

# BLACK CARBON PRODUCTION FROM PYROLYSIS AND COMBUSTION OF PYROLYSIS OIL OF ASPHALTITE, WASTE TIRE AND WOOD

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**Abstract:** Ethylene, natural gas and oil is used as the raw material in the combustion production of black carbon by soot formations. Black carbon is useful with the increased surface area as soot or activated carbon micro particles. It is traditionally believed that soot formation occurs in combustion of oil by certain amount of oxygen. As a result of this, a gas concentration gradient is needed between the carbon and the oxygen for soot production from the gaseous or liquid fuel. Therefore, certain amount of carbon can only form for soot compounds that readily provide submicron black carbon product. Black carbon involves allotropic soot produced by not full combustion and removal of carbon matter in a closed-loop circulating batch system.

This study searched that firstly, pyrolysis of low ash Şırnak asphaltite and washed with flotation device provided less ash Turkish lignites (less than 10% of the level in existing advanced clean lignite washery plant manufactured) with waste wood and tyre, secondly, combustion of pyrolysis oil of the retort for black carbon production on which the furnace gas parameters by examining the highest black carbon efficiency is obtained, and thirdly, according to the optimum design parameters of the pilot plant, gas furnace equipment investigated. The 32% soot yield from the 50% coal and 20% wood and 30% waste tyre weight rate were provided in the retort furnace.

**Keywords:** coal pyrolysis, pyrolysis oil, coal soot, lignite pyrolysis, black carbon; biomass pyrolysis oil

## INTRODUCTION

Lignite consumption of our natural resources in energy production is increasing in parallel with the increasing energy needs today (IEA, 2012). In terms of consumption and production quantities of our high-low thermal valuable lignite reserves are limited (TKI 2009, TTK 2009). Depending on the economic technology enables the production of advanced technological developments needed coal derived products. Compliance with environmental norms coal pyrolysis or gasification facility allows the production of liquid and gaseous fuels needed with today's modern technology (Bell et al. 2011). However, the method requires a variety of raw materials and chemicals and adaptation to the type of processing fuel material. It has also been shown that black carbon occurs during the time course of diesel or other liquid fuel combustion processes that are based on bad combustion (Shadle et al. 2001, Sharma et al. 2008). Most of the studies aiming at black carbon quantifying performance were performed using offline systems because of difficulties encountered in quantifying soot during combustion of ethylene or waste rubber (Amal et al. 2011). Black carbon formation is dependent on several factors including combustion parameters, temperature

and type of fuel combustion, characteristics of furnace and combustion process configuration, and it can be optimized by varying the operational conditions (Rodriguez-Mirasol et al. 1994).

Black carbon (BC) is widely used as fill for high quality tire production (Anonymous 2013, Anonymous 2009), also high conductive graphite rods production, also as soot for production of paints and construction materials resistive fire. It may also thought as soot source as activated carbon fill for treating wastewaters contaminated with phenols, volatile acids, aromatic and aliphatic organics.

Basically better quality lignite oil production, high value-added light oil, to produce black carbon products have been identified as key objectives. For the production of black carbon, high amount of oil (due to just about 7-20% production rate) is needed by the countries in the production of pyrolysis lignite oil as much clean possible. Acetylene is used to produce high quality soot black products in the reactor and (Amal et al. 2010, Guerrero et al. 2005, Guerrero et al. 2008, Mendiara et al. 2007). Therefore further cleaning of Şırnak asphaltite and Turkish lignite, clean product of pyrolysis with the production of liquid fuels will enable the development of South-East Anatolia and will also further enhance industrial

development and diversification and supply of industrial energy fuel (Tosun 2012).

In this study, we are addressing the enhancement of soot formation by pyrolysis oil and CO<sub>2</sub> gas, the definition and mechanisms of combustion, the relationship between the temperature and CO<sub>2</sub> gas partial pressure, the factors affecting soot formation, the methods for determination and quantification of soot formation and the mathematical models of combustion. Future research is still required to determine the optimum conditions for an increased performance for other types of oil. Particularly, factors such as the black carbon type, nature of the soot community and optimum process configuration need further investigation. For this purpose it is necessary to provide basic pilot knowledge of the industry working together with universities. Mainly due to improving yields in performing quality raw materials and advanced processes for performance testing processes according to the nature of our lignite-to-date with research institutions and technological applications are required. This study examined the beneficiation from our high-sulfur lignite. However, technologies are examined on the basis of lignite and lignite based on raw materials as well as the contribution of forest biomass and biogas plants waste cellulosic waste can be processed together with our lignite in the contribution rate as 15-30%.

