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MICROBIAL ECOLOGY OF RUM JUNGLE III  
LEACHING BEHAVIOUR OF SULPHIDIC WASTE MATERIAL  
UNDER CONTROLLED CONDITIONS

by

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December 1981

ISBN 0 642 59725 1

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+ Work carried out under AAEC Contract No. 74/F/40, in the School of Biotechnology, University of New South Wales. The University's Supervisor for this contract was Professor B.J. Ralph. Other work under this contract is reported in Microbial Ecology of Rum Jungle I and II, [Goodman, Khalid and Ralph 1981a,b].

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#### ABSTRACT

The discharge, into river systems, of acid and heavy metals generated by leaching of sulphidic waste materials at the abandoned opencut uranium mine at Rum Jungle, Northern Territory, is causing continuing pollution of the surrounding environment. The maximum effects of acid and microorganisms on samples from the overburden dump material, under defined and controlled environmental conditions, were assessed using reactor systems. These samples came from the overburden dump resulting from the mining of White's orebody. Similarly, the stability of tailings material under conditions of flooding and increasing acidity was determined.

At pH 2.5, metals in White's dump material were solubilised by acid attack only, whereas at pH 3.5, bacterial activity (principally that of Thiobacillus ferrooxidans) generated acidity and contributed significantly to metal release. At pH 3.5 (the average pH of soil in the dump) naturally occurring bacterial populations have the capacity to generate a maximum of  $40 \text{ kg SO}_4^{2-} \text{ tonne}^{-1}$  ore. Under microaerophilic conditions Thiobacillus ferrooxidans continued to effect metal release from the ore, but did not produce further acidity. If White's overburden is returned to the acidic, flooded opencuts, complete solubilisation of the material will occur. The exclusion of oxygen from the dump will not necessarily stop bacterially catalysed leaching processes.

Under highly aerated and agitated flooded conditions the tailings material was not active, except for copper release of about  $2 \text{ g kg}^{-1}$  ore at pH 4.0. The only deleterious element released by increasing acidity was copper, which was 100 per cent solubilised at pH 2.5. Uranium was always less than  $3 \mu\text{g kg}^{-1}$  ore, and lead was detected only at pH 2.5. Indigenous leaching bacteria did not develop.

National Library of Australia card number and ISBN 0 642 59725 1

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LEACHING ; THIOBACILLUS FERROXIDANS; TAILINGS; RUM JUNGLE; PH VALUE; PYRITE;  
SULFATES; OXIDATION; COPPER COMPOUNDS; IRON COMPOUNDS; MANGANESE COMPOUNDS;  
MAGNESIUM COMPOUNDS; URANIUM COMPOUNDS

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## 1. INTRODUCTION

The release of acid and heavy metals from the pyrite-containing waste rock dumps at the abandoned uranium mine site at Rum Jungle, NT, has had, and is continuing to have, extremely deleterious effects on the surrounding environment [Davy 1975]. If the mechanisms of the generation of these pollutants are to be understood, the rate and manner of metal solubilisation and acid production in materials typically found in the dumps must be determined under well defined and controlled conditions. In particular, it is important to know the maximum amount of metal that can be solubilised and acid produced under optimum conditions, and the likelihood that these conditions occur in the field. It is also of considerable interest to determine the role of naturally occurring microorganisms in the oxidation process.

The aim of the present investigation was to determine some of these factors. Material typical of White's overburden dump, which is the largest dump at the mine site, was leached at pH 3.5 (close to the average pH found for soil samples taken at the dump) and at pH 2.5 (the generally accepted optimum pH for leaching mineral sulphides with Thiobacillus ferrooxidans [Razzell 1962; Razzell and Trussell 1963; Duncan and Teather 1966]). Leaching was carried out, both in systems inoculated with microbial populations and in sterile controls, to quantify the effect of microorganisms on the release of metals and acid from the samples. Strict control and monitoring of environmental conditions and meaningful quantitative measurements of acid production, soluble metal concentrations and bacterial populations were achieved by carrying out the study in reactor systems. These reactor systems and their contribution to this type of study, are described in more detail in Section 2.

