Reduction of Sulfur and Ash from Tabas Coal by Froth Flotation

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ABSTRACT: *Tabas mines in Iran have coal sources which are suitable for use in metallurgy industries as coking coal. But the high sulfur content of this coal imposes severe limitations on its utilization as the sulfur oxide gases evolved from the combustion of high sulfur coals result in acid rains and corrosion of equipments. In this work, attempts have been made to reduce sulfur from high sulfur coal of Tabas by froth flotation. Laboratory tests were carried out in order to investigate the influence of various collectors, frothers, pyrite depressants and their consumption dosages on ash and sulfur reduction of Tabas coal. The use of kerosene as a collector and pine oil as a frother has decreased ash and sulfur content of coal more than other collectors and frothers. Although use of sodium polyacrylic acid as a pyrite depressant improved the total recovery of coal concentrate but did not enhance the reduction of sulfur.*

KEY WORDS: *Desulfurization, Ash removal, Coal, Froth flotation.*

INTRODUCTION

Existence of sulfur compounds in coal limits its industrial application due to environmental as well as technical problems. Sulfur is present in coal in three forms: pyrite, organic, and sulfate. The organic sulfur directly bound to the coal matrix is in the form of thiols, sulfides, disulfides, thiophenes, and cyclic sulfides. Pyritic sulfur (FeS_2) occurs in mineral phases as agglomerates of pyrite and marcasite crystals. The sulfate exists mostly as sulfates of iron and calcium. Silica in different forms such as quartz, crystobalite, etc., clay minerals such as kaolinite, illite, etc., carbonates such as calsite, dolomite, siderite, etc., sulfate and sulfides, etc., are the major ash-forming minerals in coal [1].

However, high sulfur coals can be upgraded by desulphurization through physical, chemical and bio-technological processes. One of the most suitable physico-chemical methods is froth flotation for removal of pyritic sulfur from coal [2]. Froth flotation is a fine particle separation process based on the difference in surface hydrophobicity of different components. Generally, froth flotation is the technique used for the beneficiation of coal particles below 0.5 mm in size. In this technique, separation of fine coals relies upon the wetting ability differences between the coal-rich and mineral-rich particles in an aqueous solution [3,4]. The basis of this process is the stable connection of air bubbles to the coal surface. The stability of this connection depends on a number of physical and chemical factors [5]. Flotation is

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often very effective for coal cleaning since coal is naturally hydrophobic and minerals are hydrophilic [6]. The advantages of froth flotation in coal processing are its relatively low capital and space requirements, as well as the relatively high recovery achievable under a wide range of operating conditions [7].

The conventional froth flotation method is applied to the coal for three aims: (1) to obtain a product with low sulfur and ash content by recovery of the coal in slime, (2) to diminish pollution of the environment by cleaning the process water, which is called black water and is removed from the coal preparations plant and (3) to produce coking coal by separating macerals from the coal [2].

Tabas coal is suitable for coke making, but it has high sulfur content for use in coke making industry. Total sulfur of Tabas coal is in the range of 1-6 % in the form of inorganic and organic compounds. Pyrite is the major inorganic sulfur compound in this coal.

The objective of this work was to study the possibility of cleaning this coal by the froth flotation method. For this purpose, laboratory tests were carried out in order to investigate the effect of various surfactant additions at different concentrations. The quality of the flotation products was evaluated in terms of ash and sulfur content percentage, in an attempt to understand the above effects.

MATERIALS AND METHODS

In the conventional flotation process, the pulp in a flotation cell is agitated to obtain particle suspension, and the mechanism of agitation permits entrainment of air into the cell. This air is dispersed as fine bubbles. During the flotation, the bubbles rise to the surface of the pulp, and a froth zone is taking place. In froth flotation, both water and solids are recovered into the concentrate from the level of the equilibrium froth height. Hydrophobic mineral particles are transferred up and out of cell, suspended in the water between bubbles.

Experiments have been done by Denver flotation device (Fig. 1). It has three cells (1.2, 5 and 10 lit.), a rotor with variable round (SALA Impeller) and is equipped by air valve.

A coal with less than 0.5 mm particle size from Tabas region in central east of Iran was used in experimental work. Ash and sulfur analysis was carried out according to ASTM D3174-73 and D3177 standard methods [8], respectively.

Fig. 1: Scheme of Denver flotation device.