The modeling approaches and combustion with high CO<sub>2</sub> gas content in combustion medium other than the fuel type especially wood oil and pyrolysis oil of asphaltite.

## **PRODUCTION OF PYROLYSIS TAR, HEAVY OIL FROM COAL**

Many studies have investigated the numerous advantages of adding pyrolysis oil to activated combustion systems (Kegl 2011). The presence of carbon dioxide in conventional systems improves soot settling, improves soot thickness, increases soot removal, improves removal of soot, reduces the impact of organic shock loadings, increases black carbon and oxygen concentration at the surface of soot carbon, increases removal, suppresses, improves, and reduces bulking (Neeft et al. 1997).

In BC production, chemicals is enhanced through the adsorption of inhibitory substances as in the catalytic processes, but in addition, the BC used in the furnace serves as a supporting medium for iron film colon (Liu et al. 2002, Wei-Biao et al. 2001).

## **Factors Affecting Pyrolysis and Combustion for Black Carbon**

Effective carbonization processes depend on numerous factors including coal rank in carbonization, the volatile gaseous matter of coal such as presence of hydrogen, carbonyl gas and carbonization rate (Mendiara 2007) so stabilizing the desorption, the settings of optimal diffusion conditions including structure defects (nitrogen, phosphorus, sulfur, etc.), temperature, oxygen content of coal, etc. and optimization of carbon dioxide concentration ratios (Amal 2011, Amal 2010) added the adsorption-desorption balance, the residence time and the spatial distribution of molecules in coal pores among other factors determining the efficiency of carbonization. Guerrero et al. (2008) also included the carbon reactivity, the adsorption characteristics as factors affecting the rate and extent of carbonization much dependent on the site activation, its gas desorption properties and its porosity (Bell et al 2011). Carbonization is a prerequisite step for oil generation and soot formation from tyre waste, biomass wastes and coal.

### **Coal particle size**

A major reason is that the retention time in fixed film processes is longer than in solid-gas processes. This allows more time to the carbonization for cracking to the desorbed persistent compounds. Furthermore, high rank coals allows an sufficient intimate contact between surface pores and gas atmosphere in the furnace due to more gas desorption (Kajitani et al. 2006)

### **Coal porosity**

The porous structure of activated carbon is a factor that determines to a great extent both the rate and degree of carbonization (Shadle et al., 2001). Sharma et al. (2008) found that, a mesoporous coal was more efficiently carbonized than a microporous coal.

Phenol molecules that may undergo an oxidative coupling reaction may be irreversibly adsorbed on coal, which in turn may result in low carbonization efficiency. Phenoxy radicals formed by the removal of a hydrogen atom from each phenolic molecule can participate in direct coupling with other phenoxy radicals at even room temperature, coal surface serving as a catalyst.

Carbonization efficiencies exceeding the total desorption abilities during increased fast pyrolysis on coal and wood were also reported by Tosun (2013).

## Surface properties of coal - Reactivity

(BET N<sub>2</sub>) specific surface area, total surface activity, oxygen functional groups, total surface impurities, metal concentrations, dielectric value, free radical concentration and reactivity of coal were related to the carbonization activity. However, in some investigations, the pore size distribution of coal is also greatly to affect pyrolysis kinetics (Guerrero et al. 2005).

## Combustion of Pyrolysis Oil of Waste Tire, Wood and Biomass

Soot matter removal during BC treatments results from the combined effect of adsorption and degradation. The efficiency of the combined combustion–soot formation process is higher than expected for either soot formation or production alone. Black carbon (BC) provides an attachment surface for pollutants and protects them from shock loadings of toxic and inhibitory materials, whereas the black carbon. High inert gas processes using catalyst carbon as carrier for iron film attachment are efficient soot from ethylene. However, in catalytic systems the gas attachment to surface is less efficient than in iron film or in fluidized bed reactors using CO<sub>2</sub> and pellets as iron film carrier (Jess et al. 2009, Schurtz et al. 2009). This is because, in the latter, then retention time of solids is generally much higher than in black carbon processes, allowing more time for gas attachment to BC.

