

Reprinted from

INTERNATIONAL JOURNAL OF MINERAL PROCESSING

Int. J. Miner. Process. 58 (2000) 85–97

The effect of environment, oxidation and dissolved metal species on the chemistry of coal flotation

P. Somasundaran ^a, Lei Zhang ^a, D.W. Fuerstenau ^{b,*}

^a *NSF IUCS Center for Surfactants, Henry Krumb School of Mines, Columbia University, New York, NY 10027, USA*

^b *Department of Material Science and Mineral Engineering, Evans Hall, University of California, Berkeley, CA 94720-1760, USA*

Received 4 December 1998; received in revised form 18 April 1999; accepted 27 May 1999



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INTERNATIONAL JOURNAL OF MINERAL PROCESSING

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International Journal of Mineral Processing (ISSN 0301-7516). For 2000 volumes 58–60 are scheduled for publication. Subscription prices are available upon request from the Publisher or from the Regional Sales Office nearest you or from this journal's website (<http://www.elsevier.nl/locate/ijminpro>). Further information is available on this journal and other Elsevier Science products through Elsevier's website: (<http://www.elsevier.nl>). Subscriptions are accepted on a prepaid basis only and are entered on a calendar year basis. Issues are sent by standard mail (surface within Europe, air delivery outside Europe). Priority rates are available upon request. Claims for missing issues should be made within six months of the date of dispatch.

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variety of organic and inorganic constituents in different forms. Its surface properties, such as wettability and floatability, can vary to a larger extent due to its complex nature, as well as alterations in-situ and through subsequent exposure to various environments. In this paper, the effects of oxidation, pretreatment and dissolved species on coal flotation are discussed along with mechanisms involved.

2. Coal oxidation

The atmospheric oxidation of coal by weathering or aging is a natural process that involves physical and chemical adsorption of oxygen on the surface of coal and the formation of various acidic groups, peroxides and phenols. The oxidation of coal starts with the physical adsorption of oxygen on the surface to form a complex, and then chemical adsorption to form peroxides. As oxidation proceeds, these organic materials lead to the formation of various humic acids and degradation of the humic acids into soluble acids. Oxidation of pyrite also leads to the generation of various soluble inorganics that can adsorb on the coal surface and modify its wettability and floatability (Berkowitz, 1989).

The effects of the oxygen containing groups such as phenolic and carboxyl groups on the floatability of the coal are well-documented (Fuerstenau et al., 1983). For example, the floatability of a group of western coals has been reported to decrease with the increase in percentages of either the phenolic or the carboxyl oxygen (Fig. 1). These functional groups were found to not only control the wettability through the hydrophobic/hydrophilic balance, but also control flotation kinetics by their influence on the surface charge of coal particles.

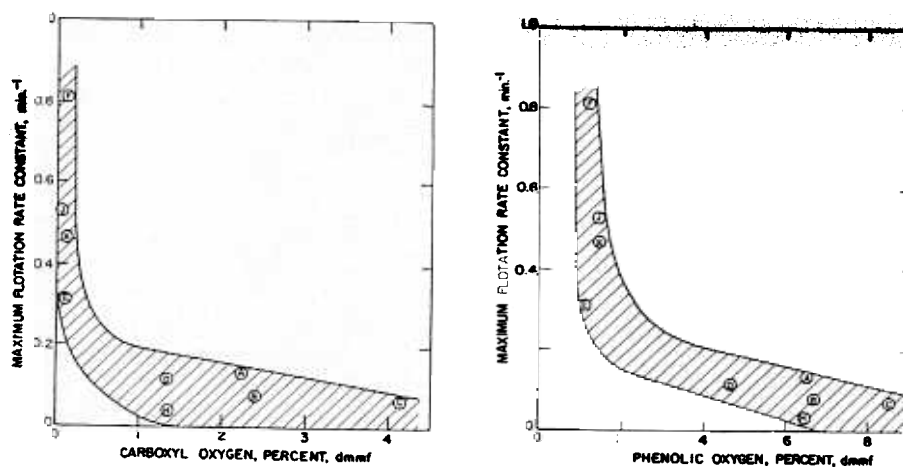


Fig. 1. Maximum flotation rate constant vs. phenolic and carboxyl oxygen contents (on a dry miner basis) for western coal (after Fuerstenau et al., 1983).