It is generally assumed that successful rehabilitation of such mine waste rock dumps can be achieved by the exclusion of oxygen, since oxygen has been thought to be essential for the oxidation of sulphides and subsequent solubilisation of metals. It is, therefore, of considerable interest to note what changes, if any, occur in metal solubilisation rates and bacterial populations when the leaching system is subjected to anaerobic, or microaerophilic, conditions after being subjected to highly aerobic conditions. Such a change in atmospheric conditions was easily achieved in the reactor systems used in this study.

The present investigation was also aimed at studying the stability of material from the tailings dam at Rum Jungle under conditions which simulate flooding of this area. Of particular interest were the metal solubilisation rate, how this rate depended on pH, and whether the tailings material could reach these acidity levels under field conditions.

Total sulphur content of the ore was determined by means of a Leco Automatic Sulphur Determinator (USA). This analysis was performed at the CSIRO Institute of Earth Sciences, North Ryde, NSW.

## 2. REACTOR SYSTEMS

Since the 1950s, there has been a rapid development of a large range of increasingly sophisticated fermentation equipment (i.e. reactors with ancillary instrumentation), which can automatically control, measure and record culture conditions. This development has been a result of the increasing awareness of microbiologists that environmental factors must be controlled (and therefore rendered non-variable) if meaningful and reproducible data are to be obtained on the growth, metabolism and kinetics of microbial activities [MacLennan 1970]. Reactor systems are now accepted as one of the most powerful tools for the study of microbial processes under strict environmental control. Although the literature on the application of reactors in many microbiological fields (both in research and industry) is vast, the use of reactors in the fields of biodegradation of mineral sulphides has so far been very limited [Torma et al. 1972; Gormely et al. 1975, Khalid 1978; McElroy and Bruynesteyn 1978; Babij and Ralph 1978; Babij, Doble and Ralph 1980], but their potential in this area is becoming increasingly apparent.

The most commonly used laboratory technique in mineral leaching is that of the shake-flask. This method is relatively cheap, simple to operate and allows simultaneous examination of a large number of samples.

However, these advantages are outweighed by those of reactor assemblies which enable:

- (a) precise control and monitoring of physicochemical parameters (the most important of which are pH and dissolved oxygen);



- (b) sampling without disturbance to the system under investigation;
- (c) maintenance under strict aseptic conditions for long periods;
- (d) precise reproduction of experimental conditions;
- (e) simultaneous control and test experiments under the same conditions;
- (f) unchanged initial pulp density (because particle-free leachate samples are obtained from a large volume of suspended slurry);
- (g) reduction of sampling errors (because the large working volume allows a more representative sample of ore to be analysed);
- (h) experimentation under the optimum conditions for any microbial species; and
- (i) standardisation of experimental conditions and procedures, and optimisation of a variety of parameters.

Methods other than those using reactor systems, employed in the study of mineral leaching, have no provision for standardisation or optimisation of conditions. This may explain the large amount of conflicting data in the literature.

### 3. MATERIALS AND METHODS

#### 3.1 Sample Preparation

Material from the overburden dump that resulted from White's orebody was collected by hand-picking about 300 kg of rocks from the surface of the dump and from the exposed surfaces of trenches dug to depths of 4 metres in September 1976. This material was picked with no regard for pyrite content and in this sense represented an 'average' sample. The ore was then crushed in a 'jaw-crusher', mixed thoroughly, and approximately 10 kg ground into fine material using a 'Rocklabs' ring-grinder (model CH-1). Particles of -200 mesh size (but larger than -400 mesh size) were used in reactor leaching studies. Each reactor was charged with between 300 and 400 g dry weight of ore.

Material from the tailings dam was collected, in May 1979, by hand-digging a hole to the base of the fill and taking approximately 5 kg from a depth of 0.77 m; 320 g dry weight of material was used in each reactor.