On the other hand, in suspended growth systems, the use of BC is more advantageous than Granule BC since Powder BC systems provide a uniform distribution of solids with a minimum energy requirement for grinding.

In summary, in spite of the contradictory hypotheses, based on the available literature, it appears that pyrolysis of oil can occur when the condensate is removed from the liquid phase through carbonation activity either in low or high temperature processes. Certainly, a oil concentration gradient should establish for a continuing carbonation. Pyrolysis is extremely dependent on both carbonation and cracking hydrocarbons in coal. On the other hand, cracking reactions may increase the adsorbent of carbonyls. However, available data do not allow determining if the oil generation phenomenon depends solely on a mechanism involving absorption or if it also involves pore activities. Further work is required to support either one of these hypotheses.

## Production of Black Carbon from Pyrolysis Oil

### Oil Combustion

Considerable black carbon production on oil combustion and natural gas is managed as seen in Figure 1.

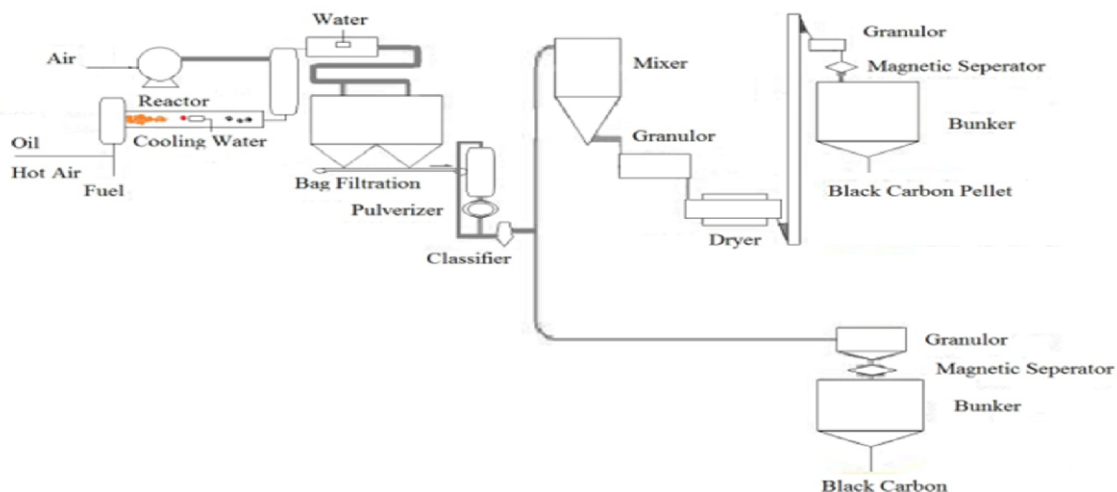


Figure 1, Use of fuel gas and pyrolysis oil subjected to black carbon production flowsheet.

The research over coal pyrolysis and gasification has been conducted over the years, but the pyrolysis results are widely dispersed because of the complex chemistry of coal (Bell 2011, Kajitani et al. 2006). Time related coal-

pyrolysis modeling assumes basically first-order kinetic equations, or less sensitive for heating rate (Donskoi et al. 1999). The other distributed activation model is dependent on the heating rate. The last two more advanced models need

three and four constants, respectively, which basically depend on the coal properties but also cover to some extent, the effect of heat-and-mass transfer phenomena (Liu et al. 2011). The reaction rate of char is influenced mainly by chemical and physical factors, which include coal type, catalytic effect of the ash and the specific surface area of char (Çakal et al. 2007), which changes during the reaction course with the development of internal pores, and finally, their destruction (Wiktorsson et al. 2000). In the case of the scaling-up procedure, the uncertainty of a complex model of the reacting system may be very high and it is reasonable under some conditions to use a methodology based on quasi-equilibrium conditions, which can be reflected at a larger scale.

