Effects of coal size, carbonization temperature and treatment time on the upgradation of coal properties

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Abstract

The Lignite coal reserves of Sindh province (Pakistan) which is usually a low-grade coal could be used in coal-fired power plants, brick kilns, steel mills and cement industries. Though, it needs upgradation of coal in order to increase the fixed carbon content and simultaneous reduction of impurities such as Sulfur content of coal. Therefore, an economic clean coal technology (i.e., carbonization) was tested to upgrade the indigenous coal quality. This study was conducted to investigate the possible outcomes of carbonization process of coal with variable parameters such as size of coal sample, carbonization temperature and time interval. Three different sizes of coal samples (i.e. -25 mm to +10 mm, -10 mm to +5 mm and -5 mm to +2 mm) were investigated at different carbonization temperatures (400 ºC, 600 ºC and 750 ºC), respectively. In order to optimize the results, all samples were remain under carbonization process for 2 h and 4 h respectively. Results suggests that size of coal sample, carbonization temperature and treatment time significantly affect the carbonization of coal. After carbonization, the calorific value improved from 11,703 to 13,432 Btu/lb, the volatile matter reduced from 60.06 to 1.19%, the fixed carbon increased from 35.13 to 94.54% and the sulfur content reduced from 6.34 to 5.56%. However, the ash content was increased from 8.23% to 14.24%, in the test no.06 where higher size, higher temperature and longer time interval were used. Since ash could be reused for many applications like in road construction or surface subsidence. This research delivered an understanding to the policy makers to use local coal reserves for energy sector including various industries.

Keywords: *Coal Carbonization; Proximate Analysis; Sulfur Analysis; Gross Calorific Value Analysis; Coal Quality Parameters*.

1. Introduction

The greater expenses of oil and gas for electricity generation have inclined us towards the alternative and economical source e.g. coal (186 billion tons) which has been estimated to fulfill energy demand for many decades in Pakistan [1]. Around 90% of global energy requirements are fulfilled by fossil fuels alone [2]. Today coal is among the major fossil fuel sources in the world. In the year 2011, the contribution of coal all over the world was 39 billion joules of the total 150 billion joules of energy demand [3-5]. The consumption of coal is expected to rise by 17.6% to meet the energy demand for almost 10 billion world population by 2040 [6, 7]. In Pakistan, the major resources of power generation are oil and gas with 47% and 34% share respectively followed by hydro/nuclear 13% and coal resources with 5% [8]. By the end of 2021, the coal-fired power plants of 4290 MW and 5201 MW are expected to be in a state to utilize indigenous and imported coal to produce economical electricity in Pakistan [9]. While, many research studies have been conducted to decrease the environmental effects of coal-fired power plants in Pakistan [10, 11].The usage of every single possible fuel reserve is the need of the hour, due to increasing population [12]. Nowadays, coal is a mineral resource of great importance [13]. Coal is considered as pillar of the Iron and steel industries. Conferring to the World Coal Association around 70% of global steel production depends on coal Low-quality coal for example lignite are rich resources worldwide but they have low calorific value and high moisture content. Carbonization could convert lignite into coke and tar [14]. Combustion of coal is an opposite function of its volatile matter [15] . The production of coke, tar and gas yields is directly proportional to the volatile matter of the coal [16, 17].
Devolatization of lignite coals can increase the burning efficiency and coal quality. Meanwhile, problem of particulate matter radiations can be controlled [1]. While burning of coal, volatile matter is the main cause of particulate matter

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radiations in the environment , it can be easily removed by carbonization process. Carbonization process of coal provides cleaner and environment friendly usage of coal [18]. Carbonization process provide enhanced results in the oxygen free environment. Oxidation process have adverse effects on coal carbonization [16]. Existing studies have defined the effect of volatile matter in different ranks of coals on emissions from ovens of various types [19]. In a new industrial society coking coal is vital, purchases are becoming progressively more expensive [20], therefore, it is time to upgrade the quality of non-coking coals to meet the energy requirements.

2. Objectives of the study

The purpose of this study was to upgrade the coal quality parameters utilizing different variation in carbonization process and to observe effects of size, temperature and time interval on carbonization of coal. This study has resemblance to the study conducted in year 2002, brown coal (lignite) was taken for laboratory scale carbonization in rotary kiln vessel tested at different temperatures, different sizes and different intervals of time [21]. The main equipment for carbonization process is combustion furnace and carbonization chamber [22, 23]. Following the Rohde et al and Zhaohang the carbonization chamber (Fig. 1) was prepared and used along with crucible furnace for carbonization tests.