Flotation experiments were made in a 1.2 liter Denver cell using 5 % solids by weight. The reagents used were kerosene and methanol as collectors and pine oil and MIBC as frothers. 60 g of coal sample was subjected to single stage direct flotation. The pulp was conditioned for 5 min. prior to any reagent addition. After adding reagents, it was conditioned for another 5 min. Then, the pulp was aerated at 1250 rpm and the froth was collected for 5 min. All experiments were conducted at natural pH around 8.0.

Both, floated (concentrate) and non-floated (tail) fractions were dried in an oven at 60 ^{o}C and then weighed to calculate flotation yield. Finally, ash and sulfur analysis carried out upon both concentrate and tail fractions.

RESULTS AND DISCUSSION

Effect of collectors on ash and sulfur recovery

Hydrocarbon oils and similar compounds have an affinity for hydrophobic surfaces such as coal surfaces. They selectively adsorb on the coal and increase its hydrophobicity. This improves the recovery of the coal and increases the selectivity between coal particles and mineral matter. In the coal flotation, non-polar oils such as kerosene, fuel oil and creosote have been used as collectors. These collectors are used to promote the rigid adhesion of air bubbles to the coal surface [5]. The flotation behavior of the coals using various collectors may change by many factors such as type and size of collector molecules, the type of bonding of collector, the structure of reagents, mixing ratio of reagents, collector electrical effect, and collector dispersion.

In this study, the improvement of the ash and sulfur recovery of Tabas coal was investigated by using kerosene and methanol as collector. The concentrations of collectors varied in the range of 125-500 g per ton of coal at low (5 %) pulp density.

Reduction of ash and sulfur and percentage of coal recovery for various kerosene and methanol concentrations are presented in Figs. 2 to 4.

Fig. 2 indicates that reductions of sulfur percentage decrease with increasing of kerosene concentration, whereas, with increasing of methanol concentration, the sulfur reductions increased, passed through a maximum of about 32 % at 250 g/t coal and then decreased.

The surface of unoxidized coal is naturally hydrophobic because coal is mainly composed of nonpolar hydrocarbons. Since water is a polar liquid, it has little tendency to wet nonpolar materials such as oil. Oils, on the other hand, have a strong affinity for nonpolar surfaces and wet them easily. The various ash minerals, such as silicates and clays, are composed of strongly polar compounds, therefore water wets them but oils do not. This basic difference in structure is then responsible for the ease of cleaning coal by froth flotation. So, it is clear that both collector and water are participated in froth flotation.

In general for both collectors, a reduction in sulfur is observed by increasing the collector concentration. At higher concentration of collectors, it seems that the collector is more attached to the pyrite surface than to the coal affected on the hydrophobia of pyrite. Therefore pyrite with coal particles floated up to the froth.

Fig. 3 shows that ash reduction is increased with concentration of both kerosene and methanol. *Yakup Cebeci* investigated the floatability improvement of Yozgat Ayridam lignite using various collectors and pointed out that ash reduces with increasing of kerosene concentration [5]. The presence of kerosene as well as methanol collector increased flotation yield resulted in high recoveries of coal and ash of the concentrates significantly by floating ash-forming materials. More than 50 % of the ash content of Tabas coal can be reduced using methanol collector at concentration of 500 g/t of coal. This indicates that methanol is an effective collector in reducing mineral compounds of the coal.

Fig. 4 shows that more recovery percentage using kerosene as collector is obtained compared with using

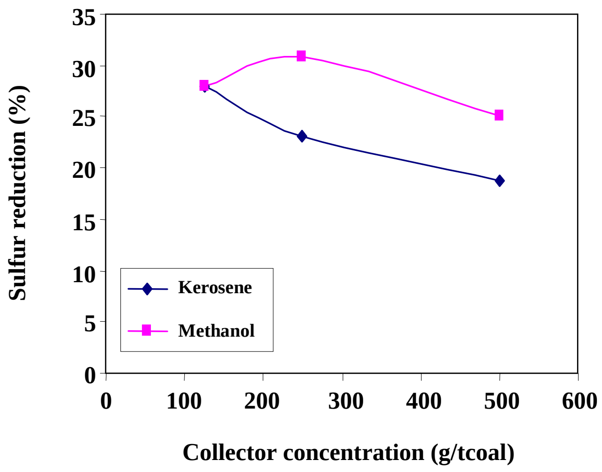

Fig. 2: Effect of collector concentration on sulfur reduction.