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^b *Department of Material Science and Mineral Engineering, Evans Hall, University of California, Berkeley, CA 94720-1760, USA*

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Abstract

The wettability and hence the floatability of coal are influenced markedly by its oxidation as well as the presence of dissolved inorganics in the system. Oxidation results in the formation of oxygenated surface sites that reduce hydrophobicity. Hydrolyzable metal species, whether released from coal itself during processing or added in the process, can drastically affect the flotation process. These phenomena are reviewed and the mechanisms involved are discussed in this paper. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: coal oxidation; coal hydrophobicity; coal flotation; coal weathering; surface functional groups

1. Introduction

Cleaning of coal has gained much more importance recently as a result of the increasing environmental concerns and the resultant requirements to provide cleaner coal for subsequent processing. Coal cleaning processes have resulted in the generation of more and more finer particles as the removal of pyrite and mineral matter requires finer grinding. Also, the use of mechanized and continuous mining techniques creates more fines than ever before. Efficient flotation, which is based on the differences in the surface chemical characteristics of coal and mineral particles, has become very important as a result of this trend. Coal is a complex heterogeneous material composed of a

* Corresponding author. Tel.: +1-510-642-3826; Fax: +1-510-643-5792; E-mail: wfuerst@socrates.berkeley.edu

Ramesh and Somasundaran (1989) proposed that the surface of coal consists of polar groups formed by the rupture of cyclic rings during oxidation, and that the density of polar groups on the surface is responsible for the observed decrease in hydrophobicity. Subsequently, Xiao et al. (1990) showed with diffuse reflectance infrared Fourier transform (DRIFT) spectroscopy that the loss of hydrophobicity upon oxidation by heating is due to the disappearance of the aliphatic CH groups by oxidation to carbonyl groups. The aromatic CH and C=C groups, on the contrary, were not susceptible to oxidation, at least at the test temperature used. Diao and Fuerstenau (1992) showed that anthracite did not oxidize when subjected to the same conditions that oxidize bituminous coals.

A number of investigators have attempted to simulate the oxidation process by gaseous and aqueous chemical treatments. Sun (1954) used heating to simulate the oxidation process and observed that floatability gradually decreased as oxidation became extensive (Fig. 2). Wen (1977) reported that coal becomes more and more negatively charged and the contact angle decreases with increasing oxidation.

To isolate the effect of oxidation from other processes such as loss of moisture or volatile matter, Somasundaran et al. (1991) conducted experiments in the presence of nitrogen as a control inert gas. From the results given in Fig. 3, it can be seen that with nitrogen, a slight increase in flotation is observed due to the removal of moisture, which renders the coal surface more hydrophobic. While the exposure to air at room temperature has very small effect, an increase in the floatability was observed at 90°C, probably due to the release of gases from the interior of the coal and increase in the hydrophobic-

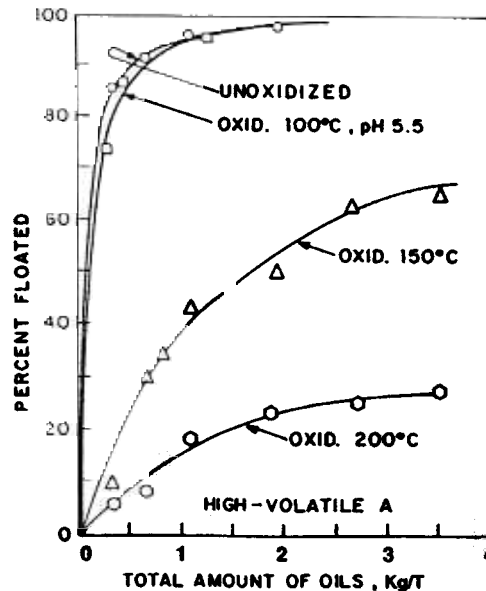


Fig. 2. Effect of air oxidation on the flotation of minus 65 mesh (minus 0.212 mm) high-volatile A coal using purified light oil (after Sun, 1954).

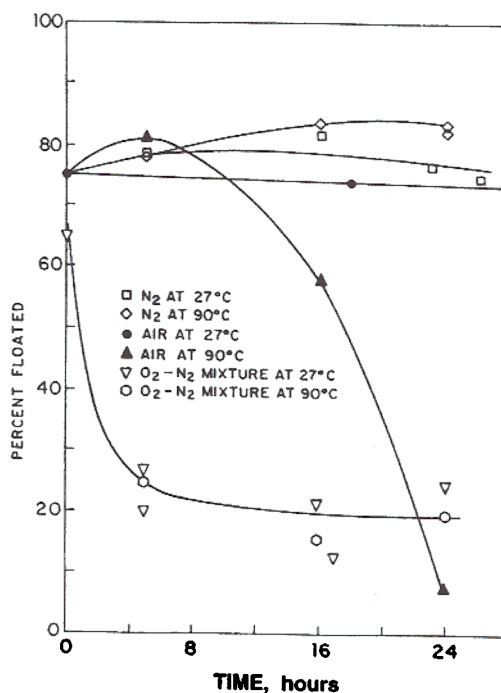


Fig. 3. Flotation of 65 × 100 mesh (0.212 × 0.150 mm) high-volatile bituminous coal treated with various gas mixtures (21% oxygen, 79% nitrogen) at 27°C and 90°C (after Somasundaran et al., 1991).