### 3.2 Element Analyses

From the 10 kg of ore ground to -200 mesh, a further 0.5 kg was ground to -400 mesh for initial metal analysis. In accordance with the method of Bernas [1968] the ore was digested in Teflon vessels in the presence of aqua regia/hydrofluoric acid, and the solubilised material analysed by atomic absorption spectroscopy, using a Varian instrument with modular updating to the Mark V level of performance. The method was modified so that any precipitate formed during the aqua regia/hydrofluoric acid digestion was re-digested in concentrated  $\text{HNO}_3$  and added to the solubilised material to be analysed. Uranium concentrations in leach liquor and solid samples were determined by the liquid scintillation method of Babij, Madgwick and Ralph [1978].

### 3.3 Reactor Runs

#### 3.3.1 White's material

Two 10 L (8 L working volume), stirred reactors fitted with temperature, pH, aeration and other monitoring and control devices, were each charged with 320 g dry weight (DW) of material from White's dump. One reactor, the test reactor, was inoculated with a particular microbial flora (see below), while the other was run simultaneously as a sterile, abiotic control. One such reactor pair has been described by Babij, Doble and Ralph [1980] and one reactor is illustrated schematically in Figure 1.

The charged reactors were sterilised by passing Sterigas 27 (CIC Ltd) through the system for approximately 1 hour, then sealed and left overnight. After flushing with sterile air, the reactors were filled with sterile 9 K medium [Silverman and Lundgren 1959], without  $\text{FeSO}_4$ , and the pH adjusted to the pre-determined set-point of either pH 2.5 or 3.5. The aeration and stirring rates were set at  $800 \text{ mL min}^{-1}$  and 200 rpm respectively and initially the slurry was leached by acid only, at  $30^\circ\text{C}$ .

For the run at pH 3.5, 50 mL of sediment slurry from each of White's and Intermediate opencuts was used as a inoculum at day 5 (point I in Figures 3

and 4) in the test reactor. For the following run at pH 2.5, an aliquot (from a stock culture), containing approximately  $10^{10}$  cells of Thiobacillus ferrooxidans [Khalid 1978] was used as an inoculum in the test reactor at day 10 (point I in Figure 2). Samples of particle-free leachate were taken regularly for metal and microbiological analysis. Aerobic conditions were maintained for 30 days, by which time copper and iron release had ceased. The run at pH 2.5 was terminated at this point since 100 per cent of the copper had been leached in the test reactor (see Results Table 2). However, for the run at pH 3.5 the reactors were subjected to anaerobic, or microaerophilic, conditions by continuous gassing with 5% CO<sub>2</sub> in nitrogen (point A in Figures 3, 4, 5, 6 and 7).

### 3.3.2 Tailings material

The reactors were sterilised as described above but the tailings material was not. Sterile distilled water followed by 320 g DW of the tailings was added aseptically to the reactors (duplicate reactors were run). The pH, bacterial flora and metal release were monitored. After 48 days the air flow was discontinued, and the CO<sub>2</sub>/N<sub>2</sub> mixture used for 25 days, after which the system was made aerobic again for a further 27 days. When no further changes took place, the pH was lowered by steps of 0.5 units to a minimum of pH 2.5. Each pH setting was maintained for three days before sampling. The temperature was maintained at 35°C.

### 3.4 Microbiological Assessment

Bacterial numbers were estimated by colony counts on selective media [Goodman, Khalid and Ralph 1981a].

## 4. RESULTS

### 4.1 White's Overburden Material

The composition of the overburden material used in this study is given in Table 1. The results of leaching at pH 2.5 and 3.5 are contained in Table 2, and Tables 4 and 6 contain the results obtained under various conditions at pH 3.5.

At pH 2.5 (Table 2), the role of bacteria in the release of soluble metals seems to have been non-significant, since both the control and inoculated reactors showed similar amounts of iron and copper release at similar rates (Figure 2 and Table 3). No sulphur was oxidised to sulphate, as acidity was not generated.

