

A preliminary survey of the corrosivity of water in South African gold mines

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SYNOPSIS

Samples of water that had been collected underground from eight gold mines in South Africa were subjected to comprehensive analysis of the quality of the waters, and their corrosivity on mild steel was measured by the laboratory techniques of extrapolation from Tafel plots and of polarization resistance.

The results indicate that, if such factors as pH, Langelier index, and degree of aeration are beyond certain specified limits, the corrosivity of the water is affected. The applicability of the corrosivity index in predictions of the degree of corrosion is questioned. Suggestions are included for future work involving the fundamental characterization of the corrosivity of mine waters.

SAMEVATTING

Watermonsters wat ondergronds uit ag goudmyne in Suid-Afrika versamel is, is aan 'n omvattende ontleding van die gehalte van die water onderwerp en die korrosiwiteit van die water vir sagtestaal is gemeet deur die laboratoriumtegnieke van ekstrapolering aan die hand van Tafel-grafieke en polariseringsweerstand.

Die resultate toon dat die korrosiewerking van die water geraak word as faktore soos die pH, die Langelier-indeks en die mate van belugting sekere gespesifiseerde perke oorskry. Die toepaslikheid van die korrosiwiteits-indeks op voorspellings van die korrosiegraad word bevestig en alternatiewe word aangevoer. Daar word ook voorstelle vir toekomstige werk in verband met die fundamentele karakterisering van die korrosiwiteit van myn-water ingesluit.

Introduction

The failure of mining equipment such as water-carrying equipment as a result of corrosion is a major problem in the South African mining industry.

The water found in mines, which is mainly due to infiltration, is used in processes and machines and as feed for refrigeration units. If a proper understanding is to be obtained of the corrosion mechanisms involved, the corrosivity of this water must be determined at various stages of treatment and use.

The motivation for the investigation described here was a need for reliable data on the quality of the water in various parts of South African gold mines. It was hoped that the information on corrosivity collected during the course of the investigation, which would be based on representative samples, would form a body of data that could be accepted with a fair degree of certainty.

Despite the importance of the problems arising from the corrosivity of naturally occurring waters, very little systematic information can be found on the relation between the rate of corrosion and the properties of the water (such as pH, conductance, total dissolved solids, and the Langelier index¹).

The Langelier index (LI) is the difference between the actual pH value of the water and the saturation pH value, i.e.

$$LI = \text{pH (measured)} - \text{pH}_s \text{ (computed)},$$

where pH_s is the saturation pH value, i.e. the pH value at which solid calcium carbonate is in equilibrium with its saturated solution.

If the LI is positive, it means that the water is supersaturated with calcium carbonate and that the deposition of a protective film should occur. When the LI is negative, any protective film that is present is stripped and

the bare metal is exposed to attack.

The Langelier equation, which is based upon the carbonate equilibrium, is

$$\text{pH}_s = (\text{p}K_2 - \text{p}K_s) + \text{pCa}^{2+} + \text{pAlk} + \log \left| 1 + \frac{2K_2}{(\text{H}^+)_s} \right|,$$

where

pCa^{2+} is the negative log of the Ca^{2+} concentration in gram molecules of calcium carbonate per litre,

pAlk is the negative log of alkalinity to methyl orange expressed as the gram-equivalent mass of calcium carbonate per litre,

$\text{p}K_2$ is the negative log of K_2 , which is defined by the equation

$$K_2 = \frac{(\text{H}^+)(\text{CO}_3^{2-})}{(\text{HCO}_3^-)}, \text{ and}$$

$\text{p}K_s$ is the negative log of solubility product, K_s , of calcium carbonate, i.e. $K_s = (\text{Ca}^{2+})(\text{CO}_3^{2-})$.

If the pH values range between 6 and 9.5, the term

$$\log \left| 1 + \frac{2K_2}{(\text{H}^+)_s} \right|$$

can be omitted from the Langelier equation.

The pH_s values used in this work were taken from the chart published by Powell².

In a survey of mine waters in Indian coal mines, Rawat³ found that the total dissolved solids ranged from 2000 to 2600 p.p.m. The LI did not correlate well with the corrosion rate (in the testing of mild steel with coupons) since a rate of about 0.1 mg/cm² was found over a saturation index from -1.0 to +1.0. It was concluded that corrosion rates depend on the amount of aggressive ions (chloride and sulphate) present. The corrosion rates in mine waters were of the same order as those of solutions with the same concentration of sodium chloride.

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The corrosion rate of iron in oxygenated water, which was thought to depend on the concentration of dissolved oxygen, drops to a low value below a certain critical oxygen level. Corrosion effects due to dissolved oxygen are reported to be enhanced in water of low pH.

Subrahmanyam and Hoey⁴ investigated the influence of water quality on the corrosion of mild steel in synthetic acidic mine waters, using measurements of loss in mass and of polarization resistance. The waters were adjusted to a pH value of 5 by the addition of sulphuric acid, and contained the ions Fe²⁺, Fe³⁺, Cu²⁺, Cd²⁺, Mg²⁺, Zn²⁺, K⁺, Na⁺, Cl⁻, SO₄²⁻, and SO₃²⁻.

The de-aeration of stirred solutions containing ferric and cupric ions lowered the corrosion rate by the same amount as did the addition of sulphite ions. Hence, it can be concluded that the sulphite ion removes dissolved oxygen if the solution is stirred sufficiently. It was concluded that the corrosion rate, in the absence of the sulphite ion, was controlled by the amount of ferric and cupric ions in solution. The presence of the chloride ion had an accelerating effect on the corrosion in stirred solutions, but this effect was obscured in the presence of ferric and cupric ions. None of the other ions investigated had any effect on the corrosion rate.

Cathodic polarization curves for mild steel in synthetic mine waters exhibited a significant amount of concentration polarization, indicating that the reduction of H⁺, O₂, Cu²⁺, and Fe³⁺ was largely under diffusion control.

Johnson and Omura⁵, also using techniques involving loss in mass and polarization resistance, measured the corrosion rates of low-carbon steel in aerated samples of ground water (natural and synthetic). The natural samples gave corrosion rates in the range 0,15 to 0,65 mm per year, whereas, in synthetic solutions containing

between 0,01 and 0,1 per cent NH₄NO₃, the corrosion rates were 0,27 to 0,5 mm per year. In general, the more corrosive waters contained higher concentrations of the anionic species Cl⁻, SO₄²⁻, and NO₃⁻.