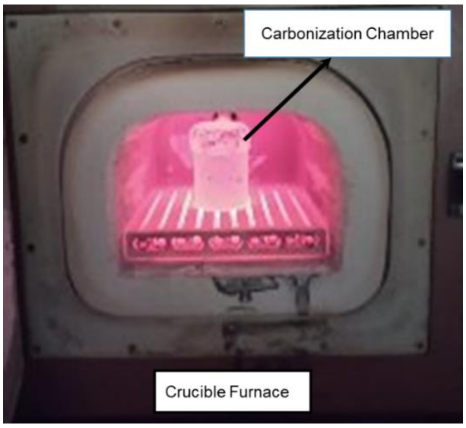

Fig. 1: Crucible Furnace and Carbonization Chamber

3. Materials and Methodology

3.1. Study Area

The Lakhra coal field was selected for this study. This field is located in the province of Sindh (Pakistan), having Latitude 25º42′ to 25º55′ (N-S) and longitude 68º0′ to 68º62′ (E-W). This field is considered as one among the largest coal producing field of Pakistan. The total coal resources of Lakhra are estimated as 1.328 billion tons with 146 million tons of mineable reserves [24]. The Lakhra coal mines have Lignite-A type of coal reserves. For this study the coal samples were taken from the mines of Indus Coal Company working in the Lakhra coal field area.

3.2. Sample Preparation

Polythene wrapped lumps of coal were carefully removed from the coating followed by the manual breaking of coal with the help of geological hammer. Samples were prepared in two phases, one for carbonization chamber and other for equipment of proximate, GCV and sulfur analysis. In first phase 18 raw coal samples were pasted in mortar and screened through -250µm sieve in order to get as-received analysis of the coal sample. In second phase 18 samples (6 samples of each size -25mm to +10mm, -10mm to +5mm and -5mm to +2mm) were prepared for carbonization chamber.

3.3. Carbonization Test Sequence

Carbonization test sequence was designed in a way that all sizes -25mm to +10mm, -10mm to +5mm and -5mm to +2mm were tested individually at three different temperatures 400°C, 600°C and 750°C for two time intervals 2h and 4h respectively in the carbonization chamber. The reason behind selecting two time intervals (i.e. 2h and 4h) was to observe that how effectively treatment time affects the coal quality parameters. The test sequence can be better understood by following Fig. 2.

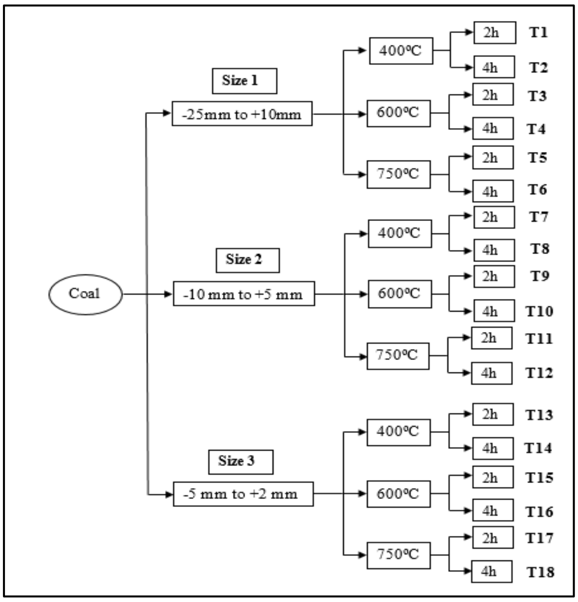

Fig. 2: Carbonization Test Sequence

4. Results And Discussion

4.1. Proximate, Sulphur and GCV Analysis

The results of proximate, GCV and sulphur analysis of raw coal samples and carbonization products were obtained through Thermogravimetric Analyzer (TGA-701), Calorific Value Analyzer (AC-500) and Sulfur Carbon Analyzer (SC-832) respectively. Total 36 number of samples were tested of which 18 were raw coal samples and 18 samples were their corresponding carbonization products as shown in Table 1.