At pH 3.5 (Table 4), microbial populations contributed to the release of iron, copper and manganese from the material. The inoculated sample released iron, copper, magnesium and manganese at rates initially about twice those of the control sample (Figure 3 and Table 5). The decrease in soluble iron in the inoculated material (after day 15) to negligible levels was probably caused by the precipitation of jarosites. Sulphur oxidation to sulphate in the inoculated reactor was calculated from the amount of 5N NaOH added (by automatic controller) to maintain the pH at 3.5. Sulphate production is shown in Figure 4 and the rates given in Table 5. No sulphate (i.e. acidity) was generated from the control ore. The total amount of sulphate generated from the inoculated material was 16.6 g, which implies a rate of about 40 kg  $\text{SO}_4^{2-}$  tonne<sup>-1</sup> material. Analysis of the initial sulphur (as sulphide) content of the overburden material (Table 1), indicates that the bacteria oxidised 54 per cent of the total sulphur in the initial sample (400 g DW). Also, 16.6 g  $\text{SO}_4^{2-}$  produced in 8 L corresponds to 2.1 g  $\text{SO}_4^{2-}$  L<sup>-1</sup> which is equivalent to 43 mg H<sup>+</sup> L<sup>-1</sup>. This amount of H<sup>+</sup> ion would have caused the pH to drop from 3.5 to 2.5 (allowing for the buffering capacity of the suspending 9K medium), if the H<sup>+</sup> produced had not been constantly neutralised.

At day 30, when the copper concentration in solution was stable, the inoculated reactor was subjected to an atmosphere of 5% CO<sub>2</sub> in N<sub>2</sub> for a further 20 days, thus generating anaerobic, or microaerophilic, conditions. Dissolved oxygen (D.O.) was below the detection limit (1 mg L<sup>-1</sup>). Copper, magnesium and manganese showed a significant further increase in leaching, but further acid was not generated (i.e. no sulphate was produced). The total release of metals into solution after aerobic and anaerobic conditions is shown in Table 6 and Figure 3. The approximate rates of metal release under anaerobic conditions were as follows: copper 0.2 mg L<sup>-1</sup> d<sup>-1</sup> for 6 days; magnesium 12 mg L<sup>-1</sup> d<sup>-1</sup> for 20 days and manganese 0.1 mg L<sup>-1</sup> d<sup>-1</sup> for 10 days. Dissolved iron precipitated and was not detected after 12 days.

Figures 5, 6 and 7 show the levels of microbial populations during aerobic and anaerobic leaching. Although the initial level of I. ferrooxidans was low (Figure 5), numbers of this organism increased a thousand-fold between

days 5 and 10, a further ten-fold by day 15, to a maximum level by day 20 (twice that at day 15). After day 20, numbers of viable cells in the slurry supernatant fluctuated but remained high during anaerobic conditions.

Acidophilic (i.e. low pH), sulphur-oxidisers (isolated at pH 3.5) showed an initial die-off, but then rapidly increased between days 15 and 20 and continued to increase more slowly until day 30 (Figure 6). These microorganisms rapidly lost viability under anaerobic conditions. Numbers of high pH sulphur-oxidisers (isolated at pH 6.2) were high initially and oscillated under aerobic conditions, whereas anaerobically their numbers remained fairly constant after an initial drop.

Intermediate pH sulphur-oxidisers (isolated at pH 4.8) increased ten-fold from the initial level and also fluctuated under aerobic conditions but numbers remained steady under anaerobic conditions (Figure 7). Acidophilic heterotroph numbers (isolated at pH 3.5), were generally low and increased only about ten-fold, with numbers fluctuating throughout the run (Figure 7).

#### 4.2 Tailings Stability

The content of uranium, copper and lead in the tailings material is given in Table 7 [Roberts 1974]. The leaching experiment was carried out in paired reactors over a period of 100 days. The maximum amounts of metals released and sulphate produced are given in Table 8. In the slurry, iron was generally below the detection limit ( $1 \text{ mg L}^{-1}$ ), rose slowly to a maximum concentration of about  $3 \text{ mg L}^{-1}$  from day 14 to 44, and then precipitated. Copper was released at a rate of about  $1 \text{ mg L}^{-1} \text{ d}^{-1}$  for the first 21 days and the final concentration remained at about  $80 \text{ mg L}^{-1}$  (initial solubilisation gave a concentration of  $60 \text{ mg L}^{-1}$ ). Lead was below detection level in the slurry throughout. Magnesium concentration remained constant, after initial solubilisation, at  $60 \text{ mg L}^{-1}$ . The uranium concentration remained at  $1\text{-}5 \text{ } \mu\text{g L}^{-1}$ . The pH of the slurry, initially 4.5, dropped to 4.2 within the first 10 days, remaining at this level for a further 30 days, after which it slowly settled to a constant 4.0. This drop in pH corresponded to the production of about  $3 \text{ mg L}^{-1}$  of sulphate.