In their work on corrosion rates on mild steel in underground waters, Clarke and Barnes⁶ took measurements of the water temperature, redox potentials, pH values, and bicarbonate and ferrous ion concentrations. Many possible solid phases were investigated, but in only two, ferric hydroxide and calcium carbonate, was there any correlation with the rate of corrosion, both solids providing partial protection.

Finan *et al.*⁷ established a quantitative correlation between the corrosion rate of mild steel and the Ryznar index (which is equal to 2 pH_s - pH) in circulating natural waters. Values of less than 6 for the Ryznar index denote a tendency towards the formation of protective calcareous scales, and values above 7 denote aggressive conditions to the presence of free carbonic acid.

In summary, attempts at the correlation of the composition of natural water with corrosivity led to the conclusion that the LI is not a reliable guide to the corrosion rate. Although factors such as pH, dissolved oxygen, and various ions (chloride, sulphate, nitrate, iron, and copper) were established as having some influence on corrosion rates, no comprehensive, systematic study of the effects of these factors has been reported, even for mild steel. It was therefore decided that the present investigation should start with a survey of the corrosivity of mine waters on mild steel as determined by the use of electrochemical methods involving extrapolation from Tafel plots and measurements of polarization resistance.

TABLE I
THE SAMPLING POINTS

Mine	Shaft	Level	Type of water
Western Deep Levels	3	87	(i) Make-up* (ii) Chill* (iii) Machine
Elandsrand		Pipeline on surface	Machine
Free State Geduld	2	53	(i) Machine (ii) Drain
President Brand	2 4	44 46 37	(i) Make-up* (ii) Chill* (iii) Dam
Grootvlei	1	Kimberley	(i) Machine (ii) Dam
Leslie		10	(i) Machine (ii) Drain (iii) Fissure
Stilfontein		Surface	(i) Machine (ii) Fissure
E.R.P.M.	Hercules	68	(i) Pump (to surface) (ii) Condenser*

*With reference to refrigeration plants

Experimental Procedures

Location of Sampling Points

The study was based on small areas within eight gold-mines, which covered the whole geographical distribution of the South African goldfields. The location and types of samples taken from the mines are listed in Table I. A minimum of three visits was paid to each sampling site over an overall period of some eight months.

Chemical Analysis

At each sampling point, tests were carried out on pH, conductivity, temperature of sample and air, and dissolved oxygen. Also, samples of the water containing 'fixed' dissolved oxygen were taken for titration by the Winkler method. Samples were stored at 4°C until they were required for testing, and were submitted for analysis as soon after sampling as possible. The following list gives the analyses that were carried out:

Alkalinity (methyl orange)	S ²⁻
Hardness (as CaCO ₃), Ca, and Mg	pH
Cl ⁻	Conductance
SO ₄ ²⁻	Chemical oxygen demand
NO ₃ ⁻ (as N)	Na
NO ₂ ⁻ (as N)	Fe
NH ₃ (as N)	Cu
Total dissolved solids	Mn
Total suspended solids	Al
Total organic suspended solids	

Procedure for Electrochemical Tests

The Princeton 350A Corrosion Measurement Console was used to record Tafel plots and plots of polarization resistance for samples of mild steel with an exposed area of 2.1 cm² immersed in mine water. The analysis of the mild-steel rod used is given in Table II. The samples were polished to a finish of 3 μm and ultrasonically degreased in methanol before being immersed in mine water, which had previously been saturated with air or high-purity nitrogen at 25°C. The gas was bubbled in slowly throughout the course of the experiment. Tafel plots were measured over a range of 500 mV at a sweep speed of 0.1 mV/s, starting from an initial potential of -250 mV from the corrosion potential after 30 minutes of equilibration. Four separate Tafel plots were recorded for each sample under identical experimental conditions for the aerated and the de-aerated states.

TABLE II
ANALYSIS OF MILD STEEL

Element	Concentration %
Carbon	0,14
Sulphur	0,068
Phosphorus	0,031
Manganese	0,75
Chromium	0,13
Silicon	0,26
Copper	0,094
Nickel	0,056
Aluminium	< 0,005
Tin	< 0,01

The rate of corrosion was calculated from the extrapolation of the Tafel lines (the linear sections of the anodic and cathodic branches of the polarization curve where E is proportional to log current density) to the corrosion potential E_{CORR} . For each experimental run, a second estimate of the corrosion current was obtained from the slope of the graph for potential versus current density recorded within 15 mV from the corrosion potential. This is known as the polarization resistance, R_p . The curve for potential versus current density is linear to a first approximation in this region, and the slope R_p is related to the corrosion current I_{CORR} by the Stern-Geary equation

$$I_{CORR} = \frac{1}{2,3R_p} \left(\frac{b_a \times b_c}{b_a + b_c} \right),$$

where b_a and b_c are the anodic and cathodic Tafel constants respectively. Hence, for each mine water, eight separate values of I_{CORR} were recorded for the aerated and for the de-aerated states, and the results were averaged. E_{CORR} values for each experimental run were also recorded and averaged.

Results and Discussion

Water Quality

The distribution of pH values found in the samples of mine waters is shown in Fig. 1. Except for a few samples, the range in pH was from 6 to 9, which is an indication that the control of pH carried out within most mines is satisfactory. The concentrations of dissolved oxygen (Fig. 2) found at the sampling points were generally within the range 5 to 9 mg/l, which is close to 100 per cent saturation. A few very low concentrations (less than 4 mg/l) of dissolved oxygen were determined, chiefly in areas of fissure waters with high sulphide concentrations.

