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Upgradation of coal by froth flotation method; A case study of Akkakhel, Akhorwal and Shekhan coal mines

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Abstract

This study has been carried out on the upgradation of low-grade coals of Akkakhel, Akhorwal and Shekhan areas in western Khyber Pakhtunkhwa, Pakistan. The coals of these areas, because of high ash and sulfur contents and low Gross Calorific Values (GCV) are being mined on small scale for domestic purposes only. In this work, attempts have been made to reduce sulfur and ash contents from high ash and sulfur coal which will ultimately increase the fixed carbon and GCV of the studied coal from aforementioned coal mines by froth flotation technique. During this technique, the use of kerosene as a collector and pine oil as a frother at various particle sizes (i.e., -80, -100 and -140 mesh) has decreased ash and sulfur contents in the studied coal samples. The optimum process parameters were established at -140 mesh particle size with 20 % solids in floation pulp at pH 9. Under these conditions ash was reduced from 40.6 to 13.8% (66 % reduction) and sulfur from 3.97 to 1.51% (62 % reduction) while the GCV was enhanced from 7070 to 7963 kcal/kg in the coal samples of Akkakhel area. In the coal samples of Akhorwal area, ash was reduced from 35 to 18.16% (48.11% reduction) and sulfur from 6.31 to 3.51% (44.37% reduction) while the GCV was enhanced from 5500 to 6108 kcal/ kg. Similarly, in the coal samples of Shekhan area, ash was reduced from 41.6 to 20.1% (51.68% reduction) and sulfur from 3.1 to 1.5% (51.61% reduction) while the GCV was enhanced from 3995 to 4941 kcal/ kg. The final upgraded product was found suitable for use in power plants, boilers, and cement industries etc.

Keywords: Desulfurization, Ash removal, fixed carbon, GCV, Coal, Froth flotation.

1. Introduction

The present study has been carried out on the coals of Akkakhel in Bara, District Khyber, Akhorwal, District Kohat and Shekhan in Lower Orakzai District located in the western parts of Khyber Pakhtunkhwa, Pakistan (Fig. 1). The study area of Akkakhel Bara is situated between latitude 33o 50′ to 33o 53′ N and Longitude 71o 25′ to 71o 27′ E. The study area of Akhorwal is located between latitude 33o47 to 33o 50 N and Longitude 71o 20′ to 71o 24′ E while, the study area of Shekhan is located between latitude 33o 44′ to 33o 46′ N and Longitude 71o 13′ to 71o 17′ E. Geographically the above-mentioned study areas are bordered to the north by Peshawar to the east by Kurram Agency, to the west by Landi Kotal and to the south by Kohat and are easily accessible through Kohat pass and Darra Adam Khel road.

Various mineral phases are present in coal that can be removed from coal and their quality may be upgraded (Aksoy and Sagol, 2016). The abundant mineral phases in coal are sulfide minerals, which include pyrite (FeS2) and marcasite (FeS2) that cause boiler tube corrosion, fouling and pollution by emisssion of sulfurdioxide, when coal is burned (Xia et al., 2015) . Additionally, some other minerals i.e., carbonate minerals include calcite (CaCO3) and siderite (FeCO3) are also dominently present in coal (Saikia et al., 2014). Clay minerals are the most common occurring inorganic constituents of coal and generally count for 50 to 80% of the total mineral contents (Saho et al., 2017). In the present research work, froth flotation technique has been adopted to upgrade the coal from the studied areas. Froth flotation is a physico- chemical process which can separate mineral gangue from the desired coal and almost 60% of the worldwide coal has been upgraded through this process (Cebeci, 2002; Harbor, 2008). This process is based on the difference in surface hydrophobicity of different components and are used for the separation of a large range of sulfides, carbonates and oxides prior to further refinement. (Petokhov, 1995; Felici, 1996; Saleh, 2000). The advantages of froth flotation in coal processing have relatively low capital and space requirements, as well as the relatively high recovery achievable under a wide range of operating conditions (Demirdas, 2002; Erol, 2003).

