Acta Geodyn. Geomater., Vol.2, No.4 (140), 91-96, 2005

INVESTIGATIONS OF COKING COAL BLENDS CONTAINING A COAL TAR PITCH-BASED CARBONACEOUS WASTE COMPONENT TOWARDS UTILIZATION IN COKE OVENS

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(*Received* June 2005, *accepted* November 2005)

ABSTRACT
Herein, are presented results of laboratory-scale investigations of co-pyrolysis of artificial coking components made of carbonaceous waste materials and a coal tar pitch (labeled synthetic coking coal or SCC) in admixture with a typical coal blend used in coke manufacturing in Poland. The solid SCC component is prepared by combination of organic wastes and a molten coal tar pitch at elevated temperatures in a laboratory mixer followed by water quenching to room temperature. The results indicate the utmost utility of a range of otherwise environmentally troublesome carbonaceous wastes in the classical coking process.

KEYWORDS: organic wastes, waste utilization, pitch, pyrolysis, coking, smokeless fuel

INTRODUCTION

The environmental pollution and protection problems are closely and inseparably related to ever increasing volumes of industrial and municipal waste materials. Especially, such carbonaceous wastes as plastics pose acute problems due to their relatively slow intrinsic disintegration processes (Górka, 1997).

The specific recovery and recycling as well as combustion processes are perhaps the most obvious options to-date to utilize such wastes. In this regard, especially, the combustion of municipal wastes seems to be a preferred method in many industrialized countries (Stelmach and Sobolewski, 1999). However, in Poland more than 90 % of the wastes are deposited in open air waste grounds.

It appears that pyrolytic methods of organic waste utilization have recently become a subject of increased scrutiny for potential applications. This trend dwells on hopes for a more effective utilization of high carbon containing materials. Many research centers have been especially interested in pyrolytic methods applied to plastic wastes since their volume has been increasing systematically in recent decades. In this respect, the use of heavy liquid carbochemical and/or petrochemical fractions as co-additives in such systems seems to be very promising (Nomura et al., 2003; Ku-Joo et al., 1999; Wasilewski and Sobolewski, 1999).

Coal tar pitches resulting from thermochemical processing of coal tars constitute mixtures of numerous compounds of various chemical nature and diverse molecular structures. It has been established that the pitches' average chemical composition as well as some specific properties are similar to the respective values of vitrinites in hard coals of medium degree of metamorphism (coking coals). These are the features that should potentially enable the mixtures of carbonaceous wastes and pitches to yield a useful "artificial" coking component. Such a component as an additive in a coal blend could, in principle, be utilized in a typical coke-oven to eventually yield, on the one hand, a valuable smokeless coke fuel while, on the other hand, the associated lighter by-products such as gas and tar could be appropriately processed in the existing coke-plant treatment sections.

In this report, are presented laboratory-scale investigations of (i) the solid component called synthetic coking coal, SCC that is prepared by mixing selected carbonaceous wastes with a coal tar pitch and (ii) composite mixtures of SCC with a typical coking coal blend (CB) used in the stamp charging method of coke manufacturing in Poland.

EXPERIMENTAL

PREPARATION OF SYNTHETIC COKING COAL, SCC

A coal tar pitch with a softening point T_{Mett} of 82 ˚C and Coking Number LK of 44 % (specific carbonization yield) was used in admixture with the following materials:

- wood mash/saw dust;
- used fabrics (used clothing);
- used insulating graphite fill from Acheson furnaces;
- crumbled waste plastics.

The materials were mixed in the following proportions (weight percentages): wood mash/saw dust – 16 %, used fabrics – 15 %, insulating graphite fill – 38 %, crumbled waste plastics – 8 %, and coal tar pitch – 23 %. The fabrics and plastics were cut into pieces smaller than 5 mm prior to mixing. The components were placed in a heated rotary mixer (5 kg nominal charge) and homogenized in a medium of the molten pitch at 130 ˚C for 30 minutes. Subsequently, the hot liquid mixture was discharged and quenched with water. The solidified product was then crushed in a splitter followed by sampling for individual investigations. For subsequent studies of coking and caking material properties, typical coal blends used in the stamp charging method were used both in admixture with the waste/pitch SCC additive prepared as above, and as an unaltered reference coal blend system. The 5 wt. %, 10 wt. %, and 20 wt. % proportions of the SCC component were used with the initial coal blend.