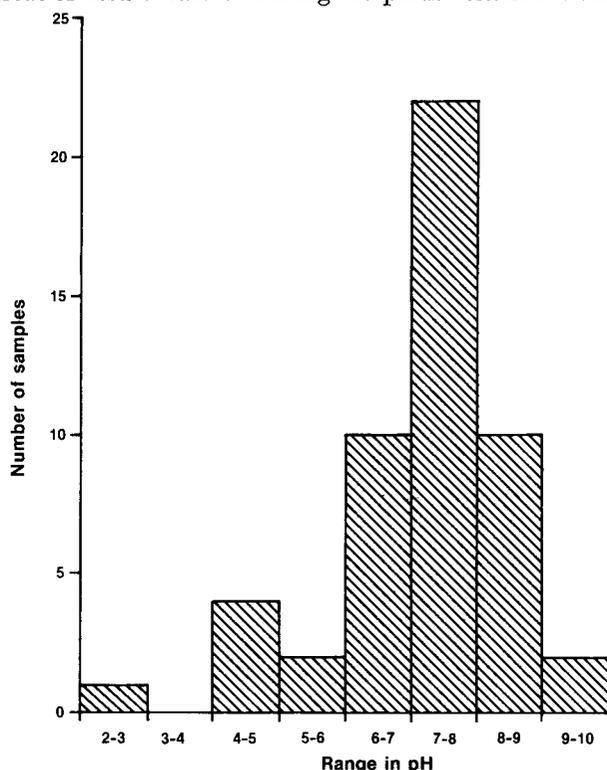


Fig. 1—Distribution of pH range in samples of mine water

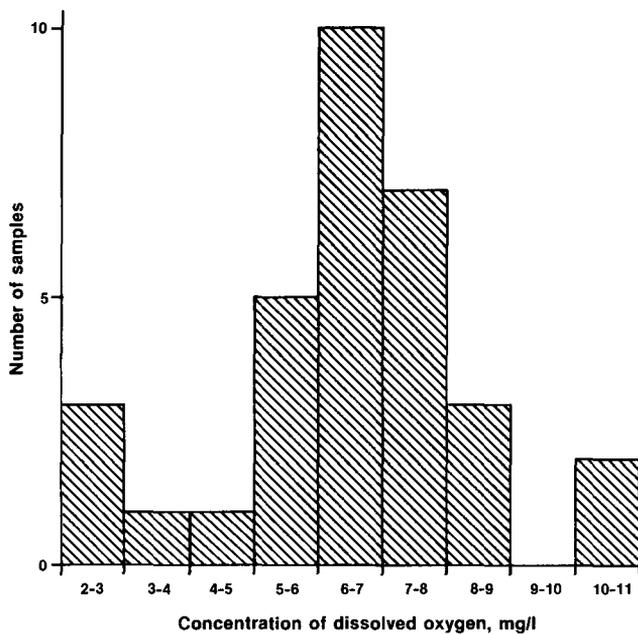


Fig. 2—Distribution of dissolved oxygen in samples of mine water

The concentrations of total dissolved solids (Fig. 3) showed values of up to 8 g/l, with the majority of samples within the range 1 to 3 g/l. The total dissolved solids content consisted mainly of sulphates (Fig. 4) and chlorides (Fig. 5). The range of total hardnesses, expressed as grams of calcium carbonate per litre, is shown in Fig. 6.

Minor constituents of most of the samples of mine waters were found to be nitrate (usually below 50 mg/l but up to 150 mg/l expressed as N) and ammonia (usually below 20 mg/l but up to 50 mg/l expressed as N). These constituents are thought to result mainly from the spillage of an explosive based on an ammonia salt.