As per U.S standards (ASTM D 388), the rank of coal is mostly calculated on mineral matter (ash) free basis, fixed carbon, calorific value and can be classified as lignite, sub bituminous, bituminous, semi anthracite and anthracite (Crozier and Klimpel, 1989). The coal fields of western Khyber Pakhtunkhwa are hosted by Early Paleocene Hangu Formation that extend up to Cherat, Khyber Agency, Kohat, Districts of Hangu and Tribal Orakzai Agency (Ahmad et al., 1986). The Coal seams are developed at the top and bottom contact of the Hangu and Lockhart Limestone (5-7m) in the Kohat Hangu area (Shah et al., 1993). These coal fields contain 44 million tons reserves of low quality (lignite to sub-bituminous) coal (Ahmad et al., 1986, Shah et al., 1993, Malkani, 2012). These coals as of having high sulfur and ash contents are not suitable for use in power plant, railway, cement, and coke making industry. Therefore, the upgradation of these coals is necessary to be utilized in a best way.

No detailed work has been done to assess the quality and upgradation of coal of Hangu Formation on the basis of detailed proximate analysis (Fixed carbon, moisture, volatile matter, ash and sulfur reduction and GCV. Therfore, froth floatation technique has been adopted in order to decrease the ash and sulfur contents and to enhance the fixed carbon and GCV in the studied coal at different particle sizes (i.e., -80, -100 and -140 mesh).