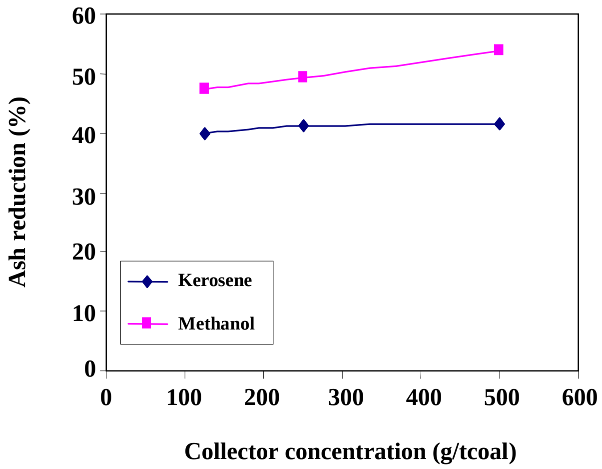

Fig. 3: Effect of collector concentration on ash reduction.

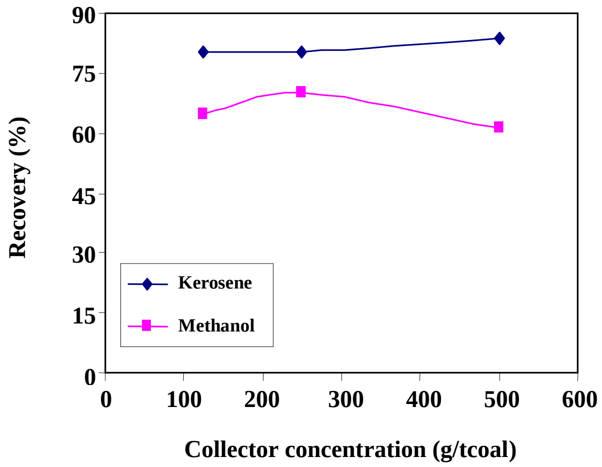

Fig. 4: Effect of collector concentration on coal recovery

methanol at the whole range of the concentrations used.

This figure indicates that kerosene has more desirable hydrophilic effect to the coal surface than Methanol.

Effect of frothers on ash and sulfur recovery

Frothers act to stabilize the air bubbles so that they will remain well-dispersed in the slurry, and will form a stable froth larger than can be removed before the bubbles burst. The most commonly used frothers are alcohols, particularly MIBC (methyl isobutyl carbinol) or any of a number of polyglycols [9].

In order to increase the flotation yield, 50-200 g/t of either pine oil or MIBC was used in this work as frother with kerosene collector that was kept constant at 125 g/t.

The behavior of pine oil and MIBC as frother at various concentrations on reduction of sulfur content and ash are shown in Figs. 5 and 6 respectively.

The performance of these frothers is compared in terms of coal recovery percentage in Fig. 7. Both frothers increased flotation yield and recovery percentage, but pine oil is more effective than MIBC. At very low frother dosage, the strength of the air bubbles was so weak due to the insufficient frother that coal particles could not be carried to the froth phase and resulted in lower yield and recovery.

Figs. 5 and 6 shows that the reduction of sulfur and ash decrease with increasing the concentration of frother for both pine oil and MIBC. The ash reductions decrease uniformly with the increase in frother concentrations. In study of *Tao* and *Li* [6] on Kentucky coal, low yield (5-15 %) and low combustible recovery (6-23 %) and high product ash (about 22 %) were obtained when MIBC was used as frother and fuel oil as collector. *Kimpel* [10] found that the use of different frothers produced changes in the flotation rate and recovery values in coal flotation and pointed out that regardless of frother type, increasing the frother dosage to increase recovery always leads to less selective flotation.

Also *Errol et al.* studied the effect of various reagents on flotation of Turkish coal and concluded that the ash rejections decrease uniformly with the increase in initial MIBC loadings and when the initial concentration of MIBC is increased, the combustible solid recoveries increase as well [7].