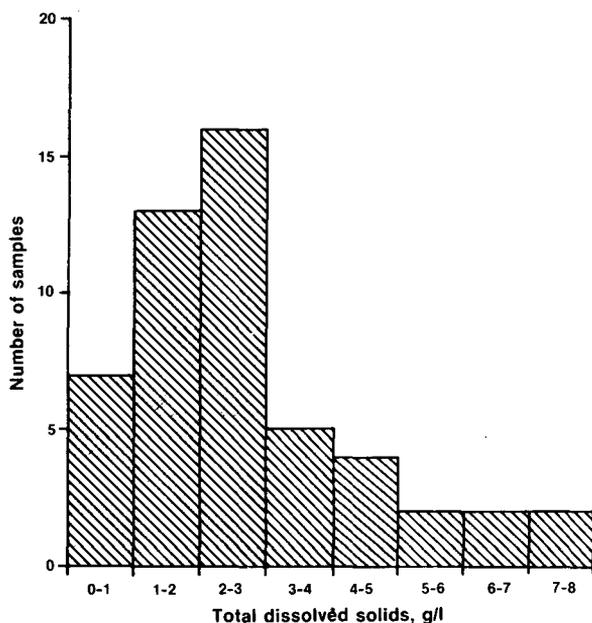


Fig. 3—Distribution of total dissolved solids in samples of mine water

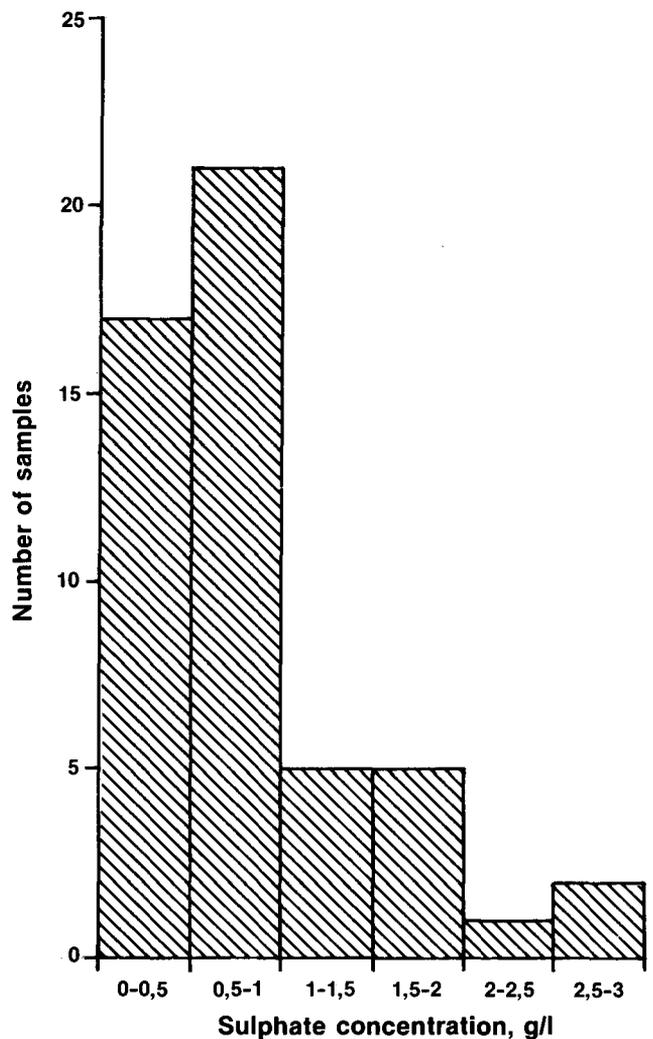


Fig. 4—Distribution of sulphate in samples of mine water

Fig. 7 shows that, in general, a linear relationship exists in the samples of mine water for the variation of total dissolved solids versus conductance with a slope of 0,76. This linear relation is generally applied in mine-refrigeration technology, where the measured conductance is used in the calculation of the total dissolved solids in the system. This enables blowdown to be carried out before the concentration of total dissolved solids in the water rises to an unacceptable level, where scaling and precipitation, especially on heat-transfer surfaces, are probable. The good correlation shown in all the waters sampled leads to the conclusion that this can be taken as an accurate relation throughout South African gold mines.

The variation of LI with pH is shown in Fig. 8. This linear relation is described by

$$LI = (1,15 \times \text{pH}) - 8,7.$$

Owing to the nature of the evaluation of the LI, a relation of this type would be expected. Moreover, this means that a good estimate can be made of the LI from a knowledge of the pH value alone. This information should prove useful to operators of refrigeration plants on which a pH probe is provided and where plant-design