ity. As the oxidation proceeds, the effect of oxidation becomes dominant. Interestingly, an oxygen–nitrogen mixture (21% oxygen, 79% nitrogen) at a ratio close to that of air causes a drastic decrease in flotation from the beginning itself, probably due to enhanced oxidation as a result of the absence of moisture in the N₂–O₂ mixture. Diao and Fuerstenau (1992) found one hydrophilic western bituminous coal that also exhibits an increase in hydrophobicity upon heating under oxidizing conditions.

Diao and Fuerstenau (1992) conducted a detailed study of the oxidation of a variety of coals under controlled conditions. Fig. 4 presents film flotation plots for 100 × 150 mesh (0.105 × 0.158 mm) particles of a New Zealand bituminous coal of extremely low ash (0.78% dry basis) that was oxidized for 19 h at different temperatures in air. In this paper, only the results for 19 h of oxidation will be presented. With increasing oxidation, particularly above 100°C, film flotation results clearly show that the coal becomes hydrophilic. Using a standard barium hydroxide/sodium acetate titration procedure, they determined the carboxylic and phenolic functional group content of the coal under different oxidation conditions, and the results for 19 h of oxidation are presented in Fig. 5. Phenolic groups begin to form appreciably when the oxidation temperature is increased above 150°C. As can be seen from the results given in Fig. 5, the carboxylic content of the coal becomes significant only after the oxidizing temperature is raised above 200°C. Zeta potential measurements were also carried out with coal particles that

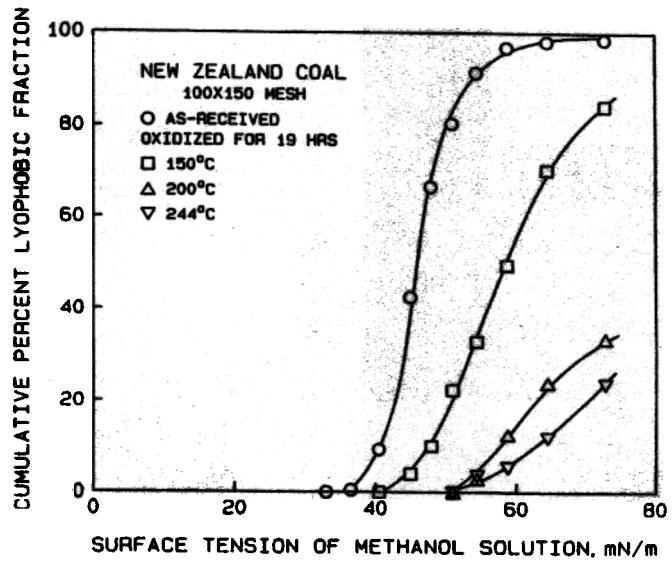


Fig. 4. Film flotation partition curves of New Zealand coal, as-received and thermally oxidized for 19 h at 150°, 200° and 244°C (after Diao and Fuerstenau, 1992).

had undergone the same degree of oxidation. The results given in Fig. 6 show that the zeta potential becomes increasingly negative in solutions at acidic pH's with increasing oxidation. Since this coal is virtually ash-free, increased oxidation must give rise to

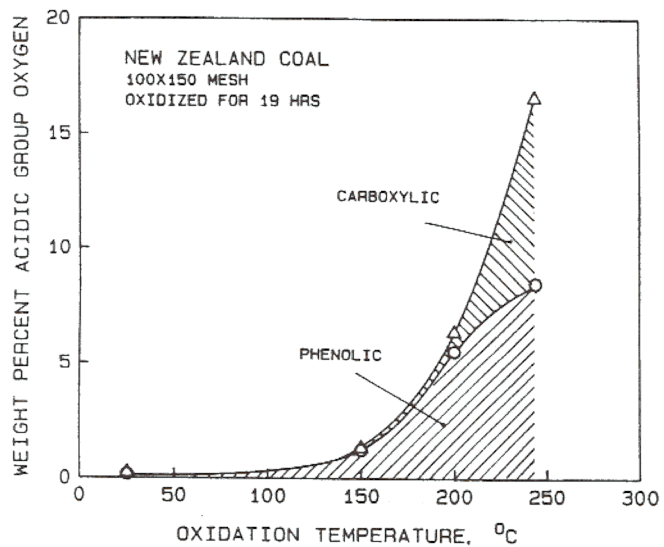


Fig. 5. The effect of oxidation temperature on weight percent oxygen in the form of carboxylic and phenolic groups on the surfaces of New Zealand coal oxidized for 19 h (after Diao and Fuerstenau, 1992).