Since pine oil as frother also has collectoric property, it may have negative effect on pyrite surface at higher

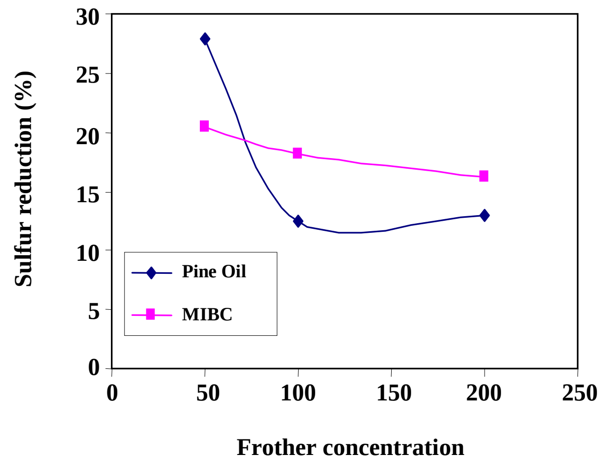

Fig. 5: Effect of frother concentration on sulfur reduction.

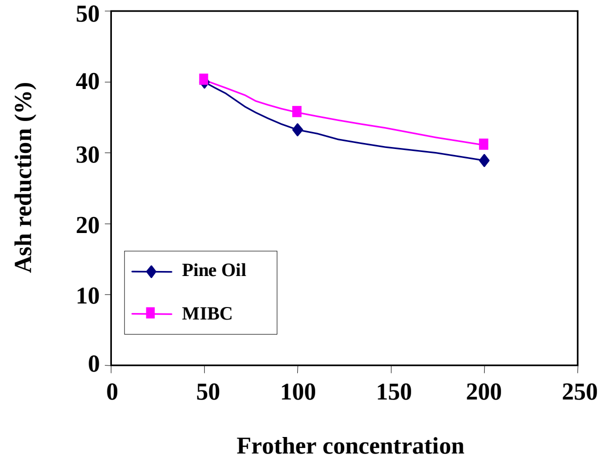

Fig. 6: Effect of frother concentration on ash reduction.

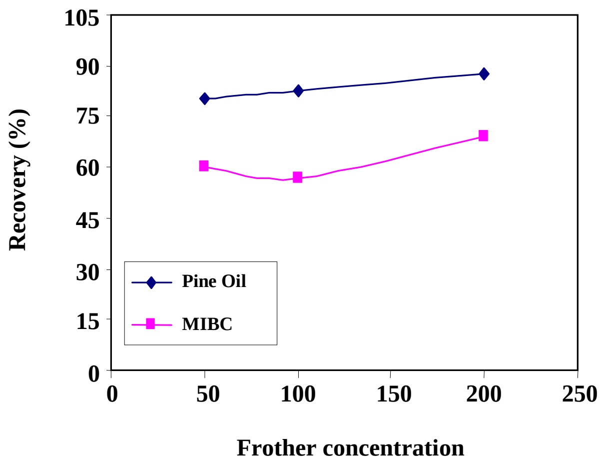

Fig. 7: Effect of frother concentration on coal recovery.

concentrations, preventing a good separation between hydrophilic pyrite and hydrophobic coal. Therefore the lowest recommended dosage of frother (50 g/t coal) is used in the next experiments of this work.

Effect of pyrite depressants on ash and sulfur recovery

Pyrite has been found to interact with the compounds that are commonly used as coal collectors [11], which could conceivably result in the pyrite being recovered along with the coal. Based on this assumption, many investigators have developed depressants that are supposed to prevent pyrite flotation. None of these depressants have ever been successfully used on an industrial scale [12].

The separation of coal from pyrite is enhanced by the use of an effective amount of a polymeric acid or salt therefore as a pyrite depressant in conventional flotation process [13].

Polyacrylamide and sodium polyacrylic acid were used as pyrite depressants, with an average molecular weight of about 9000. The kerosene is added in an amount equivalent to 125 g of collector per ton of raw coal feed, pine oil is added next in an amount equivalent to 50 g/t and slurry is conditioned for 5 minutes at 1250 rpm.

In Figs. 8 to 10, the ash and sulfur reduction and recovery percentage as a function of depressant concentration are presented. Almost no improvement effect is observed in sulfur and ash reduction of Tabas coal by adding the above depressants. The reason for this effect is that most of the time, the pyrite in coal will not be hydrophobic in the first place and adding pyrite depressants will have no effect. While depressants can conceivably help when the conditions are right for pyrite flotation, adding them is normally only a precautionary measure. Since adding depressants can more than double the total reagent consumption of coal flotation, the occasional possible benefits are not enough to justify the continuous cost.