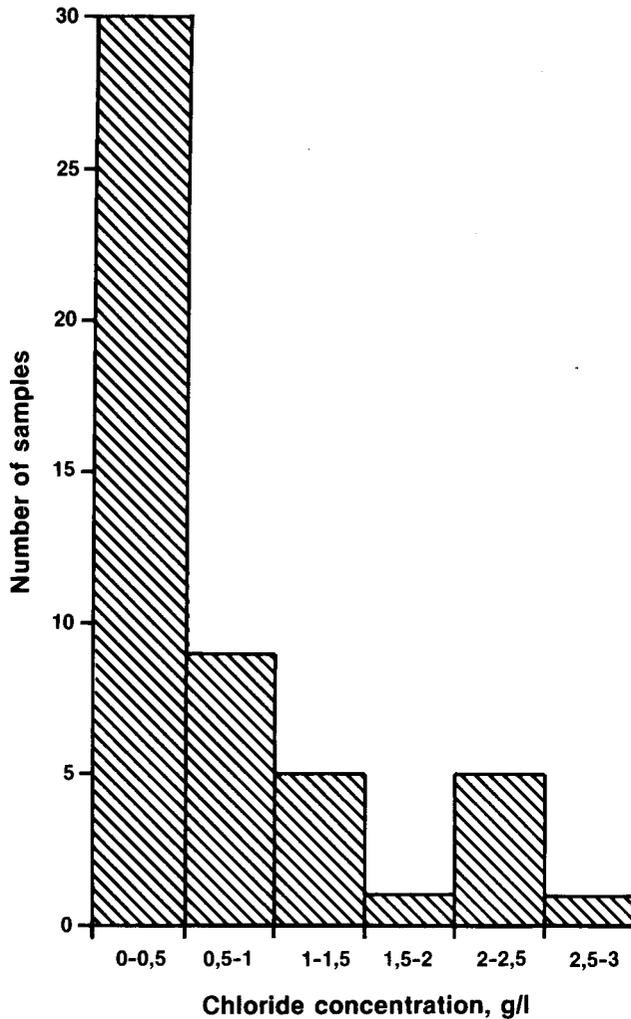


Fig. 5—Distribution of chloride in samples of mine water

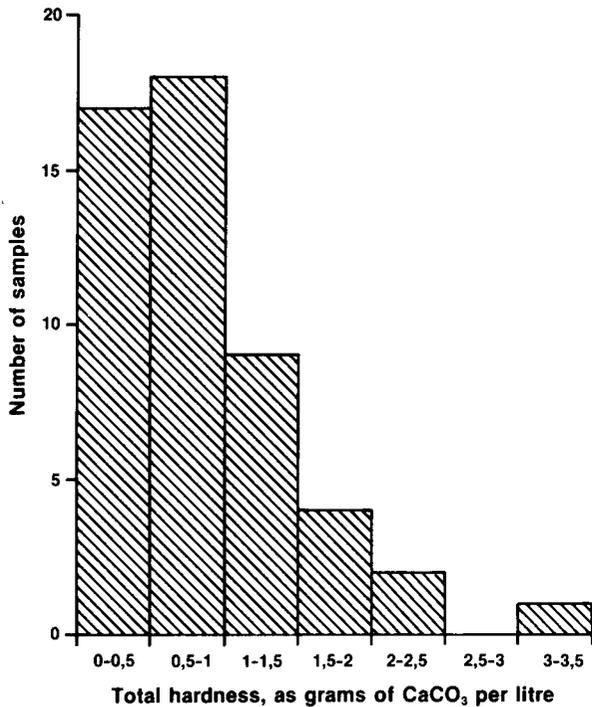


Fig. 6—Distribution of total hardness in samples of mine water

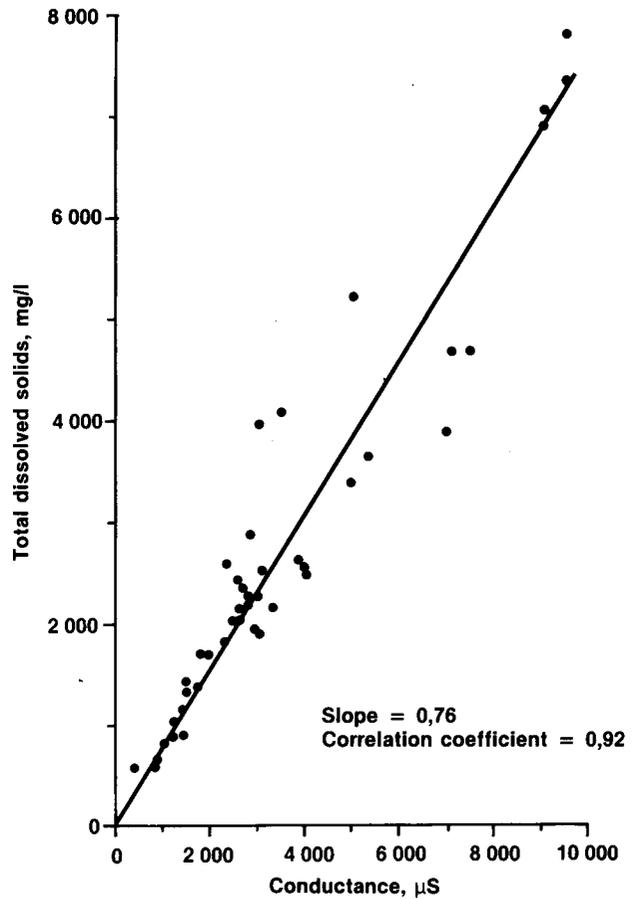


Fig. 7—Variation of total dissolved solids with conductance

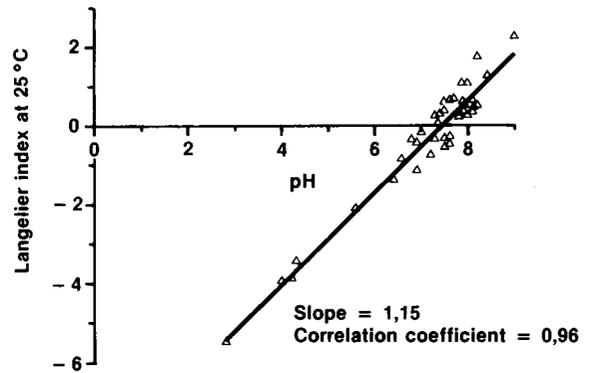


Fig. 8—Variation of Langelier index with pH

criteria require the plant to adhere to a specified value of LI.

Corrosion Rates

Fig. 9 shows typical Tafel plots for mild steel in mine water in the aerated and de-aerated states. De-aeration results in a decrease in the corrosion rate and a negative shift in the corrosion potential. The high cathodic Tafel slope (i.e. $dV/d\log i$) for the aerated solution (approximately 0,52 V) indicates that the cathodic reaction is largely diffusion-controlled. This is consistent with the cathodic reaction being the reduction of dissolved molecular oxygen, i.e.

$O_2 + 2H_2O + 4e \rightarrow 4OH^-$,
because of the low solubility of oxygen in natural waters.

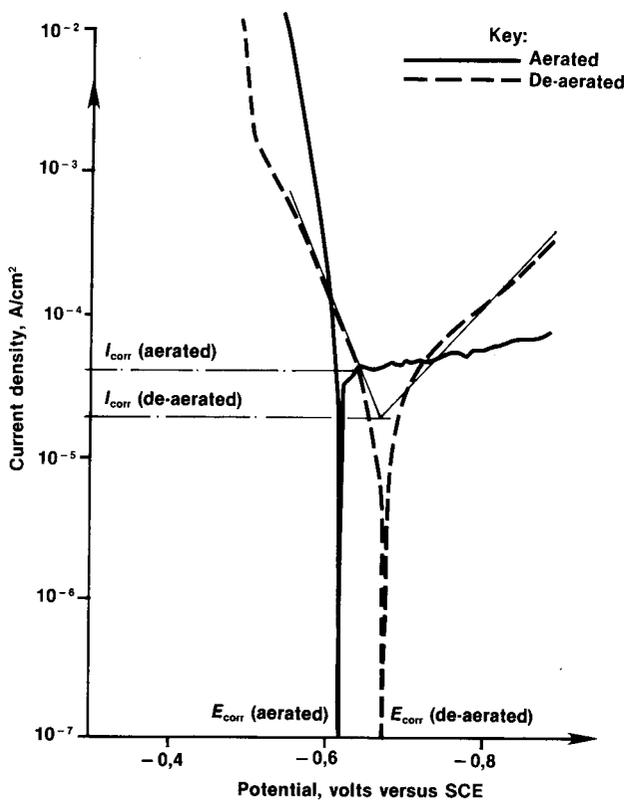


Fig. 9—Tafel plots for mine water, showing the effect of aeration

For the de-aerated solution, the value of the cathodic Tafel slope is somewhat smaller (0,2 V). The cathodic reaction in this instance is presumably the reduction of hydrogen ions to gaseous hydrogen, and the cathodic Tafel slope does not generally reflect the activation-controlled value of 0,12 V because of the relatively high pH values of these mine waters.

The anodic (positive) Tafel slope is generally near the activation-controlled value for the dissolution of iron (0,04 V) for both aerated and de-aerated mine waters. High values for the anodic Tafel slope and curvature of the anodic part of the polarization curve at high current densities generally reflect some limitation on the rate of dissolution due to the formation of dissolution products.

Fig. 10 shows typical plots of polarization resistance for mild steel in mine water in the aerated and de-aerated states. The higher corrosion rate for the aerated solution is represented by its lower value for polarization resistance (i.e. the lower value of dV/di near E_{corr}). Values found for the proportionality factor in the Stern-Geary equation, i.e.

$$\frac{b_a \times b_c}{b_a + b_c}$$

where b_a and b_c are the anodic and cathodic Tafel slopes respectively, were usually approximately 4×10^{-2} . There is greater curvature (i.e. greater deviation from the Stern-Geary relation) for the aerated solution. This is probably due to the large differences in value between the anodic and the cathodic Tafel constants, the cathodic process being diffusion-controlled.

Figs. 11 and 12 are block diagrams showing the distribution of corrosion rates (expressed in millimetres of penetration per year) in aerated and de-aerated mine waters respectively. Corrosion rates of as much as 0,6 mm per year were measured in aerated mine waters, the most frequent range being 0,2 to 0,25 mm per year. The corrosion rates in de-aerated mine waters were generally much lower, the maximum measured rate being below 0,3 mm per year and the most frequently measured range being 0,025 to 0,05 mm per year.