## MATERIALS&METHOD

The representative samples were taken from local areas of the lignite. Fundamentally, the conditions regarding better desulfurization way, high quality lignite oil production, high value light oil, coal tar and gas products were determined at the goal of high fuel producing yield.

This study examined the high sulfur and ash types of Kütahya Gediz lignite, Soma lignite, Şırnak asphaltite and lignite by TGA analyzer as illustrated in Figure 2.

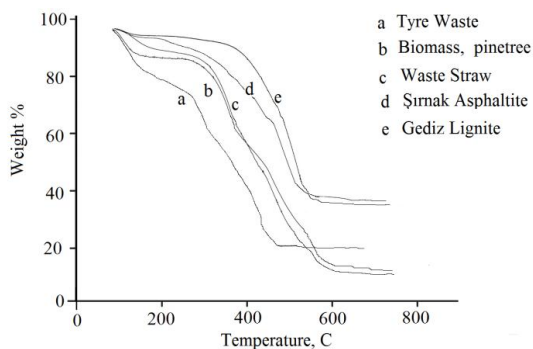


Figure. 2 Combustion of Coal gas and Waste Tyre oil and Biomass oil to black carbon

Proximate analysis of studied Turkish lignites and Şırnak asphaltite and waste wood and tyre are found as given in Table 1. Studied coals and biomass, tire wastes carried out on slow pyrolysis and pyrolysis oil subjected to black carbon production in retort, as shown in Figure 3.

The country needs the cleanest fuel to be produced providing the essential oils and gases. For this reason, gasification of Kütahya Gediz, Soma lignite, Şırnak asphaltite and lignite may be so feasible at the side of cost and production high amount of gaseous fuels instead of importing natural gas.

Table 1., Proximate Analysis of Turkish Lignite and Asphaltite. (ADB:Air dried base. DB:Dried base, DAB:Dried ashless base).

Coal Type	Ash, % ADB	Moisture, % ADB	TotalS,% DB	Volatile Matter, % DAB
Şırnak Asphaltite	6.3	0.1	5.7	52.6
Kütahya Gediz	26.0	11.7	4.6	42.7
Soma Lignite	13.8	14.0	2.2	40.4
Waste Wood	0.2	51.7	0.6	22.7
Waste Tyre	13.8	0.01	0.2	60.4

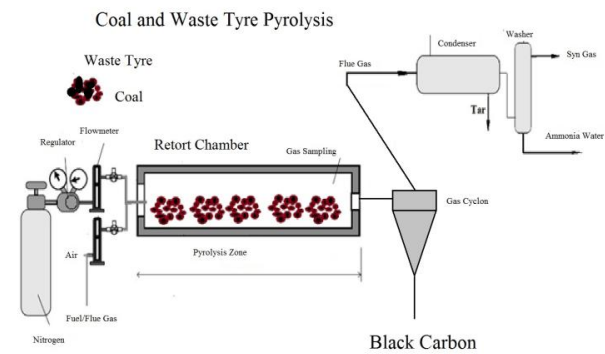


Figure 3 Pyrolysis of Coal and Waste Tyre to black carbon

This work is based on the assumption that the final process temperature is a decisive factor for the required volatile-matter content in the char being in a quasi equilibrium state with respect to the gas temperature. Instead of fluid bed combustion, packed bed gasification of coarse size coals is governed by chemical reactions on particle gas reactions. Combustion rates of Şırnak asphaltite were lower in comparison with biomass and lignite as seen in Figure 4 so that mass diffusion rates of gaseous materials to Şırnak asphaltite particle fundamentally control reaction kinetics.