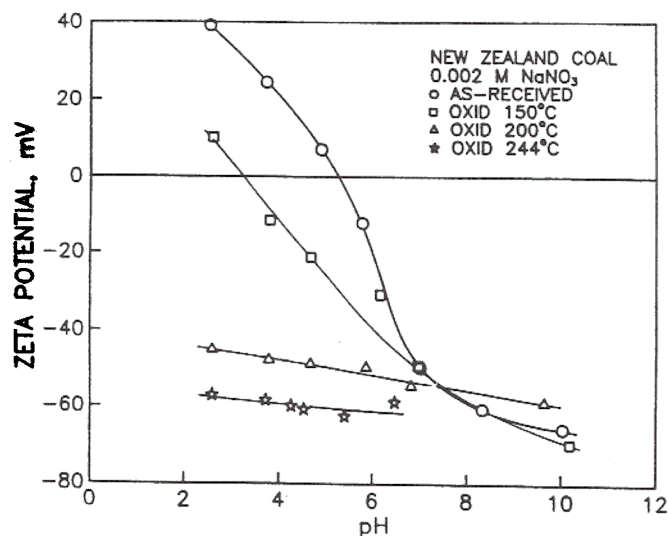


Fig. 6. The effect of oxidation temperature on the zeta potential of New Zealand coal oxidized for 19 h (after Diao and Fuerstenau, 1992).

anionic functional groups that are not amphoteric since the zeta potential of highly oxidized coal remains negative even down to pH 2. These results are in general consonance with those reported by Sun and Wen.

While most research using coal have been done with samples oxidized under controlled laboratory conditions, Fuerstenau et al. (1994) studied the flotation behavior of coal exposed to natural weathering for a period of 13 months under both open and inert (storage under argon atmosphere) conditions. The flotation response of one coal sample as a function of weathering time is illustrated in Fig. 7. Generally, as expected, the longer the weathering of the samples, the lower was the flotation yield. Even the sample stored under inert condition show signs of oxidation, as indicated by the reduced flotation yield, suggesting that this sample was susceptible to oxidation by the very small residual oxygen in the coal pores and in the purged storage container.

It was also observed that natural oxidation is mainly limited to the external surfaces. Coal stored at coarse size (0.60×6.4 mm) and then ground to minus 28 mesh (0.60 mm) before flotation exhibited a higher flotation yield than that stored as fines (minus 28 mesh). Evidently, grinding generates sufficient fresh surface that minimizes the effects of the external oxidized surface. The implication of this on processes involving fine particles treatment is to be noted.

The effect of weathering on the wettability of coal is illustrated in Fig. 8. Induction time is the minimum time necessary for the film of liquid between a bubble and a coal particle to thin and rupture, leading to stable bubble-particle contact. Film flotation is a method to quantify the wettability by determining the surface tension of a liquid that just wets the coal particles. The results given in Fig. 8 show that both film flotation and standard flotation yield, as well as the induction time, decrease with weathering time.

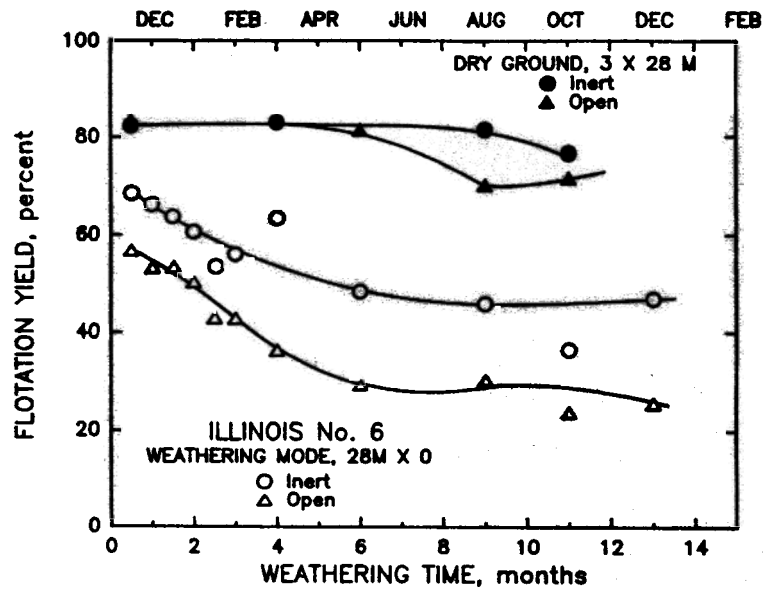


Fig. 7. Effect of weathering time and regrinding on the flotation yield of coal stored under inert and open modes (after Fuerstenau et al., 1994).