Table 1: Proximate, Sulphur and Gross Calorific Value analysis of raw coal and carbonized product

S NO:	Volatile Matter (Dry Basis) (%)		Fixed Carbon (Dry Basis) (%)		Ash Content (Dry Basis) (%)		Sulfur Content (Dry Basis) (%)		Gross Calorific Value GCV (Dry Basis) (Btu/Lb)	
	Raw Coal	**Product**	**Raw Coal**	**Product**	**Raw Coal**	**Product**	**Raw Coal**	**Product**	**Raw Coal**	**Product**
1	56.01	28.69	30.99	64.99	11.37	13.07	7.78	12.05	10,304.40	13,077.17
2	63.26	24.04	31.77	71.60	9.01	13.02	7.09	10.66	11,575.08	13,434.33
3	56.32	8.52	30.94	81.12	12.92	19.95	8.67	8.02	10,375.43	12,451.40
4	60.93	5.51	31.64	88.65	9.94	17.21	8.58	9.06	11,194.14	13,224.76
5	55.16	2.23	29.96	81.36	13.88	26.12	9.22	8.14	10,015.47	11,460.29
6	60.06	1.19	35.13	94.54	8.23	14.24	6.34	5.56	11,703.72	13,432.52
7	55.10	27.62	33.48	66.24	10.77	13.32	8.89	11.90	10,633.93	13,114.80
8	50.98	24.93	30.22	69.31	10.03	14.27	12.26	12.85	9,317.93	13,205.72
9	55.28	6.70	32.44	84.41	10.96	17.25	8.06	7.80	10,462.57	12,654.23
10	61.68	5.34	32.00	89.56	9.74	16.29	7.70	8.50	11,380.93	13,336.81
11	54.59	1.41	28.94	87.47	17.19	18.74	9.15	5.85	9,791.28	12,260.11
12	60.32	0.40	34.86	94.79	8.47	15.36	7.44	5.61	11,698.67	13,355.85
13	56.53	26.77	29.48	67.08	11.76	13.14	8.22	11.61	10,099.42	13,106.52
14	60.47	24.15	33.05	69.97	10.21	15.21	8.23	13.89	11,401.81	13,206.90
15	53.63	6.44	31.60	81.44	11.88	21.00	8.15	9.89	10,051.51	12,142.68
16	62.05	5.38	34.08	89.75	5.76	14.56	7.63	8.16	11,769.92	13,335.40
17	55.38	2.33	32.72	82.10	12.54	21.73	8.54	7.31	10,564.67	11,502.54
18	59.24	0.48	34.85	93.32	9.45	16.85	7.23	6.92	11,542.05	13,134.11

In order to compare the results of raw coal samples and carbonized products, the average value of 18 raw coal samples was taken as standard raw coal analysis and was compared with each carbonization product separately. Table 2 shows average values of raw coal analysis represented as 18RC in following graphs.

Table 2: Average Raw Coal Analysis (18RC)

VM (%)	AC (%)	FC (%)	SC (%)	GCV (BTU/LB)
57.61	10.78	32.12	8.29	10,771.27

4.2 Parametric Variation after carbonization treatment

4.2.1 Volatile Matter Analysis (VMs)

The carbonization process significantly reduce the volatile matters (VMs) as illustrated in Fig. 3. The increase in temperature heated the coal samples during carbonization which instantaneously decreased the volatile matter due to evaporation of volatiles in the form of coke oven gases. Similar behavior for VMs of coal was observed in previous studies conducted by Najam ul Saqib et al. (2019) and Irwansyah et al. (2012) during hydrothermal carbonization (HTC) and Carbonization respectively [25, 26]. In this study it can be stated that at high temperatures volatile matters tend to vaporized and released into the environment, consistent to the study conducted by Putra et al. (2018) [27].

4.2.2 Fixed Carbon Analysis (FC)

The carbonization process enhanced the fixed carbon content of the coal. The carbonization time and temperature were found to be essential factors during carbonization process and indicated direct relation with fixed carbon content as shown in Fig. 4. As all the moisture and volatiles escaped off due to increase in temperature therefore remaining sample was found to be rich in fixed carbon content. In this study fixed carbon and volatile matters have shown inverse relation to each other which is consistent to the studies conducted by Kalderis et al. (2014) and Irwansyah et al. (2012) carbonization of coal [25, 28].