CHARACTERIZATION METHODS

The following characterization methods were used for: water content – Polish Standard PN-80/G-04511, ash content – Polish Standard PN-80/G-04512, volatile matter content – Polish Standard PN-98/G-04516, Roga Index – Polish Standard PN-81/G-04518, Free Swelling Index – Polish Standard PN-81/G-04515, dilatation properties – Polish Standard PN-81/G-04517, and yield of low temperature carbonization products – Polish Standard PN-75/G-04540.

RESULTS AND DISCUSSION

The major physicochemical properties of the synthetic coking coal, typical (unaltered) coal blend, and composite mixtures of both feed stocks are included in Table 1. Table 2 presents the characteristic dilatation temperatures while Figures 1 and 2 show the derived dilatation curves. In particular, Figure 1 includes the curve for SCC and Figure 2 shows the curves for the reference coal blend and its mixtures with SCC.

The data in Table 1 suggest that even the highest 20 wt. % addition of SCC does not significantly influence the ash content (A^d) and volatile matter content (V^{daf}) in the composite blend. Thus, the initial coal blend has A^d of 6.2 % and V^{daf} of 32.4 % to be compared with the respective values of 6.4 % and 31.1 % in its composite with SCC. Somewhat bigger impact of SCC is noticed in the values of Roga Index and associated caking properties. The 5 wt. % and 10 wt. % proportions of SCC in the mixtures cause a relative decrease of Roga Index by approximately 5 units while the 20 wt. % SCC content is accompanied by the more pronounced decrease of 17 units.

The investigations of the dilatation properties show a diverse influence of the SCC content. The proportion of SCC appears to affect also the shape of the dilatation curves. For example, the least deviation from the curve for the unaltered blend has a curve for the 5 wt. % SCC mixture while 20 wt. % of SCC results in substantial changes of both dilatation and contraction. Thus, at the 5 wt. % and 10 wt. % levels of SCC, the dilatation is merely increased by 1 % and 5 %, respectively, while at the 20 wt. % level of SCC it is relatively increased by some 22 %. It also follows from Figure 2 and Table 2 that the increased contents of SCC result in the simultaneous decrease of the melting point, T_I, and temperature of contraction, T_{II}, as well as in widening of the temperature range of coal plasticity, T_{III}-T_I.

The SCC component itself is characterized by, relatively, a low water content (hydrophobic material) and a low volatile matter content. In general, the coal blend with 5 wt. % SCC appears to have the closest characteristics to the unaltered initial blend. On the other hand, the increased proportions of SCC result in the visibly decreased values of Roga Index (RI) and Free Swelling Index (SI) of the composite blends.

Table 1 Characteristics of initial coal blend (unaltered), synthetic coking coal (SCC), and composite mixtures of both components

Sample		Initial coal blend	SCC component	Proportion of SCC in coal blend		
				5 wt. %	10 wt. %	20 wt. %
Water	W^a [%]	7.5	0.2	-	-	-
Ash	A^d [%]	6.2	6.7	6.3	6.3	6.4
Volatile matter	V^{daf} [%]	32.4	23.2	31.9	31.6	31.1
Roga Index	RI	77	24	71	72	60
Free Swelling Index	SI	7.5	1	7	6.5	6
Dilatation characteristics	Contraction [%]	32	20	30	32	19
	Dilatation [%]	71	-	72	76	93

Table 2 Characteristic dilatation temperatures

Sample	TEMPERATURE [˚C]			
	Melting T_I	Contraction T_{II}	Dilatation T_{III}	Plastic state T_{III}-T_I
Initial coal blend	370	420	485	115
SCC component	75	105	-	-
Coal blend/5 wt. % of SCC	360	415	475	115
Coal blend/10 wt. % of SCC	350	410	470	120
Coal blend/20 wt. % of SCC	320	400	485	165