Correlations between Corrosion Rate and Water Quality

Fig. 13 shows the general trend of the values for corrosion rate as a function of pH for aerated and de-aerated mine waters. Above a pH value of around 5,5 the corrosion rate is fairly independent of pH for aerated and de-aerated solutions, the former having the average value of 0,237 mm per year and the latter 0,067 mm per year. Below a pH value of around 5,0, the corrosion rate rises linearly to a first approximation, with decreasing pH for aerated and de-aerated mine waters. The behaviour in this region is described by the following equations:

$$\text{Corrosion rate, mm per year} = \frac{5,43 - \text{pH}}{3,56} \text{ for aerated mine waters,}$$

$$\text{Corrosion rate, mm per year} = \frac{5,14 - \text{pH}}{5,01} \text{ for de-aerated mine waters.}$$

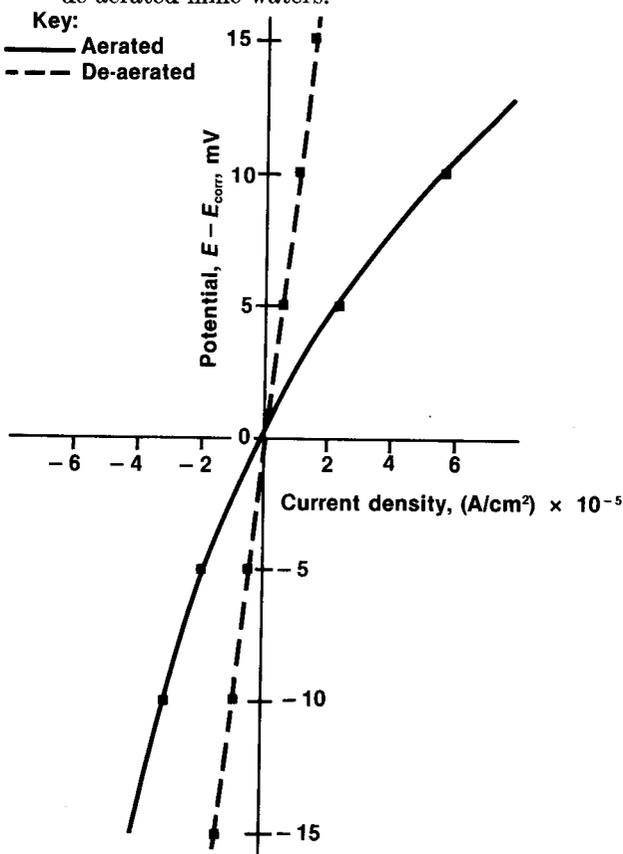


Fig. 10—Polarization-resistance plots for mine water, showing the effect of aeration

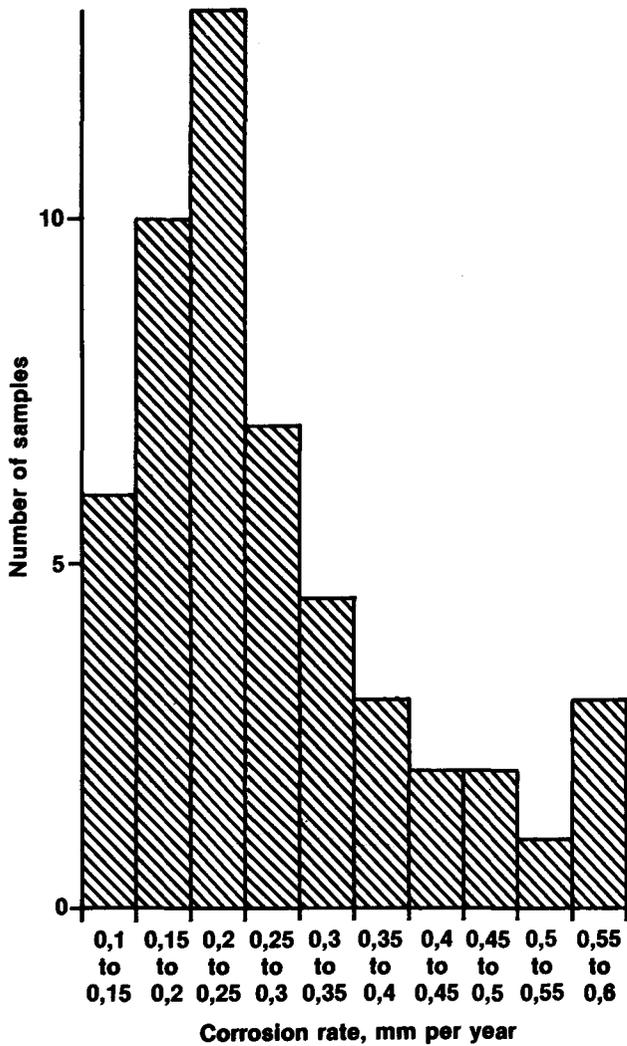


Fig. 11—Distribution of corrosion rates for aerated mine waters

Measurements of the dissolved oxygen content *in situ* before a polarization run showed it to be around 7 mg/l for aerated solutions and less than 0,1 mg/l for de-aerated solutions. De-aeration generally causes large reductions in the corrosion rate of mine waters with pH values greater than 5,0 (an average of 74 per cent) and smaller reductions for mine waters with pH values less than 5,0.