4.2.3 Gross Calorific Value (GCV) Analysis

The GCV of all the carbonization products was increased in comparison with raw coal sample. The carbonization time and temperature were found to be critical parameters in terms of the GCV see Fig. 5. The increase in fixed carbon content is considered as important quality parameter in the coal rank which gives higher calorific value during burning. A strong positive correlation was found between GCV and fixed carbon and negative correlation was found between GCV and VMs, which are consistent to the observations found in the studies conducted by Najam ul Saqib et al. (2019) and Irwansyah et al., 2012 [25, 26].

4.2.4 Sulfur Content Analysis (S)

The sulfur content of the coal was increased at lower temperature while decreased at higher temperature as shown in Fig. 6. The reduction of sulphur at high temperature is due to excessive devolatilization and dehydration of coal samples, these results are consistent to the studies conducted by Irwansyah et al. (2012) and Yuuki Mochizuki et al. (2013) [25, 29].

4.2.5 Ash Content Analysis (AC)

Fig. 7 illustrates that Ash content during carbonization is slightly increased in all products as compared to raw coal samples average value as shown in Fig. 7. Treatment time have played an important role in the ash content. Time interval 4h gave good results as compared to 2h. With increase in temperature ash content was also increased consistent to the study conducted by Irwansyah et al. (2012) [25].

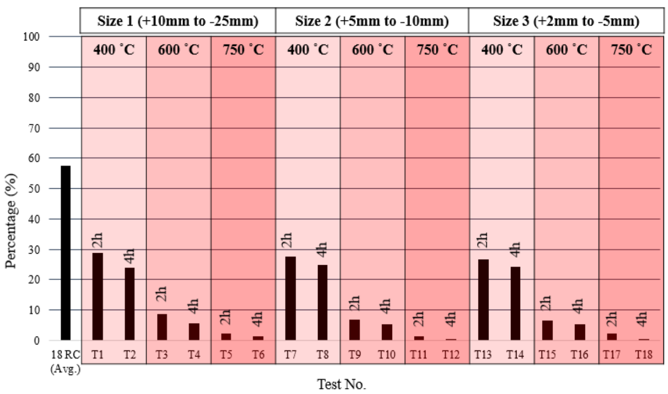

Fig. 3: Volatile Matter Analysis (VM)

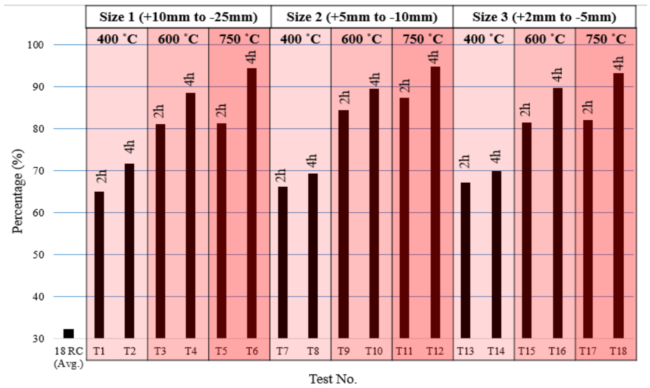

Fig. 4: Fixed Carbon Analysis (FC)

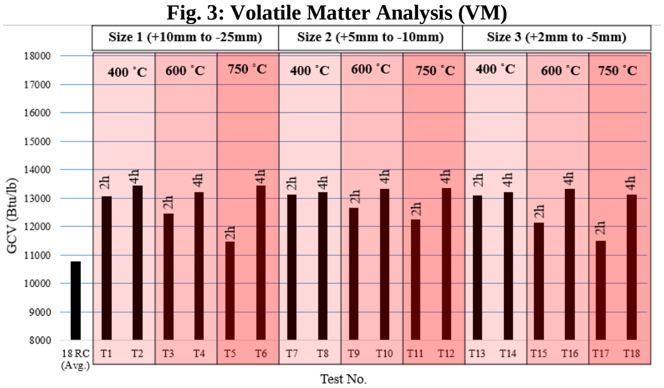

Fig. 5: Gross Calorific Value Analysis (GCV)