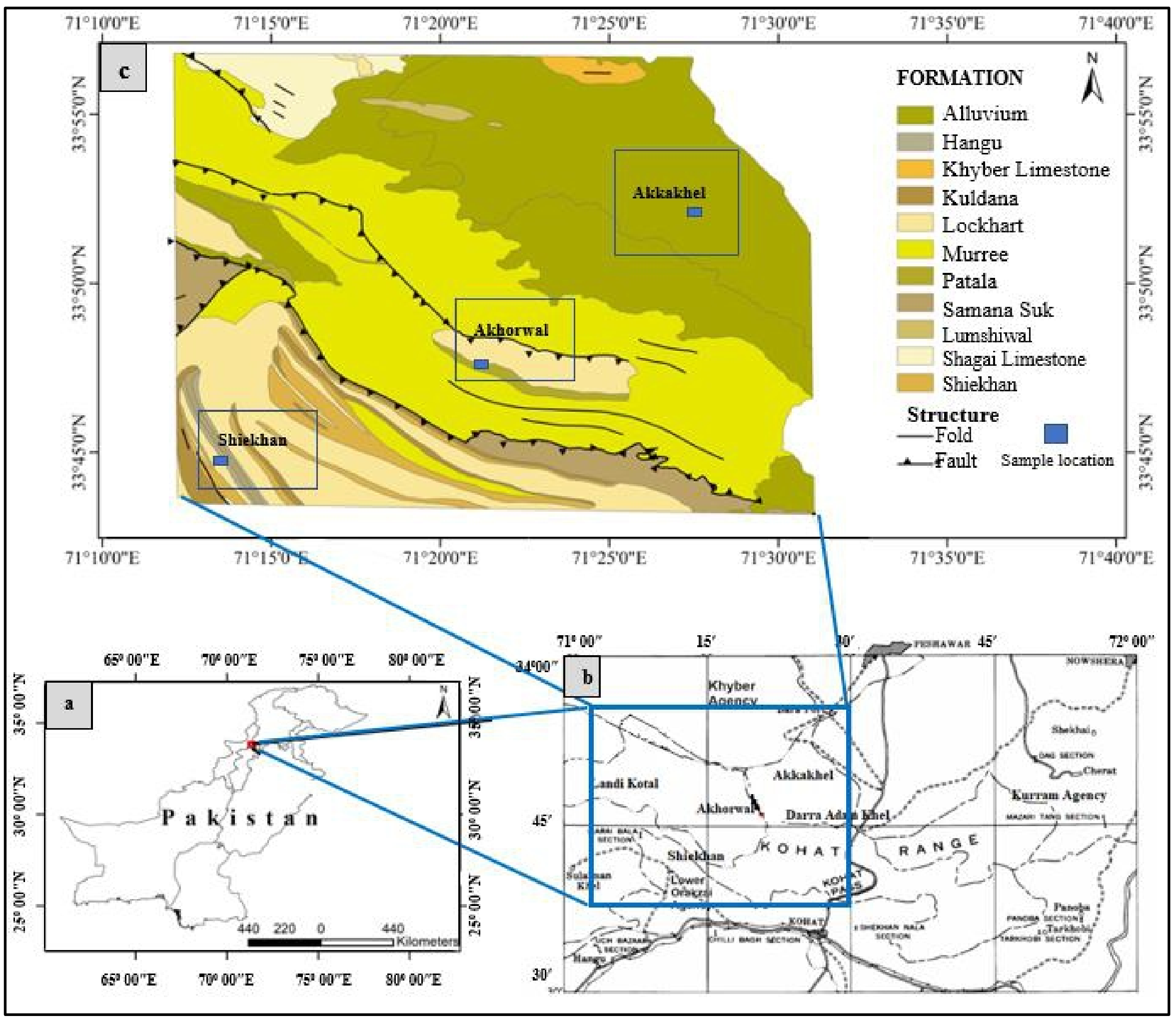

Fig. 1. (a, b, c). Geological map showing different lithological units and locations of the study areas (Rectangle) in Akkakhel, Akhorwal and Shekhan respectively (after Latif et al., 2002).

2. Methodology

During field work a total of about 5 kg each representative coal sample was arranged and packed in polythene bags by putting the specific mine name and locations . For the collection of samples the ASTM D 346-04 procedure was followed. Three samples were collected (one from each study area i.e. Akkakhel, Akhorwal and Shekhan) and were subjected to froth flotation procedure in the Chemical laboratory of "Centre for Coal Technology (CCT), University of the Punjab" for detailed proximate analysis. As per procedure, the lot of 0.5 kg was ground by the end runner mill to -80 (-180 microns), -100 (150 microns) and -140 mesh (106 microns) was used for detailed proximate (Fixed carbon, moisture, volatile matter, ash, sulphur and GCV) before and after froth flotation technique.

2.1 Upgradation of Coal using Froth Flotation Technique

Flotation treats a mixture of finely divided mineral solids, e.g., a ground ore, suspended in a liquid where a portion of such solids is separated from other finely divided solids, e.g., clays like materials present in the ore (Kawatra, 2001). In this process a gas or air is introduced in the liquid slurry that push a frothy mass containing economic mineral on the top of the liquid and leaving suspended waste material at the bottom of the agitating cell (Petukhov, 1995). When air is bubbled through the suspension that contains the coal particles, coal being hydrophobic will float on the surface and mineral matter will remain in the suspension.

2.2 Flotation equipment

Flotation can be performed in mechanically agitated cells or tanks or in tall flotaion columns and several other units including jamson cell. For froth flotation technique, computerized laboratory flotation cell was used (Fig. 2A). It has 1 to 2 litre tanks and having rotary mixer and diffuser (Fig. 2B). It has an air injector which injects air through rotary mixer (Fig. 2A). The digital screen controls the function of froth flotation cell. Flotation column use air spargers to introduce air at the bottom of column which provide mixing action into small bubbles. The mineral concentrated as froth is collected from the top of the cell while the gangue lift behind at the bottom of the tank.