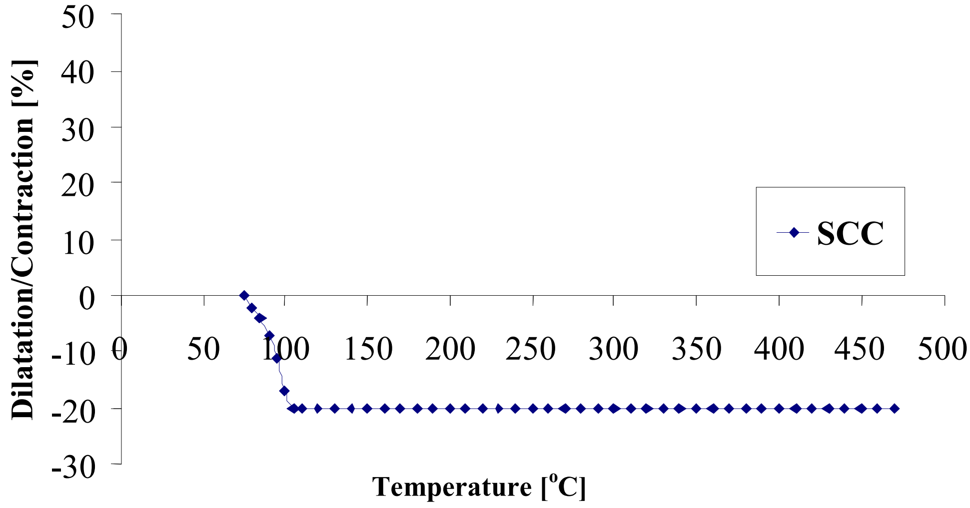

Fig. 1 Dilatation curve for the synthetic coking coal (SCC)

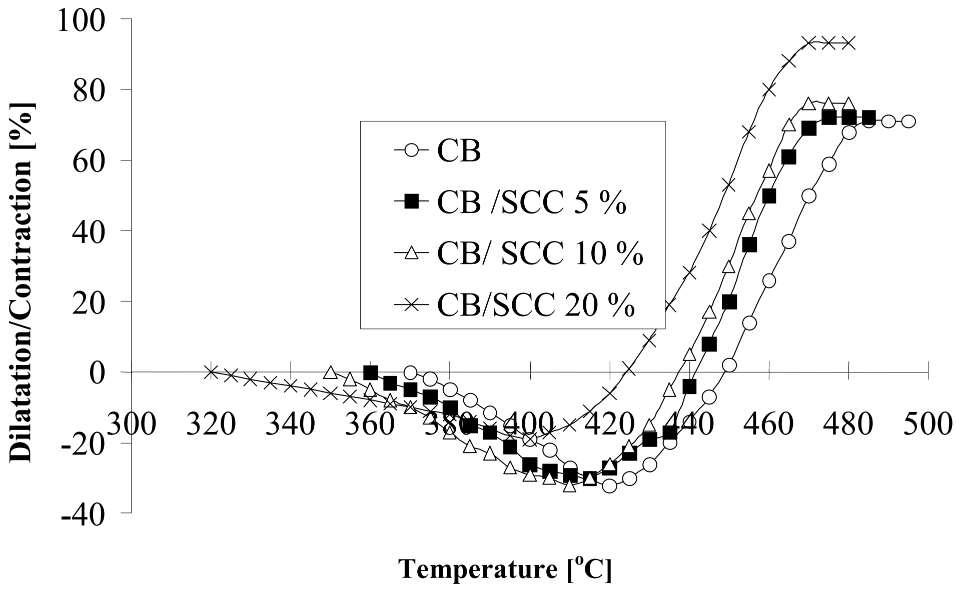

Fig. 2 Dilatation curves for the initial coal blend (CB) and its mixtures with 5 wt. %, 10 wt. %, and 20 wt. % of synthetic coking coal (SCC)

Table 3 Yield of low temperature carbonization products

Sample	Yield [wt. %]			
	Coke	Tar	Water	Gas
Initial coal blend	77.6	11.7	8.6	2.1
SCC component	82.5	12.3	4.4	0.8