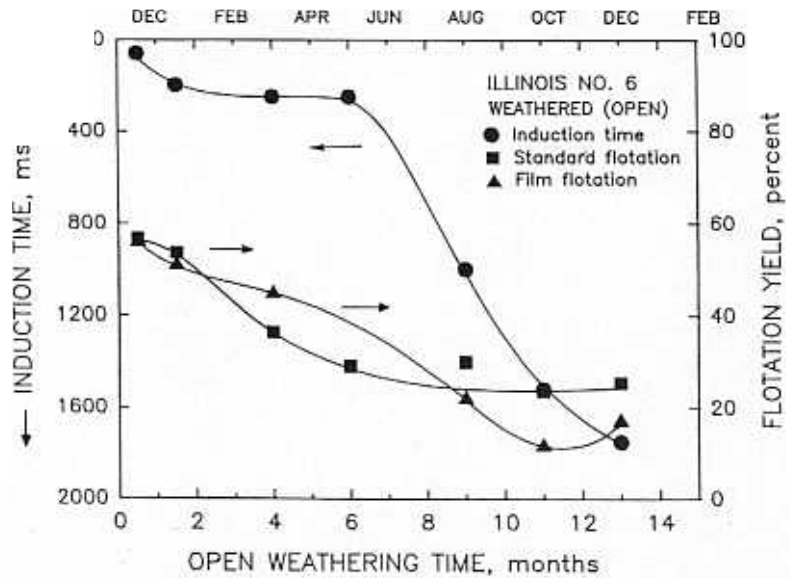


Fig. 8. Induction time, standard flotation and film flotation yields for Illinois No. 6 coal weathered sample stored under open mode as a function of weathering time (after Fuerstenau et al., 1994).

Generally, it can be concluded that oxidation changes the surface composition of coal and affects its hydrophobicity. This effect on coal processing, and particularly the treatment of fine particles, is to be noted.

3. Effect of multivalent ions

Chemical oxidation by oxidants such as KMnO_4 and H_2O_2 has also been used in the study of oxidation effects on coal. Celik and Somasundaran (1980) reported that treating the coal with either an oxidizing or reducing agent, or even with an acid or alkali in concentrations in excess of 10^{-2} mole/l can decrease the floatability. Thus, the effects of these reagents cannot be totally due to oxidation. Reagents such as KMnO_4 and SnCl_2 were also found to exert an influence on the surface properties due to possible adsorption or precipitation of various inorganic species.

The effects of different hydrolyzable multivalent ions were studied in detail (Celik and Somasundaran, 1986). The adsorption of aluminum and calcium as a function of pH is given in Fig. 9. Adsorption of calcium increases slightly with pH up to about 8 and then sharply above that value, while that of aluminum exhibits a sharp increase around pH 3–5. The sharp uptake of these metal ions appears to be governed by the formation of CaOH^+ and AlOH^{2+} . The percentage of coal floated in the presence of aluminum and iron is presented in Fig. 10 as a function of equilibrium pH. In the system with added iron salts, flotation goes through a distinct minimum at about pH 2.5 where the maximum amount of FeOH^{2+} and Fe(OH)_2^+ are expected to form (Fuerstenau and Palmer, 1976). At higher pH, the flotation dependence on pH was similar to that

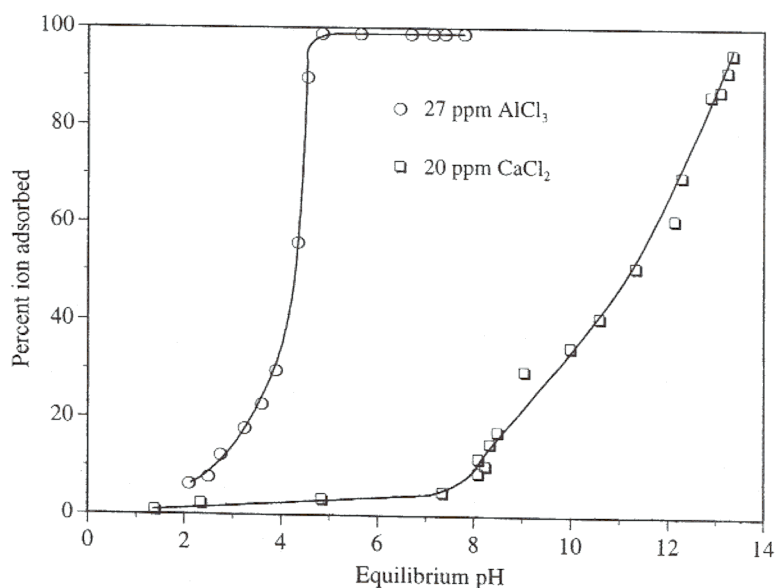


Fig. 9. Adsorption of calcium and the aluminum from coal at 25°C (after Celik and Somasundaran, 1986).