2.3 Flotation experiment

Flotation experiments were carried out in a computerized laboratory flotation Jamson cell with a 2-litter tank using 20 % solids by weight (Fig. 3A). The reagents used were kerosene as collectors and pine oil as frothers as per ASTM D 3174-73 and D 3177 standard methods. To make the pulp of 20: 80 a total of 400 g of coal sample with 1600 ml of water was subjected to single stage direct flotation in a mechanical agitating cell. About 5 drops of kerosene oil were added as collecting reagent with the help of dropper. The collector adheres on hydrophobic areas of coal surface and increase the hydrophobic character of coal. Pulp was agitated for 5-10 minutes to condition the pulp. The three drops of pine oil were added to pulp as frothing reagent and again conditioned for 2-3 minutes. The air valve, mounted tangentially on the shaft of the impeller, was opened and because of air, bubbles were formed, stabilized with the action of pine oil frothed and picked up the coal particles that were obtained as froth on the surface of the pulp (Fig. 3B). The froth, rich in coal particles, was removed with the help of scrapper. The part of the mineral, which did not float, remained in the container, and was removed as tailings. Both concentrate and tailings were filtered, air dried and weighed (Figs. 3C,-D). Using calcium hydroxide as pH stabilizer (pH 9) flotation tests were performed with -80, -100 and -140 mesh particle sizes.

3. Results and Discussion

The coal has two major phases, organic phase containing volatile matter and fixed carbon, while the inorganic phase is composed of mineral matter and are separated from coal through froth flotation technique (Yorulmaz, 1983; Cavallaro et al., 1991).

Considering the previous research, the effect of particle size of the studied coal samples during froth flotation was also judged by dividing the studied coal samples into -80,

-100 and -140 mesh particle size. These coal samples of different size fractions collected from the Akkakhel, Akhorwal and Shekhan areas have been upgraded through froth flotation technique by adjusting the values of fixed carbon, volatile matter, moisture contents, ash, sulfur contents and GCV using various reagents such as kerosine oil as collector and pine oil as frother reagent at pH 9. During this technique, variable change in various parameters (i.e., fixed carbon, sulfur reduction, ash reduction and GCV has been noticed at the impellar speed of 1200 RPM for -80, -100 and -140 mesh particle sizes (Table 1).

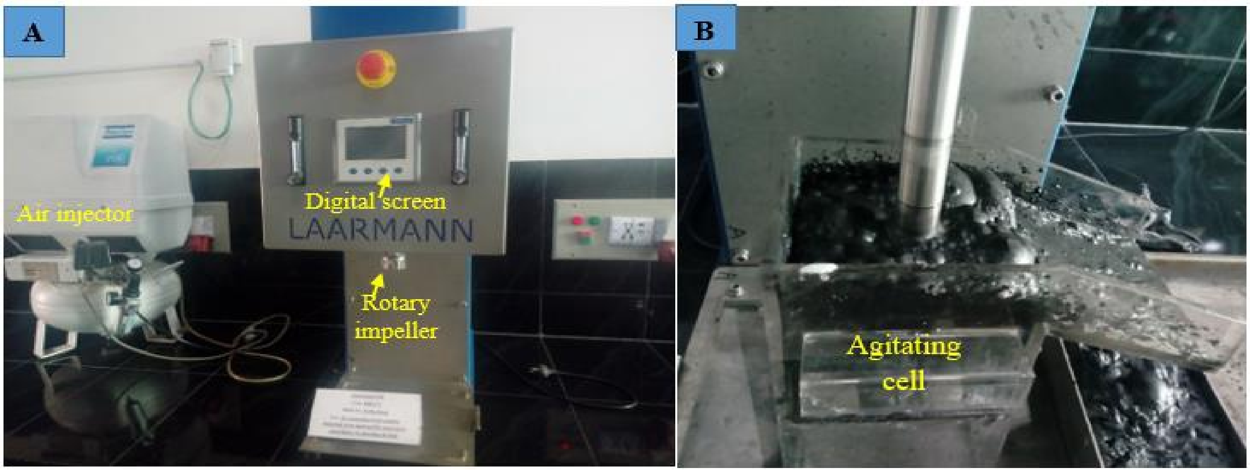

Fig. 2. (A) Showing the computerized laboratory flotation cell, (B) showing formation of froth/concentrate in a mechanically agitating cell.

