



# Co-pyrolysis of waste tire/coal mixtures for smokeless fuel, maltenes and hydrogen-rich gas production



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## ABSTRACT

The processing of waste tires with two different types of bituminous coal was studied through the slow co-pyrolysis of 1 kg of waste tire/coal mixtures with 15, 30 and 60 wt% waste tires on a laboratory scale. The waste tire/coal mixtures were pyrolysed using a quartz reactor in a stationary bed. The mixtures were heated at a rate 5 °C/min up to the final temperature of 900 °C with a soaking time of 30 min at the required temperature. The mass balance of the process and the properties of the coke and tar obtained were evaluated, further, the influence of the admixture in the charge on the amount and composition of the obtained coke and tar was determined. It was found that the smokeless fuel/carbonaceous sorbent and a high yield of tar for further use can be obtained through the slow co-pyrolysis. The obtained tars contained mostly maltenes (80–85 wt%). FTIR analysis showed that the maltenes from the co-pyrolysis of coal/waste tires exhibited significantly lower aromaticity as compared with that from coal alone. The gas obtained from pyrolysis or co-pyrolysis of waste tire/coal mixtures contained a high amount of hydrogen (above 60 vol%) and methane (above 20 vol%).

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## 1. Introduction

A major global economic and environmental burden is caused by the disposal of waste tires – they are non-biodegradable and their amount continuously increases with the growing production of the automobile industry [1]. Waste-tire production in Europe is estimated to be around 3.3 million tons per year [2,3], with some states, such as e.g. Bulgaria or Cyprus, only depositing them at landfills. The Czech Republic annually produces approximately 60–70 thousand tons of waste tires, and this number continues to increase [4]; about 30% of this waste is landfilled. Therefore, it is important to deal with its use, e.g. in the construction industry [5], or with its effective chemical treatment. Since waste tires usually contain 45–48% of elastomer, 21–22% of soot, 15–25% of steel reinforcement, 0–5% of textile, 1–2% of ZnO, 1% of sulfur and 6–8% other chemicals (accelerators and activators, fillers, pigments, plasticizers, antioxidants, etc.), they are a valuable source of products with a high utility value.

An effective industrial treatment process is pyrolysis, which is used to transform waste tires and industrial plastic wastes into fuel oil (40–45%), carbon black (30–35%), steel wires (10–15%) and gas (10–12%) [6]. The main product obtained through waste-tire pyrolysis is oils, which have broad application. High oil yields from the

pyrolysis of waste tires alone were achieved by e.g. Fernández et al. [7], who studied the influence of temperature on product yield. Other than classic pyrolysis, it is possible to use co-pyrolysis either with coal [8] or with waste of vegetable origin [9,10]. Acevedo et al. [11] pyrolyzed 6 g of a mixture of coal and scrap tires in the ratio 1:1 at a temperature of 850 °C and a heating rate of 5 °C/min, obtaining up to 37 wt% of tar. Co-carbonization for blast furnace coke production was also studied as a possible route for the recycling of waste tires [12]. The possibilities of using waste tires for the production of hydrogen at low manufacturing costs [13] and co-gasification of waste tires with lignite [14] were investigated as well. Waste plastics can be treated in the same way [15,16].

The literature insufficiently stresses the importance of the preparation of waste tires prior to processing. Because this waste contains steel components, they must be separated by a suitable method [17]; furthermore, it is necessary to select a proper grain size corresponding to the method of thermal treatment and scale of processing. If the feedstock is well prepared, co-utilization of coal with waste tires offers a number of advantages. It is possible to obtain a hydrogen-rich gas [8]; the current study showed a synergetic effect that result in improving the quality as well as quantity of tar [18]; therefore, alternative fuels can be obtained in order to reduce the consumption of fossil fuels. Another advantage is that the co-pyrolysis and co-gasification of waste tires can be successfully carried out on a commercial scale [14].

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This paper studies the co-pyrolysis of waste tires with two different types of bituminous coal on a laboratory scale and examines the properties and composition of the coke as a smokeless fuel or carbonaceous sorbent, the tar and gas obtained as well as their possible uses.

## 2. Experimental

### 2.1. Materials

Waste tires with a grain size of 1–5 mm containing less than 50% of natural rubber were used for the co-pyrolysis of model mixtures. Steel parts were removed from the waste tires by magnetic separation [17]. The tires were subjected to thermal decomposition with two types of bituminous coal from the Upper Silesian Basin. One of them was Polish gas coal (A) from the Rydułtowy-Anna Coal Mine (36.4% of volatile matter), the other was Czech coking coal (B) from the ČSM Mine, Ostrava-Karviná District (29.2% of volatile matter). The grain size of the coal was 0.5–4 mm (close to grain size of waste tires). Table 1 shows the results of the proximate and ultimate analyses of input bituminous coals and waste tires.

Proximate and ultimate analyses were carried out according to ISO standards (687:2010, 562:2010, 1171:2010, 333:1996, 334:2013, 625:1996, 157:1996). For all the samples below, the oxygen content was determined by recalculation to 100%. For the samples of coal, waste tires and coke, the higher heating value was determined calorimetrically and the lower heating value was calculated according to the ISO 1928:2009 standard.