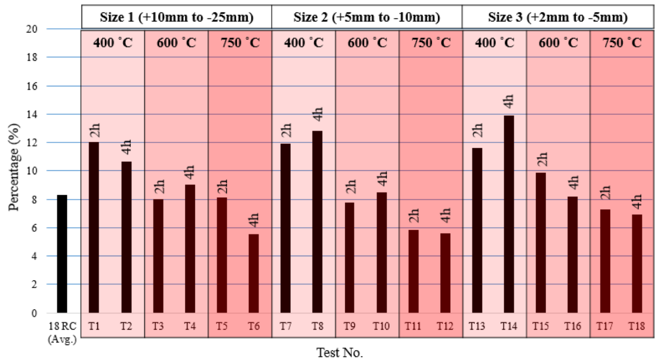

Fig. 6: Sulfur Content Analysis (S)

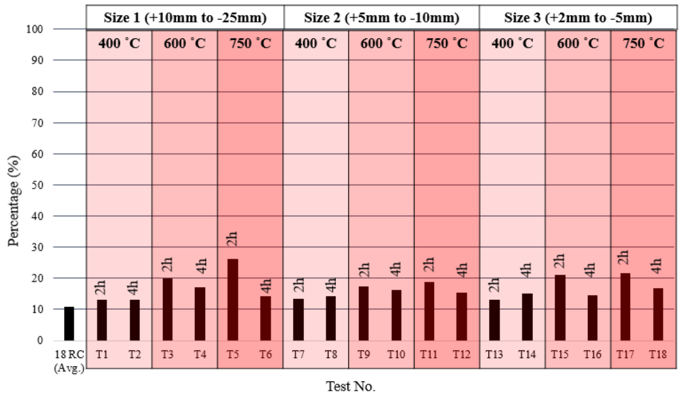

Fig. 7: Ash Content Analysis (AC)

4.3. Effect of Size on Carbonization of Coal

In order to understand the effect of coal size on carbonization process the average of all the tests conducted on each size was calculated and compared to each other. Results revealed that Size 2 (-10mm to +5mm) gave better results as compare to other two sizes. Therefore Size 2 (-10mm to +5mm) was considered as optimum size in this project.

4.4. Effect of Temperature on Carbonization of Coal

Temperature plays a vital role in the carbonization process. Based on temperature, carbonization is divided into two categories, one is Low Temperature Carbonization (LTC) which ranges from 500-700 ºC and other is High Temperature Carbonization (HTC) which ranges from 900-1200 ºC [30]. The variation of temperature in both types of carbonizations shows that the temperature has great significance in carbonization process. Average value of all the tests conducted on each temperature range was calculated in order to find out the optimum carbonization temperature. Results revealed that

Temperature 2 (600 ºC) gave the suitable results as compare to other two temperatures therefore in this study temperature range 600 ºC is the optimum carbonization temperature.

4.5. Effect of time interval on Carbonization of Coal

Time plays a significant role in every process. Therefore, a complete understanding of time dependent behavior of materials is very necessary to evaluate the merits and demerits of any project. In this study there were two time intervals 2h and 4h respectively. Each time interval has changed the key parameters of coal in carbonization process. Results were improved when time was increased from 2h to 4h. Therefore, in this study 4h time interval was considered as optimum time interval for this project.

5. Conclusions

In conclusion, it is stated that the carbonization of coal is always affected by size, temperature and time interval. Carbonization process with variable parameters played a vital role in the upgradation of coal. The results of the carbonization products were much better than raw coal sample analysis and could be compared with semi-coke properties stated by different researchers[31, 32]. The main objective of this project was to investigate the optimum coal size, carbonization temperature and treatment time for carbonization process of lignite coals. The results have shown that with the variation of coal size, carbonization temperature and time interval, the properties of carbonization product also vary. It was observed that coal size 2 (-10mm to +5mm) gave better results as compared to other two sizes (-25mm to +10mm and -5mm to +2mm) and was considered as optimum size. Variation in carbonization temperature varies the coal properties in carbonization process. The better results were obtained at 600ºC as compared to other two temperatures (400 ºC and 750 ºC) and was considered as optimum temperature. Two carbonization treatment times (2h and 4h) were utilized in this study. The increase in time improved coal quality parameters and it was concluded that 4h time interval was optimum as compared to 2h time interval. This study has provided an insight to policy makers that through carbonization process we can turn our huge lignite coal reserves into a valuable industrial product which could be used in many industries (e.g., coal fired power plants, steel mills and cement industries). The advantage of this project is that it is entirely indigenous and independent from any risk of foreign exchange variations.

Acknowledgment

Authors would like to acknowledge Department of Mining Engineering, Mehran University of Engineering and Technology, Jamshoro for coal laboratories having facilities to conduct this study.

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