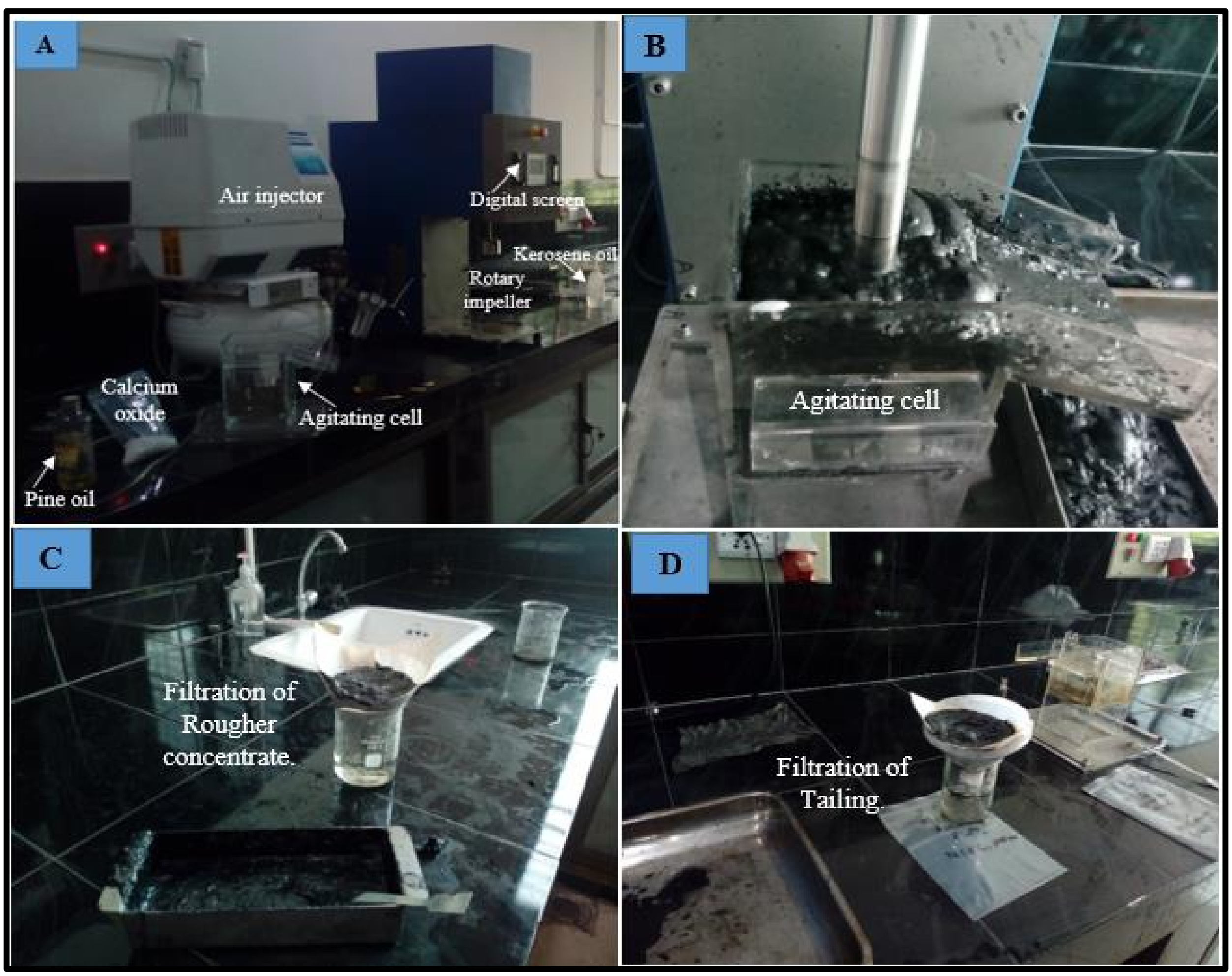

Fig. 3 (A) Showing Froth flotation Column, (B) showing collection of froth from the flotation column (Agitating cell), (C & D) showing filtration of concentrate and tailings.