### 2.2. Methods

The slow pyrolysis and comparative analyses of coal alone and their mixtures with 15, 30 and 60 wt% waste tires were performed on a laboratory unit with a fixed bed and a quartz reactor (outer diameter 100 mm, inner diameter 95 mm, length 500 mm) placed horizontally in an electric resistance furnace. Mixtures of coals A or B with 15, 30 and 60 wt% of waste tires were co-pyrolyzed. The weight of the charge was always 1 kg. Based on previous experience [8,15], the charges were heated at a heating rate of 5 °C/min up to the final temperature of 900 °C with a soaking time of 30 min at the required temperature. Volatile products from slow pyrolysis/co-pyrolysis were released during thermal decomposition. (The gas products released from about 200 °C, the tar formed at about 300–400 °C [8], therefore, the main pyrolysis reactions started at about 300 °C (waste tires, see below) and 400 °C (coal, see below).) The volatile products were led from the reactor into a water cooled flask (Fig. 1, position 3) and further in a vertical cooler (Fig. 1, position 4) with cooling water (Fig. 1, position 5). The temperature of cooling water ranged from 8 °C to 12 °C. The liquid portion (water and tar) formed during the experiment was captured and collected in a water-cooled flask after its separation from the stream of raw gas (Fig. 1, position 3). The generated gas was continuously analyzed and subsequently burned in an auxiliary burner (Fig. 1, position 6). For analysis of a gas, a 350 dm<sup>3</sup> gasholder (Fig. 1, position 6a) was incorporated in the apparatus

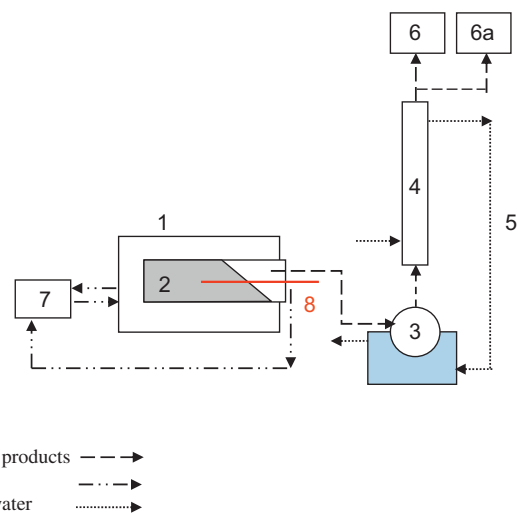


Fig. 1. A functional schema of the laboratory unit with a horizontal bed. 1 – electric furnace, 2 – quartz reactor with a charge, 3 – cooled flask for liquid products, 4 – cooler, 5 – cooling water, 6 – burner, 6a – gasholder, 7 – programmable furnace heating, and 8 – thermocouple.

for a total gas analysis (the auxiliary burner was replaced with gasholder). The gas from pyrolysis of coals A and B and their mixtures with waste tires (T) was analyzed. Water from the resulting liquid portion was separated by azeotropic distillation with n-hexane. The coke obtained was characterized, in addition to common parameters, also by its true and apparent density, porosity and resistivity. This solid product was characterized by the aforementioned analysis and can be considered as smokeless coke. A functional schema of the co-pyrolysis laboratory unit is shown in Fig. 1.

Because of the multicomponent character of the tars obtained, they were divided into groups of chemically similar compounds, with asphaltenes and maltenes being separated from the tars. The asphaltenes were prepared by precipitation with n-hexane in two stages [19,20]. In the first stage, most substances soluble in n-hexane with a fiftyfold excess were removed from the tars [21,22]. The suspension was filtered for 24 h. The residues on the walls of the container were dissolved in dichloromethane, and asphaltenes were again precipitated from the solution by n-hexane. Afterwards, the dichloromethane was distilled off and the asphaltene floccules were filtered off. The n-hexane was removed from the maltene solution obtained in a vacuum evaporator and its residues additionally in a vacuum drier at 70 °C and a pressure of 4 kPa. Under these conditions, volatiles, i.e. the BTX (benzene, toluene, xylene) fraction and the oxygenates, mainly phenol, were removed as well.

The chromatographic fractionation of the maltenes obtained was performed in a glass column of a length of 700 mm and an inner diameter of 6.9 mm, provided with brass caps at the ends. The bottom half of the column was filled with fully activated alumina (150 mesh, activated for 8 h at 400 °C) and the upper half

Table 1

The properties of the coals and waste tires used. A – Anna coal, B – ČSM coal, T – waste tires. V – volatile matters, HHV – higher heating value.

Sample	Proximate analysis (wt%)				Ultimate analysis (daf, wt%)					HHV (MJ/kg)
	Water <sup>a</sup>	Ash <sup>d</sup>	S <sub>t</sub> <sup>d</sup>	V <sup>daf</sup>	C	H	N	S <sub>o</sub>	O <sub>dif</sub>	
A	3.23	7.55	0.54	36.38	84.10	5.43	1.53	0.50	8.44	34.16
B	0.76	8.06	0.57	29.15	86.89	5.30	1.45	0.45	5.91	35.01
T	1.04	6.70	1.09	70.20	88.22	8.18	0.47	0.80	2.33	38.21

daf – dry ash free basis; a – analytical; d – dry; t – total; o – organic; dif – by difference.

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