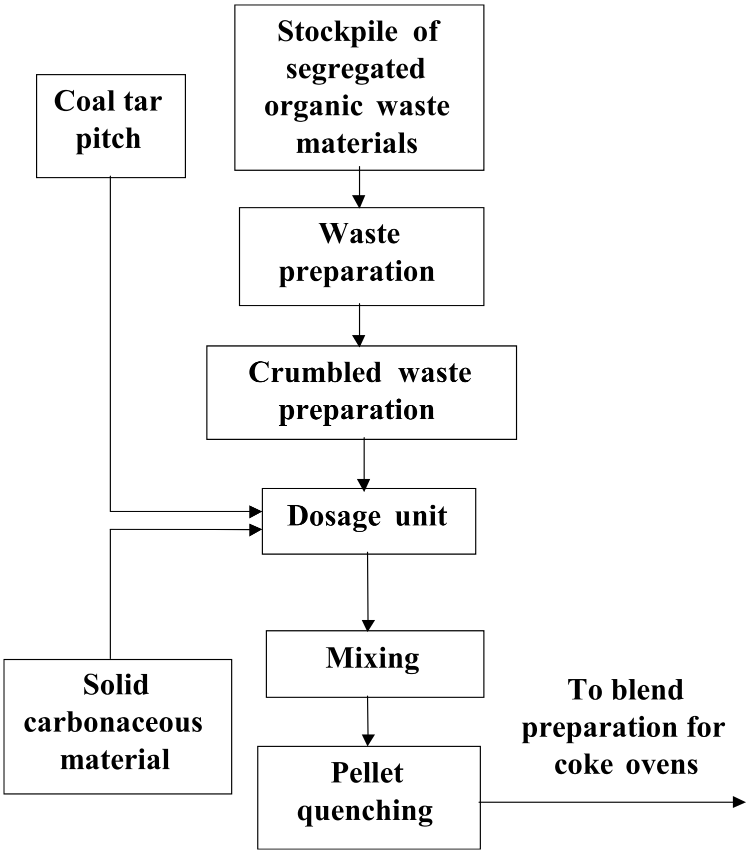

Fig. 3 Schematics of the process for utilization of carbonaceous waste materials in classical coke manufacturing

The data in Table 3 for the low temperature carbonization products indicate the product diversified yields upon comparison of the initial (unaltered) coal blend and the individual SCC component. The latter material is characterized by a significantly higher yield of coke and, consequently, by lower yields of the lighter carbonization by-products.

In this area, further studies are planned to be focused on investigations of coke properties manufactured from coal blends containing the synthetic coal component. However, the data discussed here form a solid basis for our proposition of the process for organic waste utilization as summarized below.

PROCESS FOR MANUFACTURING THE SYNTHETIC COKING COAL FROM COAL TAR PITCHES AND CARBONACEOUS WASTE MATERIALS

Based on the laboratory-scale studies, herein, we contest that there is a new potential for a feasible utilization of a range of industrial and municipal carbonaceous waste materials. The proposed process is aimed, first, at converting the waste materials into a solid by-product that is similar in its characteristics to typical components of coking coal blends and, second, utilizing it in this form as a suitable and valuable additive in classical coke manufacturing.

The method includes the initial class/type segregation of the wastes followed by mixing them in appropriate proportions with a coal tar pitch. After this stage, one obtains a solid, mechanically stable component (SCC) of suitable coking properties (Żmuda et al., 1998a; Tora and Żmuda, 1998; Żmuda et al., 1998b; Żmuda et al., 1998c; Żmuda et al., 1999; Żmuda and Minkina, 1999). The final stage of the process consists of controlled admixing of SCC to a coal blend charge to be used in classical coke ovens.

Both the classical stamp and free charging methods of coke manufacturing are suitable for the utilization of SCC without any additional and/or crucial process adjustments. Additionally, in the former method, the SCC "pellet" component may positively inflict upon increasing the overall density of the charge. In the coke oven, the SCC additive is subsequently co-pyrolyzed with other components of the charge to yield a mechanically stable coke and raw gases. The pyrolytic gases are treated in the appropriate gas condensation/utilization sections of the plant. Concerning the coke product, its properties will depend, among others, on the characteristics of the coal blend including the quality and quantity of the SCC additive.

Figure 3 presents the schematics of the technological process for the preparation of synthetic coking coal as the means for the utilization of a range of carbonaceous waste materials.

The major advantages of the process can be summarized as follows.