O.S Origional Sample F.C Final concentrate

It has been found that maximum gaining of fixed carbon and GCV and maximum reduction in sulfur and ash contents took place at -140 mesh particle size after froth flotation (Table 2). It is discussed as follows: the value of fixed carbon increase from 55 to 80 % (44.4 % enhancement) in Akkahel area, 52 to 72 % (38.5% enhancement) in Akhorwal area, and 42 to 60.2 % (43.3% enhancement) in Shekhan area (Table 2). The maximum reductions in ash contents of coals in Akkakhel area were found as 66% with a reduction from 40.6 to 13.8%, 48.11% in Akhorwal area with a reduction from 35 to 18.16% and 51.68% in Shekhan area with a reduction from 41.16 to 20.6% (Table 2).

Similarly the reductions in sulfur contents of coal samples in Akkakel area were found as 61.96% with reduction from 3.97 to 1.51%, 44.37% in Akhorwal area with rduction from 6.31 to 3.51% and 51.61% in Shekhan area with reduction from 3.1 to 1.5%. (Table 2). The GCV increased from 7070 to 7963 kcal/kg in Akkakhel area, from 5500 to 6108 kcal/Kg in Akhorwal area and from 3995 to 4941 kcal/Kg in Shekhan area (Table 2).

It has also been noticed during this study that the particle size has greater effect in the upgradation of coal during froth flotation technique. It can, therefore, be investigated that during the froth floatation of the studied coal samples of different size fractions, the finer coal particles such as -140 mesh size as of more hydrophobic (dislike water) are attached to air bubbles and are hence carried to the surface of the pulp in relatively cleaner form. During this process, most of the impurities that are unusually associate with coal acted as hydrophilic (like water) and hence remained as suspended matters which have been discharged as dilute slurry waste. The better quality of -140 mesh size coal fraction as compared to that of -80 and 100 mesh size further suggest that the maximum impurities have been liberated from the finer fraction (i.e., -140 mesh) during froth flotation.

It has also been observed that with the increase in fixed carbon and decrease in ash and sulfur contents, there is an increase in the GCV of the studied coal samples (Table 2 & Fig. 4). The reason for higher GCV of the Akkakhel' s coal mine area as compared to that of Akhorwal and Shekhan coal mine is that, it contains higher fixed carbon contents (79.44 %) and lower contents of ash (13.8%) and volatile matter (6.06 %) as given in the Table 2. This is also consistant with the observation of Hower and Eble (2006).

3.1 Comparison of upgraded coal with Turkish and Chinese coal

Similar studies have been conducted by Ozturk and Temel (2013); Acikkar and Sivrikaya (2018) on malatya-arguvan lignite to sub-bituminous coal of Paleogene coal deposits in Turkey and by Xia and Peng (2015) on Paleocene sub-bituminous coal deposits in Ordos basin, western China, using the same experimental conditions by froth floatation processes. By comparing the increase in fixed carbon and GCV and the reductions in ash and sulfur contents of the studied coal samples with that of Turkish (Temel, 2013; Acikkar and Sivrikaya, 2018) and Chinese (Xia and Peng, 2015) coals, it was found that the fixed carbon values in the coals of Akkakhel area (53%), Akhorwal area (41%) and Shekhan area (43%) are less than those of Turkish (60 %) and Chinese (70%) coals (Table 3 & Figure 5).The reduction in ash contents in the coals of Akkakhel area (66%), Akhorwal area (43%) and Shekhan area (51.2%) is less than those of the Chinese (74%) coal while the coal of Akkakhel area has higher reduction and that of Akhorwal area has lesse reduction in ash contents as comapred to that of Turkish coal. However, the coal of Shekhan area has similar reduction in ash contents as compared to that of Turkish coal (Table 3 & Fig. 5). Similar reduction in sulfur contents was observed in the studied coals and those of Turkish and Chinese coals as was noticed in the case reduction in ash contents (Table 3).