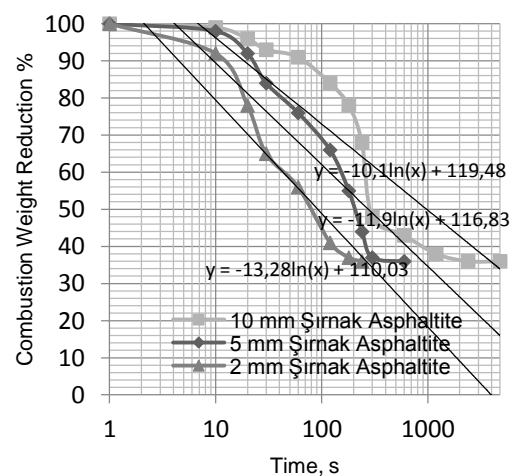


Figure 4, Combustion kinetics of Şırnak Asphaltite for black carbon production

A kiln reactor was used in coal pyrolysis heated till 600°C with a rate 7-10°C/min by fuel. The process was tested at a scale of 2–3 kg/h; collecting operational and design data to build an industrial installation. A technological diagram of the coal pyrolysis process developed unit is made.

Thermal destruction almost observed at temperature increase from 350 °C to 400°C with pyrolysis rate of 60-70% and with simultaneous dilution of oil products by condenser distillate. To achieve this, it is necessary to create conditions of internal circulation without the transported coal and char in rotary kiln, where the average concentration of solids amounts to 0.1- 0.2 kg/m<sup>3</sup>.

it is necessary to create gaseous conditions for black soot formation in retort without the transported coal and char back in retort, where the average concentration of solids amounts to 0.1- 0.2 kg/m<sup>3</sup> and the conditions for residence time are long enough for the combustion of coal and intensive gas mixing so enhancing mass and heat transfers.

Thermal pyrolysis commenced by oil and tar fuel burning into the retort firstly and then CO<sub>2</sub> gas evolution followed and circulated into the retort for three hours. When it is observed a temperature increase from 600 to 700°C without fuel addition, injected cooling water at a volume rate of 2/1 and air with 3lt/min. Gaseous products with simultaneous dilution of oil products by condenser distillate are collected. To achieve this, it is necessary to create conditions of internal circulation without the transported coal and char in retort; about 10-20% very low conversion yields to black carbon were observed at the end of pyrolysis.

### The Kinetics of Combustion to Soot

Reactivity can be determined from CO<sub>2</sub> conversion of solid carbon source in black carbon production medium for batch black carbon combustion. This approach is accurate, fast, and allows determination of time-course experimental parameters. In this case, loaded BC is centrifuged and transferred to batch cell. Controls of unloaded BC with and loaded BC without are run in parallel. When the CO<sub>2</sub> curves reach a stable level indicating that reactivity had ceased, the carbon consumed.

Oxygen Uptake also shows the kinetics of indirectly soot conversion and directly reaction rate of combustion and control, even cooling water addition and fuel gas inlet.

Quantification based on measurement collected the weights of black carbon and char products are determined at each combustion and pyrolysis tests.

Flue gas soot conversion by CO<sub>2</sub> medium and temperature control in fluidized chamber are carried out. Produced pyrolysis oils were combusted in the reactor as illustrated in Figure 5. in a batch black carbons. This approach is accurate, fast, and allows determination of time-course experimental parameters. In this case, loaded BC is centrifuged and transferred to batch cell. Controls of unloaded BC with and loaded BC without are run in parallel. When the CO<sub>2</sub> curves reach a stable level indicating that reactivity had ceased, the less carbon dioxide consumed.

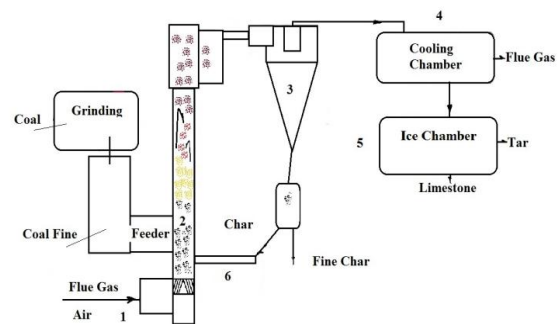


Figure 5, Combustion of pyrolysis oil of Coal, Waste Tyre oil, Biomass oil and flue gas to black carbon

Pyrolysis oil and tar of Kütahya Gediz lignite Şırnak asphaltite and waste tyre were combusted in flash chamber each separately and conversion to char determined as weight collected in the cyclone downstream. The results are illustrated in Figure 6.