- Thanks to the specific properties of coal tar pitches (relatively high softening temperatures, good rheological properties upon melting, relatively high coke yields upon carbonization), one can use them in the system with carbonaceous wastes to prepare solid by-products of suitable coking properties to be utilized in classical coke manufacturing.
- The synthetic coking coal SCC is obtained in a simple way by mixing the wastes with the pitch at relatively low temperatures above the pitch's melting point.
- The pitch does not undergo any pyrolytic decomposition during mixing and, therefore, not contributing to environmental pollution at that stage.
- Thanks to the relatively high softening temperatures of tar pitches, the pellets of SCC are compact and sufficiently mechanically stable at ambient conditions to be transported without any special precautions.
- It stems from the above that no otherwise energy consuming pressing and/or briquette forming stages are necessary.
- The matrix of the solid carbonizate formed during pyrolysis of the pitch advantageously binds the solid particles of the organic wastes during intermediate stages of the process.
- The SCC pellets can be forecast as a valuable component of coking coal blends with no deleterious effects either on the classical coking process or essential coke/gas products quality parameters.
- The process enables some control over ash content, ash composition, and volatile matter content in SCC by adjusting the type and amount of the waste.
- Combining the carbonaceous waste utilization with classical coke manufacturing is thought to result in significantly reduced relative waste utilization costs while, at the same time, yielding the known valuable products (smokeless coke fuel, gas, tar, etc.).

The proposed process can be looked upon as an alternative if not replacement for the existing methods of thermal waste utilization, especially, those relying on pyrolytic waste decomposition. In our proposition, existing coke plants can include the synthetic coking coal as an additive or a substitute in the coal blend up to several weight percentages. The possibility to control the input physicochemical properties of SCC (e.g., ash, volatiles) adds an additional flexibility to process characteristics in classical coke manufacturing. Finally, the SCC additive appears to be especially well suited for the production of smokeless coke fuel and/or metallurgical coke with demanding mechanical properties.

ACKNOWLEDGMENT

This work was sponsored by Polish Commitee for Scientific Research KBN/AGH University of Science and Technology grant No. 11.11.210.62.

REFERENCES

Górka, K.: 1997, Ekonomiczne aspekty gospodarowania odpadami przemysłowymi i komunalnym, presentation at TECH-EKO 97; University of Agriculture, Kraków, Poland; September 11-12, (in Polish).

Ku-Joo, H., Hool, J.N. and Curtis, C.W.: 1999, Determination of effective conditions for two-stage coprocessing of coal with waste plastics and petroleum residues; Energy and Fuels, 13, 1128-1134.

Nomura, S., Kato, K., Nakagawa, T. and Komaki, I.: 2003, The effect of plastic addition on coal coking properties during carbonization; Fuel, 82, 1775-1782.

Stelmach, S. and Sobolewski, A.: 1999, Piroliza – alternatywa spalania odpadów komunalnych?; Gospodarka Paliwami i Energią, 7, 17-21, (in Polish).

Tora, B. and Żmuda, W.A.: 1998, The organic waste utilization in the process of synthetic coking coal production; Proceedings of the IV International Conference on Clean Technologies for the

Mining Industry, vol. II, 921; Santiago, Chile, May 13-15.

Wasilewski, R. and Sobolewski, A.: 1999, Energetyczna waloryzacja odpadowych tworzyw sztucznych; Gospodarka Paliwami i Energią, No. 9, 22-25, (in Polish).

Żmuda, W.A. and Minkina, M.: 1999, A method for the utilization of the organic waste materials; Tenth International Conference on Coal Science – Prospects for Coal Science in the 21st Century; Shanxi Science & Technology Press, vol. II, 1219-1222; Taiyuan, P. R. China; September 12-17.

Żmuda, W.A., Długosz, A., Woyciechowski, W. and Budzyń, S.: 1998a, Metoda utylizacji materiałów odpadowych pochodzenia organicznego w procesie koksowania węgli kamiennych; Gospodarka Paliwami i Energią, No. 3, 11-13, (in Polish).

Żmuda, W.A., Komorowska-Czepirska, E. and Czepirski, L.: 1999, Laboratory preparation of carbonaceous adsorbents from waste materials; Acta Montana IRSM AS CR, Series B, 9(112), 81-85.

Żmuda, W.A., Pusz, S., Koszorek, A. and Minkina, M.: 1998b, Co-carbonization of the granulated organic wastes blended with a coal mixture; Acta Montana IRSM AS CR, Series B, 8(110), 59-66.

Żmuda, W.A., Tora, B. and Woyciechowski, W.: 1998c, The results of industrial process of utilization of organic wastes by method of hard coal coking; Symposium Proceedings, DGMK, Velen, Germany, Tagungsbericht, vol. 9802, 399-403.