The GCV found in Akkakhel coal is 7963 kcal/kg (upgraded to 13.27 % from origional value 7030 kcal/kg), in Akhorwal coal it is 6108 kcal/Kg (upgraded 11.05% from origional value 5500 kcal/kg) and in Shekhan coal it is upto 4941 kcal/Kg(upgraded to 23.67% from origional value 3995 kcal/kg) in the final concentrates (Table 3 & Figure 5). The GCV reported in the studied coals are lower than those reported by Acikkar and Sivrikaya (2018) in Turkish coal that is 8760 kcal/Kg (upgraded to 36 % from origional value 6440 kcal/kg) and Xia et al. (2015) in Chinese coal that is 9783 kcal/Kg (upgraded to 38.5 % from origional value 7062 kcal/kg) as given in the table 3. The studied areas have high moisture and volatile matter that's why its GCV is less than that of Chinese and Turkish coal (Table 2).

Table. 2. Froth flotation tests showing percentages of fixed carbon, moisture contents, volatile matter, ash reduction, GCV in the coal samples of Akkakhel, Akhorwal and Shekhan at-140 mesh (Negative sign shows reduction in contents).

Prameters	**Origional Sample (%)**	**Final concentrate (%)**	**Percent increase/dec-rease (%)**
At Akkakhel			
Fixed Carbon	55	79.44	44.4
Moisture contents	1.30	1.46	12.30 %
Volatile matter	6.13	6.06	- 1.14
Ash contents	40.6	13.8	- 66
Sulfur contents	3.97	1.57	- 60.45
GCV (Kcal/ kg)	7070	7963	12.63
At Akkhorwal			
Fixed Carbon	51.47	72.52	38.5
Moisture contents	2.53	2.87	13.4
Volatile matter	13.33	13.06	- 2.03
Ash contents	35	18.16	- 48.11
Sulfur contents	6.31	3.51	- 44.37
GCV (Kcal/ kg)	5500	6108	11.05
At Shekhan			
Fixed Carbon	42.5	60.2	43.3
Moisture contents	1.30	1.42	9.23
Volatile matter	16.2	14.2	- 12.34
Sulfur contents	3.1	1.5	- 51.61
Ash contents	41.6	20.1	- 51.68
GCV (kcal/ kg)	3995	4941	30.19

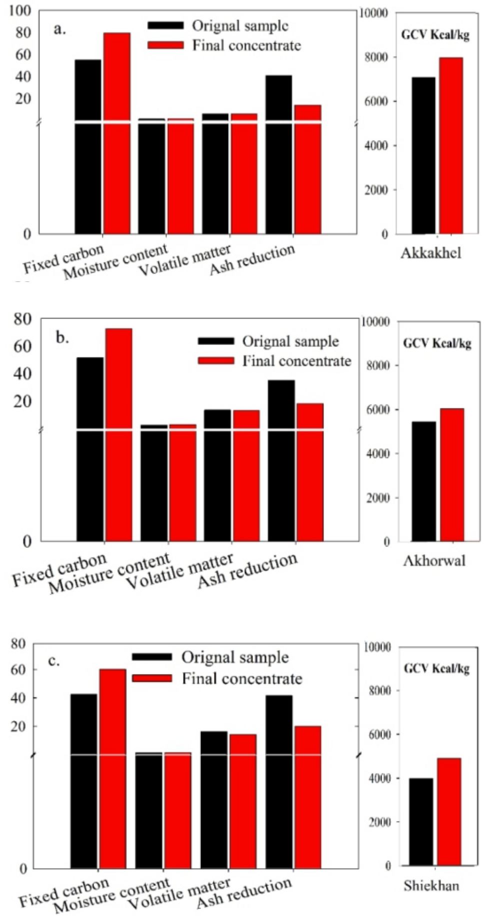

Fig. 4. Showing values of original samples and final concentrate of fixed carbon, moisture content, volatile matter, ash reduction and GCV of Akkakhel, Akhorwal and Shekhan coal mines at -140 mesh, respectively.

Table 3. Comparison of percent increase in fixed carbon and GCV and reduction of ash and sulfur contents in the final concentrates from Akkakhel, Akhorwal and Shekhan area with Turkish and Chinese coal at -140 mesh.