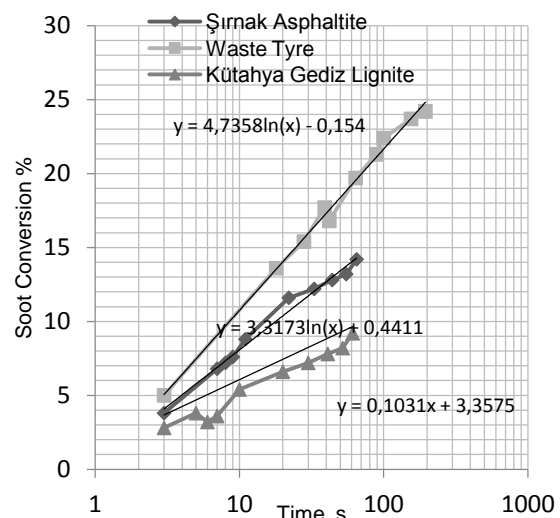


Figure 6, Pyrolysis soot conversion of Gediz lignite at equal weight rate separately in Pyrolysis Retort in comparison with Waste tyre

Oxygen consumption was the indication of soot conversion of oil combustion and change of oxygen uptake in the tests was shown in Figure 7. The results of conversion of soot and char with combustion of flue gas of pyrolysis and pyrolysis oil of coal, waste tyre and waste wood as shown in Figure 8.

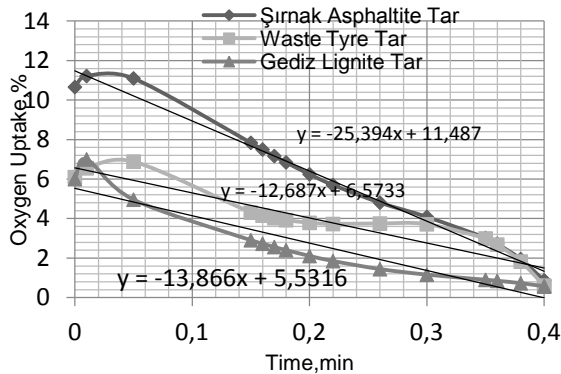


Figure 7, Combustion kinetics of Coal gas and Waste Tyre oil and Biomass oil to black carbon

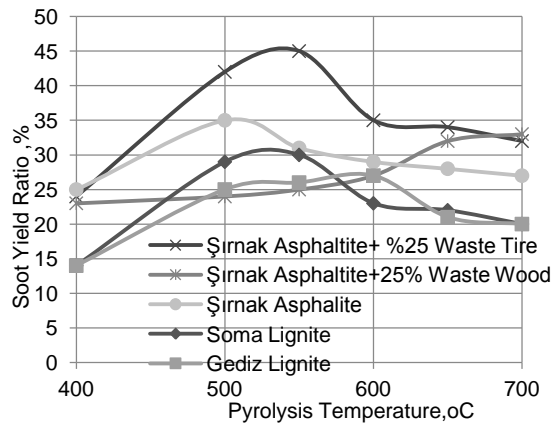


Figure 8, Combustion kinetics of Coal gas and Waste Tyre oil and Biomass oil to black carbon

### Quality of Black Carbon produced from Pyrolysis Oil

BET N<sub>2</sub> surface area analysis and pour density size analysis were determined and compared trade class black carbons in order to ascertain usage type. Based on measurement of black carbon quality over test products, ash content and sulfur contents were also determined and the results are given in Table 2.

Table 2, Black Carbon Qualities produced from Combustion of Pyrolysis Oil of Coal.

Coal Type	Trade BC	Aphaltite Tar BC	Soma Tar BC	Gediz Tar BC
N <sub>2</sub> Surface area m <sup>2</sup> /g	53	27	23	21
Ash content %	0,20	0,3	0,6	0,7
Sieve residue (100mesh)	0,01	0,01	0,04	0,04
Pour Density	340	320	380	340

## RESULTS AND DISCUSSION

In the pyrolysis experiments with addition waste tyre, the reactor temperature was changed between 400°C and 700°C and lignite samples were mixed with only by weight rate of 10% waste tyre and waste wood. Products received from pyrolysis and combustion of coal and waste mixtures were subjected to analysis for soot and char weight determination. Test results of pyrolysis by waste tyre and separately waste wood at 600 °C are seen in Figure 3.