After 100 days, the run was extended to examine the effect on metal release of lowering the pH by steps of 0.5. Table 9 gives the cumulative metal release at each pH setting. Iron and magnesium showed only slight increases above the stable pH 4.0 levels. However, 100 per cent of the copper

present solubilised rapidly at pH 2.5 (compare Table 7, the concentration of copper in the ore, with Table 9, the amount of copper solubilised at pH 2.5). Lead concentration of less than  $1 \text{ mg L}^{-1}$  was detected in the slurry at pH 2.5 only. Uranium solubility was not affected by lowering the pH and maintained a constant slurry concentration of about  $3 \text{ } \mu\text{g L}^{-1}$ . The cumulative amounts of  $\text{H}^+$  necessary to achieve the pH indicated, were  $10 \text{ mg kg}^{-1}$  for pH 3.5,  $160 \text{ mg kg}^{-1}$  for pH 3.0 and  $560 \text{ mg kg}^{-1}$  for pH 2.5.

At the end of the experiment, the slurry was maintained at pH 2.5 for a further 10 days to see if natural I. ferrooxidans populations would develop. However, no viable cells were found.

## 5. DISCUSSION

### 5.1 White's Material

Only I. ferrooxidans was used as an inoculum at pH 2.5 as it had been predominantly responsible for leaching the mineral material at pH 3.5. Water and sediment from both White's and Intermediate opencuts were used as an inoculum in the test reactor at pH 3.5 to simulate leaching by the natural microbial flora. Microbial populations were not built up from the overburden material itself probably because it had been stored for too long (two years), and also because crushing and sieving of the material had not been done under sterile conditions. Because the pH of many of the samples from White's dump (Appendix C, Goodman, Khalid and Ralph 1981a), which had been collected over a number of years, was close to 3.5, this pH value was chosen for initial study.

The rate of bacterial leaching at pH 2.5 was masked by the rate of acid leaching (Table 3). Also, total metal release from the inoculated sample was only 4, 1.5 and 5 per cent greater than from the control sample for copper, iron and manganese respectively (calculated from data in Table 2).

However, at pH 3.5 the contribution of bacterial activity to leaching was probably significant, the initial rates of metal release from the inoculated sample being about twice the rates from the control sample (Table 5). Also, total metal release from the inoculated material was 13, 95 and 25 per cent greater than from the control sample for copper, iron and manganese respectively (calculated from data in Table 4). Bacterial action effected further metal release from the sample under anaerobic conditions.

In the control sample, at pH 3.5 no sulphur was oxidised to sulphate, so that the pH remained constant. Therefore, under the abiotic conditions of this experiment, no chemical oxidation of the sulphidic (pyritic) components of the rock sample contributed to metal release. However, in the inoculated sample, bacterial action was responsible for the oxidation of about 54 per cent of sulphur to sulphate (Table 5). This corresponds to the generation, by microbes, of about 40 kg  $\text{SO}_4^{2-}$  tonne<sup>-1</sup> ore at pH 3.5. This may be compared to the estimated rate of sulphate release from the dump of about 0.7 kg tonne<sup>-1</sup>y<sup>-1</sup>. The enhanced sulphuric acid production in the reactor resulted from the fact that conditions for microbial growth were optimised which is not necessarily the case in the field situation. It should be emphasised that the reactor system used in the laboratory study was not intended to simulate conditions in the overburden dump.

Although the percentage of total metal release was higher at pH 2.5 than at pH 3.5 (compare Tables 2, 4 and 6) and only about 50 per cent of copper was extractable at pH 3.5, acid was generated by bacterial action, aerobically, only at pH 3.5. During this experiment, this acid was constantly neutralised (by automatic  $\text{OH}^-$  addition) and the pH was maintained at 3.5. If this acid had not been neutralised, then acid solubilisation of the sample would have also contributed to metal release, as the amount of  $\text{H}^+$  generated would have lowered the pH from 3.5 to 2.5.