Polyacrylamid depresses coal instead pyrite and decrease recovery percentage, severely. So this depressant is not useful. Sodium Polyacryl acid depresses pyrite slightly and separates it from coal but its sulfur reduction is not higher than without it. Altogether use of pyrite depressants does not have suitable effect on separation of pyrite from coal.

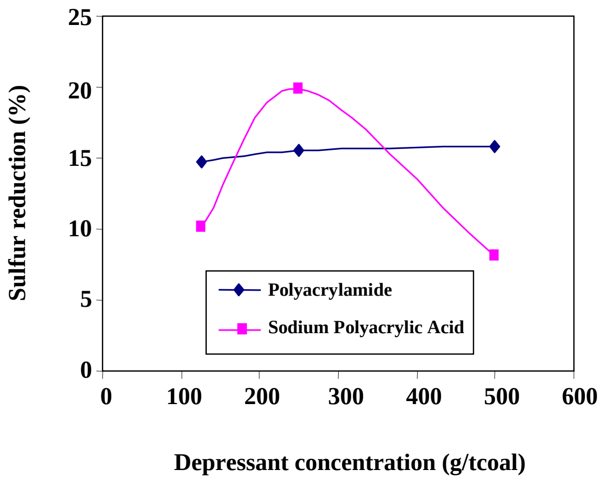

Fig. 8: Effect of depressant concentration on sulfur reduction.

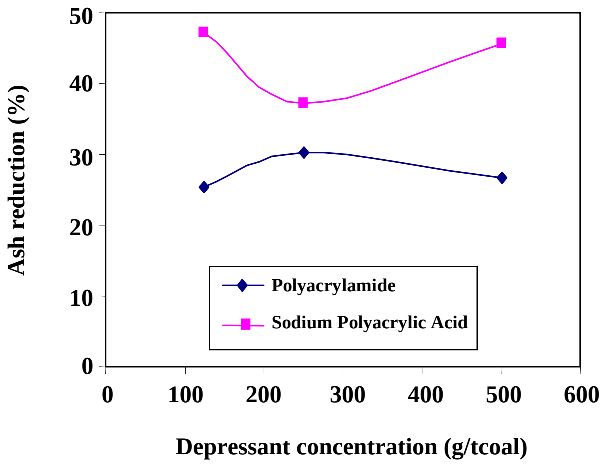

Fig. 9: Effect of depressant concentration on ash reduction.

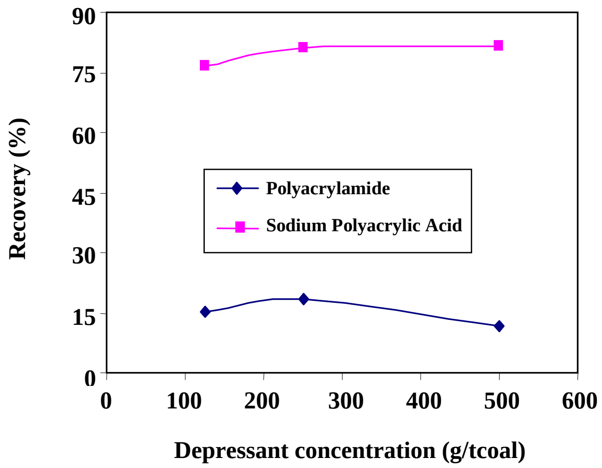

Fig. 10: Effect of depressant concentration on coal recovery.

CONCLUSIONS

Studies on the effect of reagent dosage on the floatability of pyrite during Tabas coal flotation indicated that, as the amount of frother and/or collector is increased, recovery of both coal and pyrite in the concentrates increased while no effect is observed when the pyrite depressants are used.

The use of kerosene and methanol as collectors decrease ash and sulfur content of coal about 40-50 % and 30 % respectively, but kerosene in 125 g/t dosage consumption has more recovery percentage (about 80 %) than methanol.

Both pine oil and MIBC frothers increase recovery yield but pine oil has better effect in 50 g/t coal in decreasing ash and sulfur content in coal concentrate.

The use of sodium polyacrylic acid as a pyrite depressants improved the total recovery of coal concentrate but did not enhance the reduction of sulfur.

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