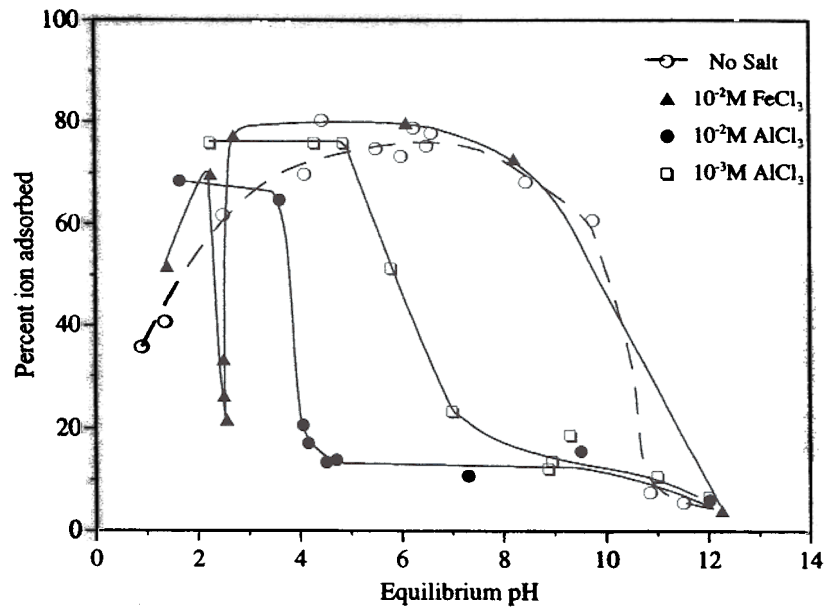


Fig. 10. The flotation of coal as a function of equilibrium pH in the presence of $FeCl_3$ and $AlCl_3$ (after Celik and Somasundaran, 1986).

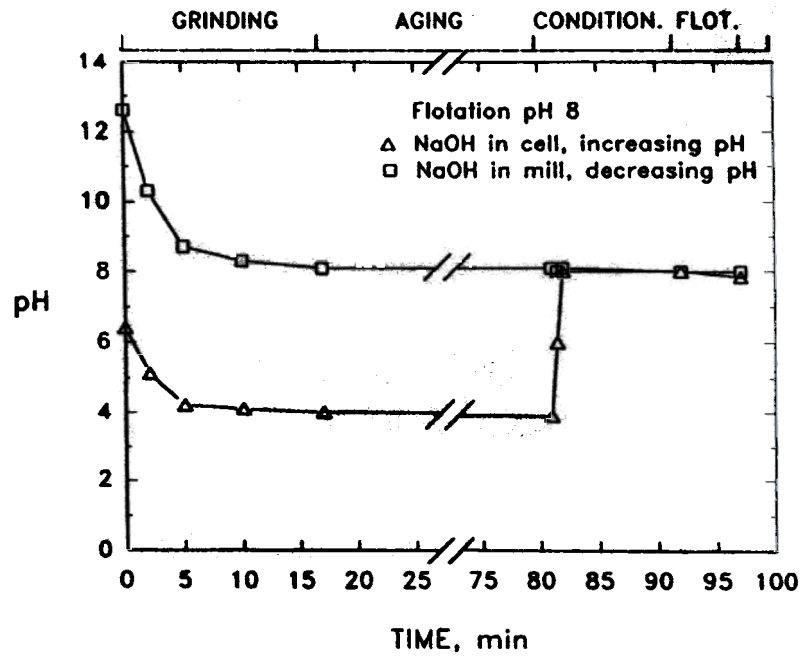


Fig. 11. pH vs. time curves for sodium hydroxide addition in the mill and in the flotation cell (after Liu et al. 1994).

obtained in water. In the case of AlCl_3 , the percentage of coal floated decreases with AlCl_3 in the alkaline region. This decrease in flotation correlates with the formation of hydroxyl complexes of aluminum as well.

The foregoing results clearly show that the adsorption of multivalent species can drastically affect the hydrophobicity of coal, with good correlation between the floatability of coal and the precipitation of metal hydroxy complexes. Depression of flotation is possibly caused by surface modification due to such surface precipitation.