From the point of view of pyrolysis and oil combustion experimentation, the resulted soot and chars quality and quantity in the pyrolysis chambers for biomass, lignite and coal-waste mixture samples were determined for different source evaluation and so we may reduce the effect of ash and sulfur content of coal samples in order to optimize pyrolysis and combustion rates of lignite samples. As given in Figure 5 gas and oil yields for lignite and coal samples were slightly similar, oil yield was lower for coal. In the pyrolysis experiments with different particle size fractions of coal specimens, at reactor temperature changed to 600°C and lignite samples mixed only by waste tyre at 10% weight rate. Products of pyrolysis of coal specimens were subjected to analysis for yield determination.

Test results of pyrolysis of Turkish lignite are seen in Figure 4. Comparison of particle size at 3-10mm additions at equal rates into the pyrolysis chamber with finer lignite showed that Gediz lignite was showed higher oil yield at near 26 % weight rate. In the gasification experiments, the experimental condition is calculated on the basis of the gas composition in the ambient state. So neither the contained water vapour nor the condensing hydrocarbons are taken into account. However, these components increased by decrease the particle size to 100 micron and oil yield was remained low 12% weight. Those values provided advantageous higher carbonaceous content to be converted to gas in gasification stage. Pyrolysis liquid and gaseous products of Şırnak asphaltite may equal to 5–20 g tar/m<sup>3</sup> and 5–10 g /m<sup>3</sup> of benzene, toluene, xylene in unit process gas. Moreover, Figure 5 showed that oil yields was slightly lower than coal char yields 27% weight. Gas yield was containing mainly steam and CO<sub>2</sub> in the pyrolizer and the amount of gas was remained between 37 and 40%. Combustion tests were carried out for pyrolysis oil and tar of Turkish lignite and Şırnak asphaltite and optimized gas inlet of 3lt/min.kg coal tar. Soot yields for Turkish lignite and Şırnak asphaltite across to temperature were

shown from Figure 8 and even other lignites showed similar trend, the higher char yields at lower combustion temperatures. It is illustrated that higher carbonaceous content to be converted to gas in gasification was managed over 650°C till 700°C. That conversion rate remained among 28-36%. Even it was observed that increased the gas's calorific value by up to approximately 3200 kJ/m<sup>3</sup> at 700°C for Şırnak asphaltite. Therefore it was supposed that porous coal layers, especially porous alkali and metal catalysts exhibit sufficient gas permeability at least for the gases of chemically inactive and sufficiently small in particle size.

## CONCLUSIONS

A modified model of kiln combustion for Turkish lignite and asphaltite used in coal pyrolysis-combustion process was found to be efficient as shown in Figure 3 and 4.

The calorific value of Şırnak asphaltite was significant for gasification. Furthermore, the results exhibited the higher combustion yields for soot in using CO<sub>2</sub> gas at the gas inlet 3lt/min.kg coal with the kiln apparatus.

Pyrolysis of different types of Turkish lignite was successfully processed in terms of desulfurization and even reduction of volatile matter. At higher rates of carbonization of different types of Turkish lignite could be obtained from the tests using low flow steam inlet at 500°C. It has been clearly determined that CO<sub>2</sub> and cooling water much beneficial in soot conversion of different types of Turkish lignite. Şırnak asphaltite should be cleaned and low ash content affected char quality and oil yield in pyrolysis.

Turkish lignite may be a regarded carbon source in the various parametric soot conversion systems. In order to receive clean soot clean liquid and gaseous products must be generated in low temperature pyrolysis. It is also advised that the high amount of formation of soot will be managed high pyrolysis temperatures over 700°C and extracts more environmental friendly gaseous products. Pyrolysis may be carried out for Turkish lignite and Şırnak asphaltite in finer - 100µm size distribution showed sufficient soot yields from Şırnak asphaltite between to 600-700 °C and even other lignite showed similar trend, the higher char yields of 42-51 % at lower pyrolysis temperatures

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