It is significant that the period of fastest metal release and sulphate production coincided with the occurrence of rapid growth of I. ferrooxidans. The fastest increase of low pH sulphur-oxidising microorganisms, probably I. thiooxidans type, also corresponded with the fastest rate of sulphate production. I. thiooxidans may have been partly responsible for the production of sulphate, as well as I. ferrooxidans, which simultaneously oxidises insoluble ferrous iron and sulphide during growth on chalcopyrite or pyrite [Duncan, Landesman and Walden 1967]. However, during this period, levels of this organism were always lower, by at least a hundredfold, than those of I. ferrooxidans. In this experiment, I. ferrooxidans was probably the organism predominantly responsible for metal release, even though I. thiooxidans has been shown to be somewhat active in the release of minerals from sulphide ores [Karavaiko and Moshniakova 1974; Khalid and Ralph 1977; Khalid 1978].

Under anaerobic conditions, I. ferrooxidans numbers in solution maintained the level reached aerobically. Pugh and Umbreit [1966] have shown

I. ferrooxidans capable of fixing CO<sub>2</sub> anaerobically, with soluble ferrous iron as the energy substrate. These authors postulate the involvement of an in vivo electron carrying system. Brock and Gustafson [1976] have shown that, under anaerobic conditions, I. ferrooxidans can oxidise elemental sulphur, with a concurrent reduction of soluble ferric to ferrous iron. Mackintosh [1978] grew I. ferrooxidans colonies microaerophilically, with ferric iron precipitating in the colonies. The precise work of Kelly and Jones [1978] on the iron oxidation kinetics of I. ferrooxidans and their demonstration of its capacity to develop in a milieu of low carbon dioxide and oxygen are highly relevant to the observed behaviour of the White's overburden material under microaerophilic conditions. During the present experiment, the further release of copper, magnesium and manganese under anaerobic conditions was most probably caused by I. ferrooxidans. The contribution to metal release under anaerobic conditions by I. thiooxidans was probably negligible because viability was rapidly lost.

Except for an initial die-off by I. thiooxidans, the fluctuations in numbers of microorganisms, after initial growth levels in the supernatant were reached, were possibly caused by attachment of the organisms to the solid particles. Numbers of I. ferrooxidans, and possibly those of I. thiooxidans, were most probably higher than estimated, as these organisms firmly attach themselves to solid substrates [Baldensperger, Guarraia and Humphreys 1974; Berry and Murr 1978], and the methods employed in this experiment accounted for numbers of viable cells in the supernatant only.

It is interesting that numbers of sulphur-oxidising organisms (isolated at pH 6.2 and 4.8) and acidophilic heterotrophs remained relatively high throughout the experiment. The sulphur-oxidisers probably include I. denitrificans type, which may grow either aerobically or anaerobically [Buchanan and Gibbons 1974] and this organism has been found in most sediment samples from the Intermediate opencut [Goodman, Khalid and Ralph 1981b]. Little work on the role of such microorganisms in mineral leaching has been done [Tuovinen and Kelly 1974; Khalid and Ralph 1977; Le Roux, Wakerley and Perry 1978; Norris and Kelly 1978]. Even less work has been undertaken on the interrelationships between such microorganisms in mixed populations [Tsuchiya, Trivedi and Schuler 1974; Trivedi and Tsuchiya 1975]. As these types of microorganisms are abundant in mine environments [Goodman, Khalid and Ralph 1981a,b] an understanding of their activities in such environments is greatly needed.