Location	Fixed carbon (%)	Ash reduction (%)	Sulfur reduction (%)	Percent increase in GCV		
				GCVs (kcal/kg)	GCVs (kcal/kg)	GCV (%)
Akkakhel	53	66	60	7030	7963	13.1
Akhorwal	41	43	44.37	5500	6108	14.69
Shekhan	43	51.21	51.61	3995	4941	23.61
Turkey	60	51	70.76	6440	8760	36
China	70	74	73.54	7062	9783	38.5

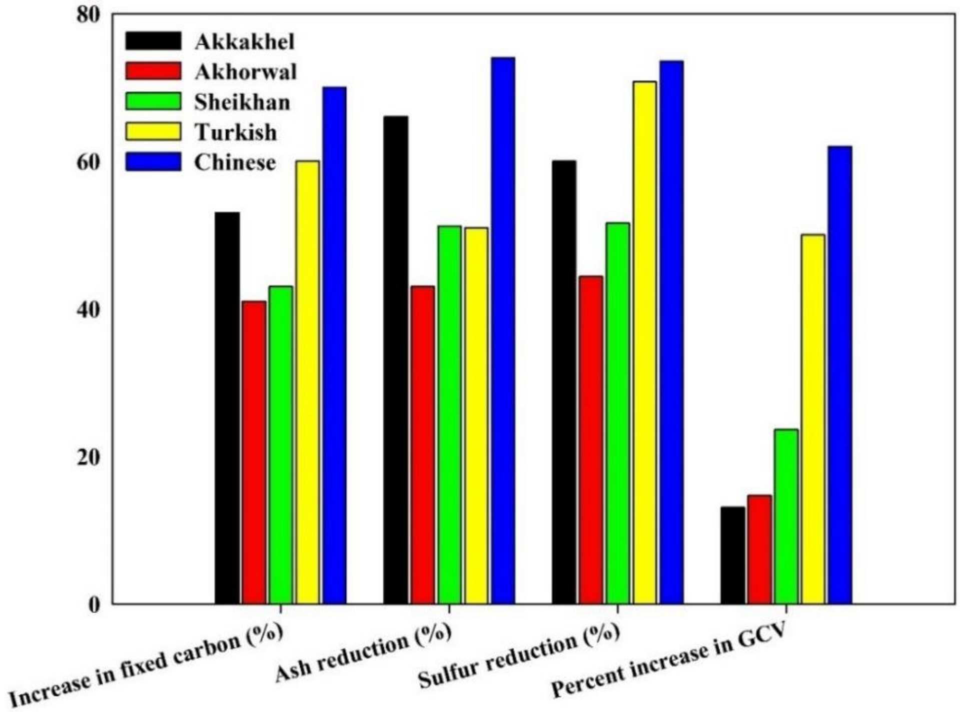

Fig. 5. Comparison of percent increase in fixed carbon, GCV and reduction of ash, Sulfur contents in the final concentrate of Akkakhel, Akhorwal and Shekhan areas with Turkish and Chinese coal at -140 mesh.

4. Conclusions

- The processing of coal samples, collected from Akkakhel, Akhorwal and Shekhan areas, through froth flotation technique results in the reduction of sulfur and ash contents and increase in fixed carbon.
- Maximum reduction of ash and sulfur contents and maximum increase in fixed carbon took place at -140 mesh particle size.
- Due to reduction of ash and sulfur contents and increase in fixed carbon, the GCV of the coal samples were enhanced in the coal of Akkakhel area, after froth flotation the upgraded GCV (7963 kcal/kg) value is higher than that of Akhorwal (6108 kcal/Kg) and Shekhan (4941 %) areas.
- By applying such technique to processing of coal will be helpful to utilize maximum amount of coal for energy purpose and will decrease the adverse effects on environment, animals and agriculture after utilizing the coal in industries.

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Author's contribution

The author "Mohibullah Khan" worked as a single author, conceived the study and were in charge of overall direction. The author contributed to sample preparation, developed the theoretical formalism, performed the analytic calculations and numerical simulations. The author designed and performed the experiments, derived the models, interpretation of the results and contributed to the final version of the manuscript.

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