4. Dissolved mineral species

Apart from the externally added organic and inorganic reagents, coal flotation is affected also by precipitation or adsorption of the dissolved mineral species from the coal itself.

The release of mineral ions was examined as a function of pH using two different pH controlling methods: NaOH addition in the flotation cell and NaOH addition in the grinding mill (Liu et al., 1994). The changes in pH of the coal slurry, during the sequence of grinding, aging of the ground slurry, conditioning (with alkali and reagents) and flotation are illustrated in Fig. 11 as a function of time for both modes of

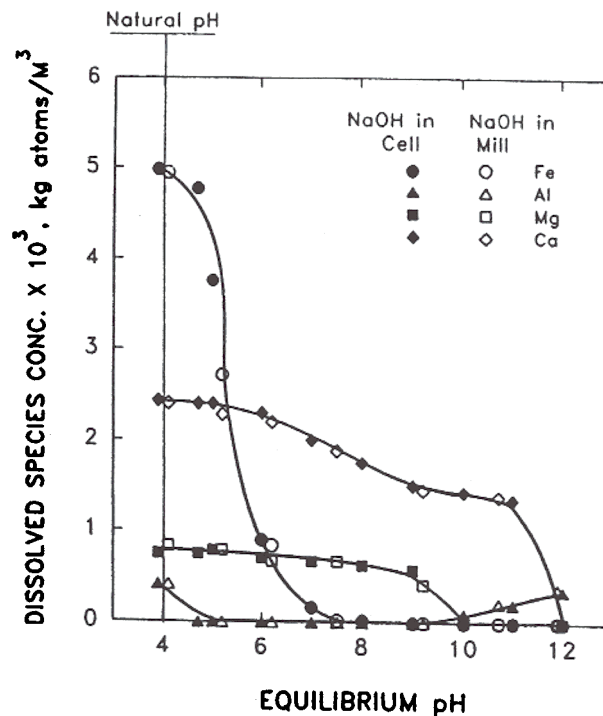


Fig. 12. Concentration of dissolved mineral species as a function of pH for sodium hydroxide addition in the mill and in the flotation cell (after Liu et al., 1994).

controlling. It can be seen that the path of approach towards the flotation pH depends strongly on the point of NaOH addition. When the sodium hydroxide is added in the grinding mill, the pH of the slurry decreases during the grinding and remains constant during the aging, conditioning and flotation steps. In this case the slurry is subjected to only pH decreases. On the other hand, when grinding is carried out in water, the pH of the slurry drops from 6.2 to 4. When NaOH is added in the flotation cell, the slurry is subjected to a pH increase. Such changes in pH can cause precipitation of metal species which affects the flotation behavior of the particles.

The concentration of dissolved mineral species from a Pittsburgh No. 8 seam sample in the ground slurry under the two different modes of pH adjustment is shown in Fig. 12. The concentrations of dissolved Fe, Al, Ca and Mg decrease as the pH is increased, with the mode of alkali addition being irrelevant. This result, together with the pH changes shown in Fig. 11, suggests that if the pH increases during coal processing, there will be precipitation of metal ion species whereas if the pH decreases, there will be dissolution of mineral species.

The effect of precipitation of the dissolved mineral species on flotation is shown in Fig. 13. Under the non-precipitation conditions (NaOH added in the mill), there is a small increase of flotation recovery with pH in the acidic region (pH 4 to 7) and a decrease between pH 8 and 10. However, under the precipitation conditions (NaOH added in the cell), the flotation recovery decreases to a greater extent in the pH range tested. This suggests that the hydroxide precipitate from the dissolved mineral species

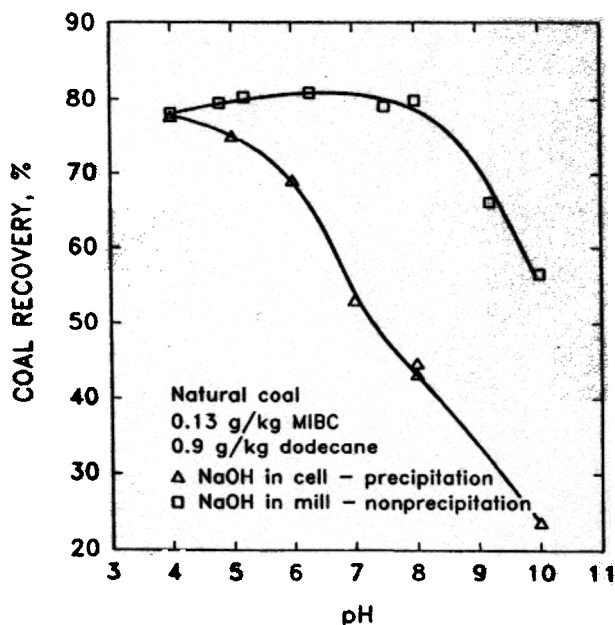


Fig. 13. Comparison of effects of sodium hydroxide addition to the mill and to the cell on coal flotation (after Liu et al., 1994).