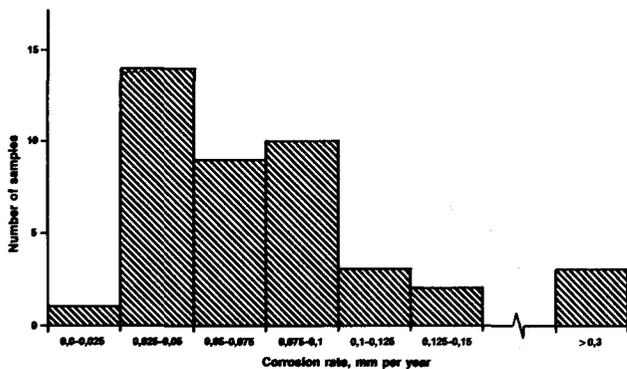


Fig. 12—Distribution of corrosion rates for de-aerated mine waters

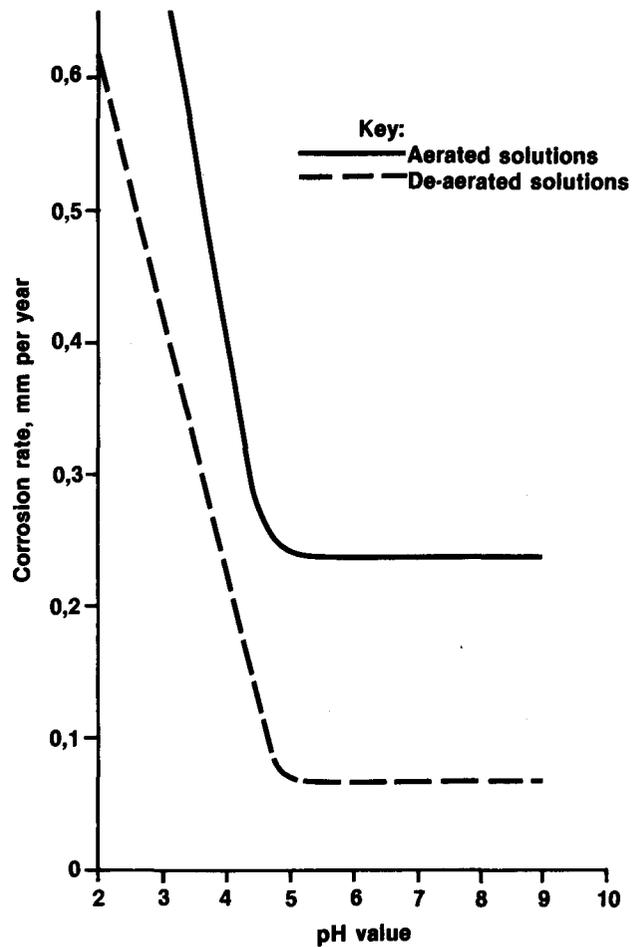


Fig. 13—Corrosion rate versus pH for mild steel in mine waters

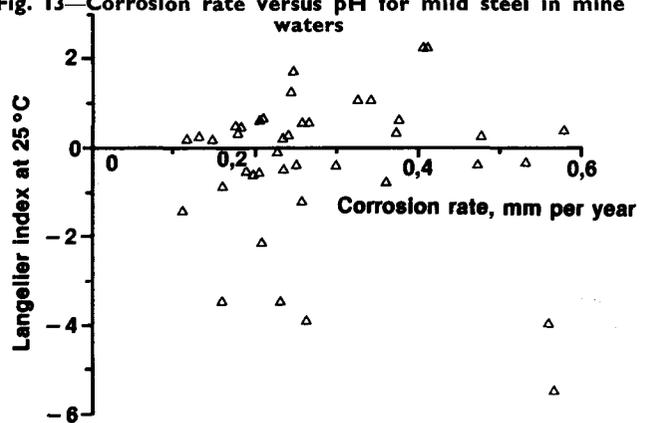


Fig. 14—Variation of corrosion rate with Langelier index in aerated mine waters

This implies that the increase in corrosion rate with aeration is due to the increased availability of the cathodic reactant (dissolved oxygen). For the mine waters with pH values of less than 5, the reduction in the corrosion rate with de-aeration is smaller because of the competition between appreciable quantities of hydrogen ions and dissolved oxygen to take part in the cathodic reaction.

Figs. 14 and 15 show LI versus corrosion rate for aerated and de-aerated mine waters respectively. For the

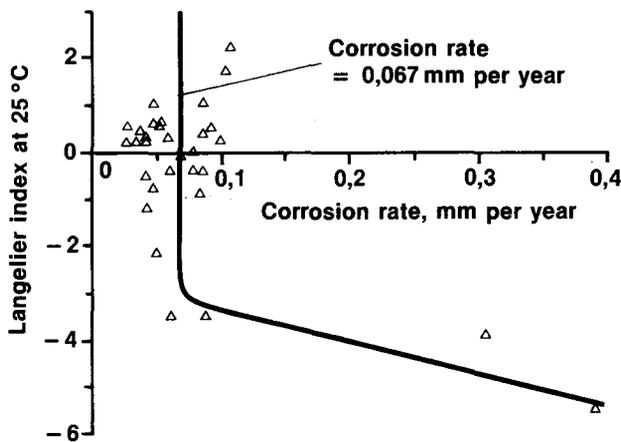


Fig. 15—Variation of corrosion rate with Langelier index in de-aerated mine waters

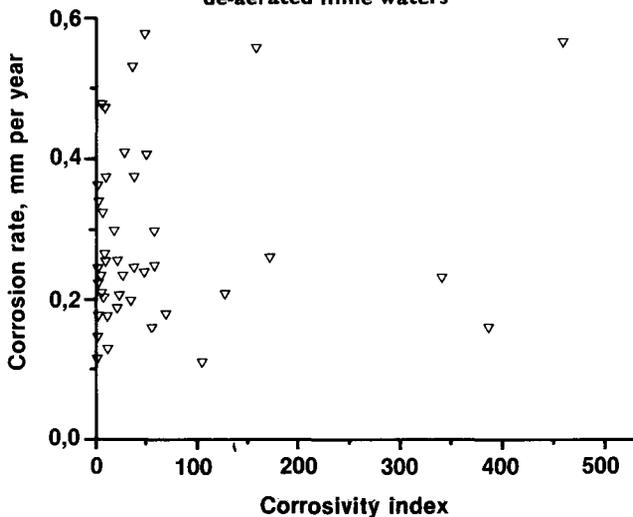


Fig. 16—Variation of corrosion rate with corrosivity index in aerated mine waters

aerated solution, there is no clear correlation between the corrosion rate and the LI for all positive and small negative values (more positive than -1), although an increase in corrosion rate with LI is evident at LI values that are more negative than -3 .

This trend of increasing corrosion rate with large negative values of LI is somewhat clearer for de-aerated solutions, being approximated by the linear relation

$$\text{Corrosion rate, mm per year} = \frac{(-2,49 - \text{LI})}{6,54}$$

for de-aerated solutions for an $\text{LI} < -3$.

Relationships of this type, between the corrosion rate and LI would be expected because of the underlying relationship between LI and pH.

Figs. 16 and 17 show the variation of corrosion rate with corrosivity index (CI) for aerated and de-aerated mine waters respectively. The CI reflects the different concentrations of aggressive and scaling ions, being defined by

$$\text{CI} = \frac{[\text{Cl}^-] + [\text{SO}_4^{2-}]}{[\text{HCO}_3^-]}$$

where the concentrations are expressed in gram milliequivalents and $[\text{HCO}_3^-]$ is given by the alkalinity with

respect to methyl orange. It can be seen from these diagrams that there is no apparent correlation between the corrosivity index and the corrosion rate for aerated and de-aerated mine waters. This result indicates that the use of the corrosivity index in the prediction of general corrosion rates in mine waters is not valid.

