar pitch. However, with petroleum pitch II, the strength of the blocks increases sharply to about double that of the ones using petroleum pitch I. This increase may be attributed to the increase in the amount of β -resins in the former pitch over that in the latter, because the β -resins are known to impart binding strength to the carbon product [1]. In batches C and E, since the β -resin contents are more or less the same, the increase in strength of the blocks using the binder

Binder	Composition of binder	Density (gcm ⁻³)	Coking value (wt%)	Ash content (wt%)	B.I. (wt%)	Q.I. (wt%)	β-resins (wt%)	C (wt%)	H (wt%)	N (wt%)	S (wt%)	C/H (atomic)
A	Coal tar pitch ($T_s = 78^\circ$ C)	1.24	58	0.29	38.0	12.4	25.6	94.72	3.60	1.13	0.05	2.19
В	Petroleum pitch I $(T_s = 78^\circ \text{ C})$	1.03	34	0.09	3.5	3.5	0.0	84.93	8.9	1.41	0.71	0.80
С	Petroleum pitch II $(T_s = 150^{\circ}C)$	1.11	53	0.14	14.0	2.5	11.5	88.85	7.31	1.35	0.71	1.01
D	PPI + 10% CB	1.03	-	_	13.2	13.1	0.1	_	_	_	_	
Е	PPII + 10% CI	31.13	_	-	22.6	12.2	12.2	_		-		-

TABLE II Characteristics of different coal tar and petroleum pitch binders

 T_s = ring and ball softening point; PPI = petroleum pitch I; PPII = petroleum pitch II; CB = carbon black; B.I. = benzene insolubles; Q.I. = Quinoline insolubles.

Binder	Composition of	Characteristics of green and baked carbon mixes							
	binder	Green density (g cm ⁻³)	Baked density (g cm ⁻³)	Crushing strength (N mm ⁻²)	Electrical resistivity (mΩ cm)				
Å	Coal tar pitch ($T_s = 78^\circ$ C)	1.75 (37%)	1.65 (35%)	37.0 (32%)	3.2(38%)				
В	Petroleum pitch I ($T_s = 78^\circ$ C)	1.67 (25%)	1.45 (25%)	12.7 (20%)	3.4(32%)				
C	Petroleum pitch II ($T_s = 150^\circ$ C)	1.67 (30%)	1.49 (25%)	24.6 (25%)	4.9(28%)				
D	PPI + 10% CB	1.68 (25%)	1.50 (20%)	24.2 (20%)	4.8(30%)				
E	PPII + 10% CB	1.68 (37%)	1.60 (33%)	41.0 (30%)	5.1 (30%)				

TABLE III Comparative data of the characteristics of green and baked carbon mixes containing optimum contents of different coal tar and petroleum pitch binders.

 T_s = ring and ball softening point; PPI = petroleum pitch I; PPII = petroleum pitch II; CB = carbon black. Figures in parentheses denote the optimum binder content.





Figure 3 Variation of crushing strengths of baked carbon mixes with various proportions of different binders.

E over that of those using[†] the binder C may be attributed to the presence of carbon black in the former binder.

Fig. 4 represents the variation of electrical resistivity of baked carbon mixes with the increasing proportions of the various binders. It is apparent that the electrical resistivity of the blocks containing coal tar pitch is the least compared to that of those using any of the petroleum pitch binders. The use of petroleum pitch II results in an increase in the electrical resistivity of the blocks

when compared with petroleum pitch I. The higher values of electrical resistivity observed with binders D and E over binders B and C respectively may be due to the effect of carbon black.

It is interesting to note from the Table III that the apparent density, crushing strength and electrical resistivity of the blocks employing binder D (petroleum pitch I with 10% carbon black) are almost the same as those of the ones employing binder C (petroleum pitch II). Thus it can be said that the effect of the addition of 10%



Figure 4 Variation of electrical resistivities of baked carbon mixes with various proportions of different binders.

carbon black in petroleum pitch I is equivalent to that of increasing its softening point from 78 to 150° C. It may be further noted that the optimum amount of coal tar pitch for a particular filler is higher than the corresponding optimum amounts of the petroleum pitch binders B, C, D and E, as estimated from the above-studied characteristics of the carbon mixes. This may be due to the inherent differences in the natures of the coal and petroleum derived pitches.

It is thus apparent from the above discussion that coal tar pitch is the most suitable binder resulting in a product of high density, high strength and least electrical resistivity. However, a petroleum pitch of high softening point may be employed in those applications of carbon products for which a relatively higher electrical resisitivity is tenable. Correlating the low values of aromaticity of petroleum pitches I and II compared to that of the coal tar pitch, with the characteristics of the corresponding baked carbon mixes, it appears that a petroleum pitch of higher aromaticity may also prove to be a suitable binder.

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References

- 1. A. DARNEY, Industrial Carbon and Graphite, (Society of Chemical Industry, 1958) p. 152.
- 2. D. McNEIL and L. J. WOOD, *ibid.* p. 162.
- 3. T. H. BLAKELY and F. K. EARP, *ibid.* p.173.
- 4. L. P. CHARETTE and G. T. BISCHOFBERGER, Ind. Eng. Chem. 47 (1955) 1412.
- 5. G. BHATIA, R. K. AGGARWAL, S. S. CHARI and G. C. JAIN, Carbon 15 (1977) 219.
- 6. G. BHATIA, Carbon 14 (1976) 311.
- 7. G. BHATIA, R. K. AGGARWAL and P. RANJAN, J. Mater. Sci. 12 (1977) 639.

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