## 5.2 Stability of Tailings Material

The tailings material was found to be inactive, except for copper release. Its dispersion in a highly aerated phase gave rise to neither the growth of indigenous leaching bacteria nor the production of acidity. Magnesium was released at a concentration of about  $1.5 \text{ g kg}^{-1}$  ore; the amount of soluble iron (because of the relatively high pH) was virtually negligible and copper, at a concentration of about  $2 \text{ g kg}^{-1}$  ore, proved to be the only deleterious element released. The concentration of uranium was always less than  $10^{-4} \text{ g kg}^{-1}$  ore. Note that mixing the tailings material in distilled water produced a soluble copper concentration of about  $2000 \text{ mg L}^{-1}$  (i.e.  $\text{mg kg}^{-1}$  ore). This result agrees with the concentration of soluble copper found in samples taken from several points (and at various depths) of the tailings dam area for microbiological analysis. However, this result is much higher than that quoted in Davy [1975] for copper release from tailings material suspended in water; soluble copper was found to have a concentration of about  $14 \text{ mg kg}^{-1}$  ore. The analysis in Davy [1975], however, was made on tailings material which had been washed from the main tailings dam area during a period of "intense rainfall" and deposited further down in the Old Tailings Creek. It is highly probable that most of the soluble copper content in this material had already been washed out by intense rain and therefore, it was not, as Davy [1975] has asserted, representative of the material in the main area of the tailings dam.

During the deliberate lowering of pH, copper was the only metal to show significant release, about 50 per cent at pH 3.5 and 100 per cent at pH 2.5 (compare Tables 9 and 7). Uranium and lead were not affected by lowering the pH. Note that the drop in pH necessary to release 100 per cent of copper (i.e. from pH 4.0 to 2.5) required the addition of the equivalent of  $0.56 \text{ kg H}^+ \text{ tonne}^{-1}$  tailings. The results demonstrate that it is unlikely that this amount of acidity can be generated from the tailings by either chemical or indigenous bacterial oxidation.

## 6. CONCLUSIONS

The pH of many soil samples taken from White's overburden dump is around pH 3.5. At this pH, bacterial activity can generate about  $40 \text{ kg SO}_4^{2-} \text{ tonne}^{-1}$  ore. The material used in this study was picked from the surface of the dump and from trenches dug to about 4 metres deep. This material had probably

already undergone leaching and, if material deeper within the heap had been exposed, the possible generation of sulphate by bacterial action would probably have been higher.

If White's dump material were to be returned to either White's or Intermediate opencuts at their existing average pH of 2.4 and 3.2 respectively, the material would completely leach by acid or bacterial solubilisation, or a combination of both.

The anaerobic leaching results show that, even if White's dump was made anaerobic, (e.g. by covering the topsoil and re-vegetating), bacterial leaching of sulphidic material would probably continue as long as water activity and CO<sub>2</sub> levels remain sufficiently high for bacterial growth. Note that, in White's dump, low oxygen tension does not imply low carbon dioxide tension because of the association of carbonate gangue (e.g. dolomite) with sulphidic minerals in the dumps. Therefore, CO<sub>2</sub> would be available to bacteria by solubilisation of soil material [Brierley 1978]. Carbon dioxide is essential for the growth of I. ferrooxidans on ferrous iron or solid sulphides, since it is the sole carbon source for the microorganism [Schnaitman, Korczynski and Lundgren 1969]. However, oxygen is not essential for the growth of I. ferrooxidans on ferrous iron, elemental sulphur or solid sulphides [Pugh and Umbreit 1966; Baker and Wiltshire 1970; Brock and Gustafson 1976; Mackintosh 1978]. Concentrations of CO<sub>2</sub> higher than the normal atmospheric concentration (i.e. 0.03 per cent) stimulated metal release from solid sulphides by I. ferrooxidans [Torma et al. 1972].

It is important that similar studies be carried out on other solid wastes at the Rum Jungle mine, e.g. Intermediate dump and the sulphide heap, to determine their 'ceiling' of degradation. The reactor system may be used to characterise a particular ore by its leaching behaviour - by bacterial, acid or chemical action, and to determine the upper limits of its degradation rates under varying conditions. Such data could be used to predict the long term pollution problem of waste ore bodies, or the fate of materials dumped in opencuts.

Study of the tailings dam material has demonstrated that flooding of the dam will neither generate acidity nor encourage the growth of leaching bacteria. The high levels of copper, even at pH 4, will require attention. Uranium was released at a concentration of  $< 10^{-1}$  g tonne<sup>-1</sup> and thus is not a major pollutant.