Other single factors that were investigated for their correlation with the corrosion rate were conductance, total dissolved solids, chloride concentration, sulphate concentration, total hardness, and alkalinity. No clear correlation was found between the corrosion rate and any one of these factors. Similarly, no correlation was found between the corrosion rate and any of the following combinations:

$$[\text{Cl}^-] + [\text{SO}_4^{2-}]$$

$$\text{total dissolved solids} \div \text{total hardness}$$

$$([\text{Cl}^-] \times [\text{SO}_4^{2-}]) \div \text{total hardness}$$

$$\text{pH} \times \text{conductance}$$

$$(\text{pH} \times \text{conductance}) \div \text{total hardness}$$

$$\text{total dissolved solids} - \text{total hardness.}$$

The results of all these correlations indicate that the concentration of dissolved oxygen is by far the largest single factor in determining the corrosion rate of mild steel in mine waters with a pH value of more than 5.0. Below that value (corresponding to LI values more negative than -3), the effect of pH becomes important.

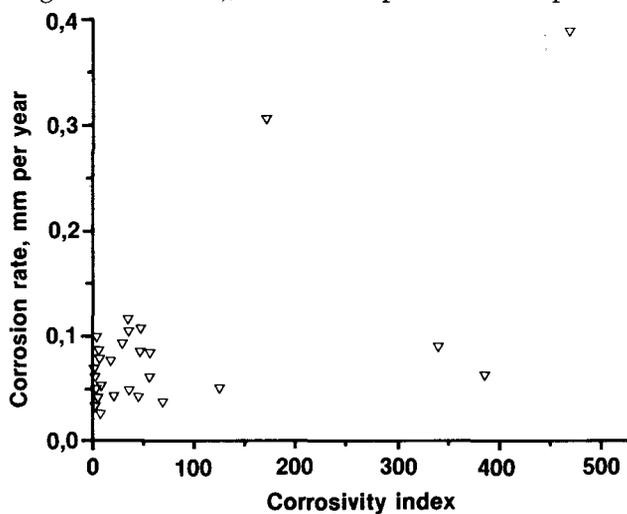


Fig. 17—Variation of corrosion rate with corrosivity index in de-aerated mine waters

Conclusions

- (1) The most common analyses for the mine waters tested were found to be in the following ranges: a pH value of 7 to 8, a dissolved oxygen content of 6 to 7 mg/l, a total dissolved solids content of 2 to 3 g/l, a total hardness of 0,5 to 1,0 g/l, a chloride content of 0 to 0,5 g/l, a sulphate content of 0,5 to 1,0 g/l, and a nitrate content of 10 to 50 mg/l.
- (2) The following two general relationships were established between water-quality parameters for these mine waters at 25°C:

$$\text{Total dissolved solids, mg/l} = 0,76 \times \text{conductance, } \mu\text{S}$$

$$\text{LI} = (1,15 \times \text{pH}) - 8,7.$$

- (3) For mine waters with pH values above 5,0, de-aeration resulted in an average decrease in the corrosion rate of 74 per cent. The corrosion rate is fairly independent of pH over this range, the average values being 0,237 mm per year for aerated water with a dissolved-oxygen concentration of about 7 mg/l, and 0,067 mm per year for de-aerated water with a dissolved-oxygen concentration of less than 0,1 mg/l. These results are consistent with diffusion-controlled oxygen reduction as being the dominant cathodic process and exercising control over the corrosion rate in this pH range.
- (4) For mine waters with pH values below 5,0, the corrosion rate rises approximately linearly with decreasing pH according to the relations

$$\text{Corrosion rate, mm per year} = \frac{5,43 - \text{pH}}{3,56} \text{ for}$$

aerated mine waters

$$\text{Corrosion rate, mm per year} = \frac{5,14 - \text{pH}}{5,01} \text{ for}$$

de-aerated mine waters.

Owing to the underlying relationship between pH and LI, the following relation was found for LI values more negative than -3:

$$\text{Corrosion rate, mm per year} = \frac{(-2,49 - \text{LI})}{6,54} \text{ for}$$

de-aerated mine waters.

In this pH range (below 5,0), there is competition between appreciable quantities of hydrogen ions and dissolved oxygen to take part in the cathodic reduction reaction.

- (5) No correlation was observed between corrosion rate and corrosivity index for aerated or de-aerated mine

waters. This indicates that the use of the corrosivity index in the prediction of general corrosion rates on mild steel in mine waters is not valid.

Further work on the characterization of the corrosivity of mine waters on mild steel is being undertaken at present. This involves a detailed investigation of the effect of the following properties on the corrosivity of a synthetic mine water:

- (i) dissolved oxygen content
- (ii) pH
- (iii) flowrate
- (iv) chloride and sulphate contents
- (v) temperature
- (vi) scale formation

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Mineral processing and extractive metallurgy

Modern plant equipment and practice in non-ferrous metallurgy is the broad theme of the joint international conference *Mineral Processing and Extractive Metallurgy*, organized by the Institution of Mining and Metallurgy and the Chinese Society of Metals, which is to be held in Kunming, Yunnan Province, People's Republic of China, from 14th to 20th October, 1984.

The Organizing Committee will welcome the submission of abstracts (200-300 words) of papers intended for publication in the preprinted volume of papers, which will be distributed to conference registrants in September 1984, and for presentation at the technical sessions.

Abstracts, in *English*, should be sent to the Conference Office, Institution of Mining and Metallurgy, 44 Portland

Place, London W1N4BR, England, *not later than 10th September, 1983*. The completed texts of selected papers will be required *not later than 10th February, 1984*.

Papers accepted for publication and presentation will be divided equally between mineral processing and extractive metallurgical subjects. It is envisaged that plenary and simultaneous sessions will be held. The Organizing Committee will welcome, in particular, the submission of abstracts on such topics as the problems that arise in the implementation of changes in practice, techniques, etc., in existing plants, innovative equipment and technology, pyro- and hydrometallurgy, energy and environmental factors, and the treatment of complex ores.