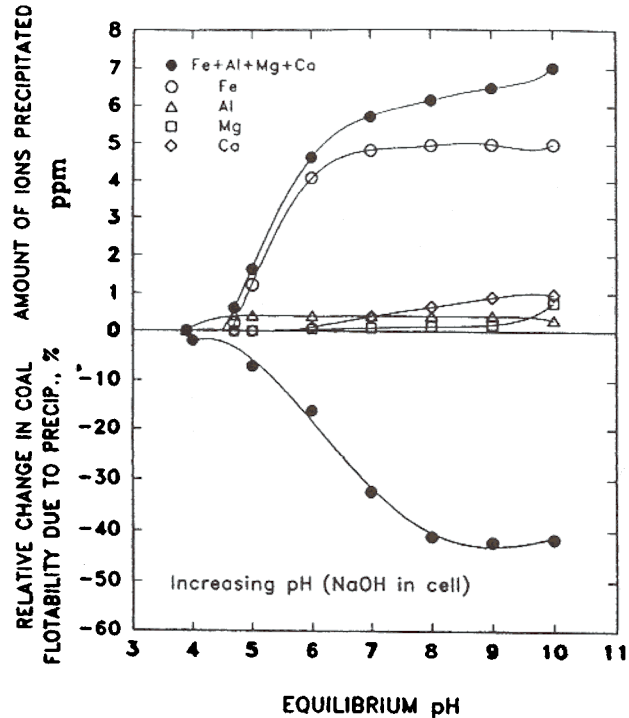


Fig. 14. Effect of different dissolved mineral ions on the floatability of coal (after Liu et al., 1994).

adsorbs on the coal surface, thereby imparting sufficient hydrophilicity to cause depression of the coal. Similar phenomena were shown for the depression of talc by hydroxylating metal ions (Fuerstenau et al., 1988).

The relative change in the floatability of coal as a function of pH is compared in Fig. 14 with the extent of overall precipitation as well as the precipitation of individual species. It can be seen that Fe species is the predominant component of the precipitate over the entire pH range tested. Also, above pH 7, there is no further increase in the Fe precipitation. A comparison of the Fe concentration curve with the floatability curve indicates that precipitation of Fe species is probably the major reason for the decreased hydrophobicity of the coal, and the effects of Ca, Mg and Al species are probably minor.

5. Summary

The flotation of coal, which depends on the surface properties of coal particles, can be drastically affected by oxidation and by the presence of various inorganics. Oxidation is a natural process that can continuously decrease the hydrophobicity of coal. In the flotation pulp, not only the added chemicals but also the dissolved mineral species released from coal itself during grinding and pulping can play an important role in

governing flotation. Understanding of the foregoing depression effects offers opportunities to enhance coal flotation. For example, oxidation can be alleviated by reducing the surface area exposed to air, and the presence of dissolved mineral species can be controlled by manipulating the pH at different stages of processing.

It might be pointed out that because of the heterogeneous nature of coals, these effects can vary to a large extent from coal to coal. A systematic study of these effects on the relevant surface properties of coal along with a thorough understanding of the mechanisms involved are necessary for better utilization of coal.

Acknowledgements

The authors acknowledge partial financial support from the National Science Foundation (EEC-9804618)

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Mineral Deposits of the World

Ores, industrial minerals and rocks

Edited by M. Vaneček

Developments in
Economic
Geology Volume 28

*Distributed in the East
European Countries,
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Mongolia, North Korea
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This overview of the world's mineral deposits lists the metallic and non-metallic mineral deposits according to continents, subdividing the latter with respect to regional metallogenic zones. The introductory chapters summarize present knowledge of the basic concepts of the geologic structure and the evolution of the continents and oceans. The description of the essential metallogenic units has been complemented by main aspects of the development of global metallogeny in time and space. The characteristics of the world's mineral wealth have been supplemented with basic data on the

amount, extent and manner of using these mineral resources. Economic geologists will find this volume of interest.

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1994 520 pages
Dfl. 375.00
(US \$ 214.25)
ISBN 0-444-98667-7

Elsevier Science B.V.
P.O. Box 1930
1000 BX Amsterdam
The Netherlands

P.O. Box 945
Madison Square
Station
New York
NY 10160-0757

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