## 7. ACKNOWLEDGEMENTS

One of the authors (AG) wishes to thank staff of the AAEC, in particular Drs A.I.M. Ritchie and R.T. Lawson, for their helpful advice and time spent in many fruitful discussions of this manuscript while she was employed at the AAEC Research Establishment.

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TABLE 1WHITE'S OVERBURDEN MATERIAL

Element	Concentration mg kg <sup>-1</sup>
Cu	520
Fe	46 000
Mg	28 800
Mn	580
Pb	160
Zn	180
S	25 700 ± 5000 <sup>†</sup>

† This is the mean of three measurements, together with the appropriate error range. Measurements of the soluble metals, on the other hand, were made by standard AAS technique where each measurement is made only once.

TABLE 2LEACHING AT pH 2.5

Element	% Released After 30 Days	
	Inoculated	Control
Cu	100	96.1
Fe	14.3	14.1
Mg	99.8	92.0
Mn	18.5	17.6
Zn	3.8	3.6
<sup>†</sup> S	0	0

<sup>†</sup>calculated from  $\text{SO}_4^{2-}$  produced



TABLE 3APPROXIMATE RATES OF METAL RELEASE AT pH 2.5

Metal	Inoculated		Control	
	mg L <sup>-1</sup> d <sup>-1</sup>	Time, d	mg L <sup>-1</sup> d <sup>-1</sup>	Time, d
Cu	0.5	5-20	0.7	5-20
Fe	5	10-15	3	5-15
	24	15-25	24	15-30
Mg	35	5-30	47	5-15
			10	15-30

TABLE 4LEACHING AT pH 3.5AEROBIC CONDITIONS

Element	% Released after 30 Days	
	Inoculated	Control
Cu	46.0	40.1
Fe	2.3	0.1
Mg	48.8	43.9
Mn	19.2	14.5
S	53.7	0

TABLE 5

APPROXIMATE RATES OF METAL AND  $\text{SO}_4^{2-}$  RELEASE AT pH 3.5

AEROBIC CONDITIONS

Element	Inoculated		Control	
	mg L <sup>-1</sup> d <sup>-1</sup>	Time, d	mg L <sup>-1</sup> d <sup>-1</sup>	Time, d
Cu	0.6	5-15	0.3	5-30
Fe	10	10-15	ND <sup>†</sup>	5-20
	0.5	25-30	0.5	20-30
Mg	45	5-10	25	5-15
	8	10-30	3	15-30
Mn	0.2	5-20	0.1	5-20
S (as $\text{SO}_4^{2-}$ )	40	10-15	ND <sup>†</sup>	
	340	15-20		
	16	20-30		

<sup>†</sup> not detected

TABLE 6LEACHING AT pH 3.5AEROBIC FOLLOWED BY MICROAEROPHILIC CONDITIONS

Element	% Released After 50 Days	
	Inoculated	Control
Cu	50.4	40.1
Fe	ND <sup>†</sup>	0.7
Mg	67.1	43.9
Mn	22.4	14.5
S	53.7	0

<sup>†</sup> Not detected because of precipitation

TABLE 7TAILINGS MATERIAL ANALYSIS

Element	Concentration
	mg kg <sup>-1</sup>
U	430
Cu	5400
Pb	830

(Roberts 1974)

TABLE 8TAILINGS STABILITY pH 4.5-4.0

Element	Amount Released After 100 Days	
	mg kg <sup>-1</sup>	%
Cu	2125	40
Mg	1500	
Fe	62.5	
Pb	ND <sup>†</sup>	ND <sup>†</sup>
U	< 0.1	< 0.02
S (as SO <sub>4</sub> <sup>2-</sup> )	82.5	

<sup>†</sup>Not detected

TABLE 9RELEASE OF METALS FROM TAILINGS BY LOWERING pH

Metal	Amount Released		
	mg kg <sup>-1</sup>		
	pH 3.5	pH 3.0	pH 2.5
Cu	2625	2950	5500
Mg	1950	2250	2500
Fe	ND <sup>†</sup>	ND	125
Pb	ND	ND	< 25
U	< 0.1	< 0.1	< 0.1

<sup>†</sup> Not detected

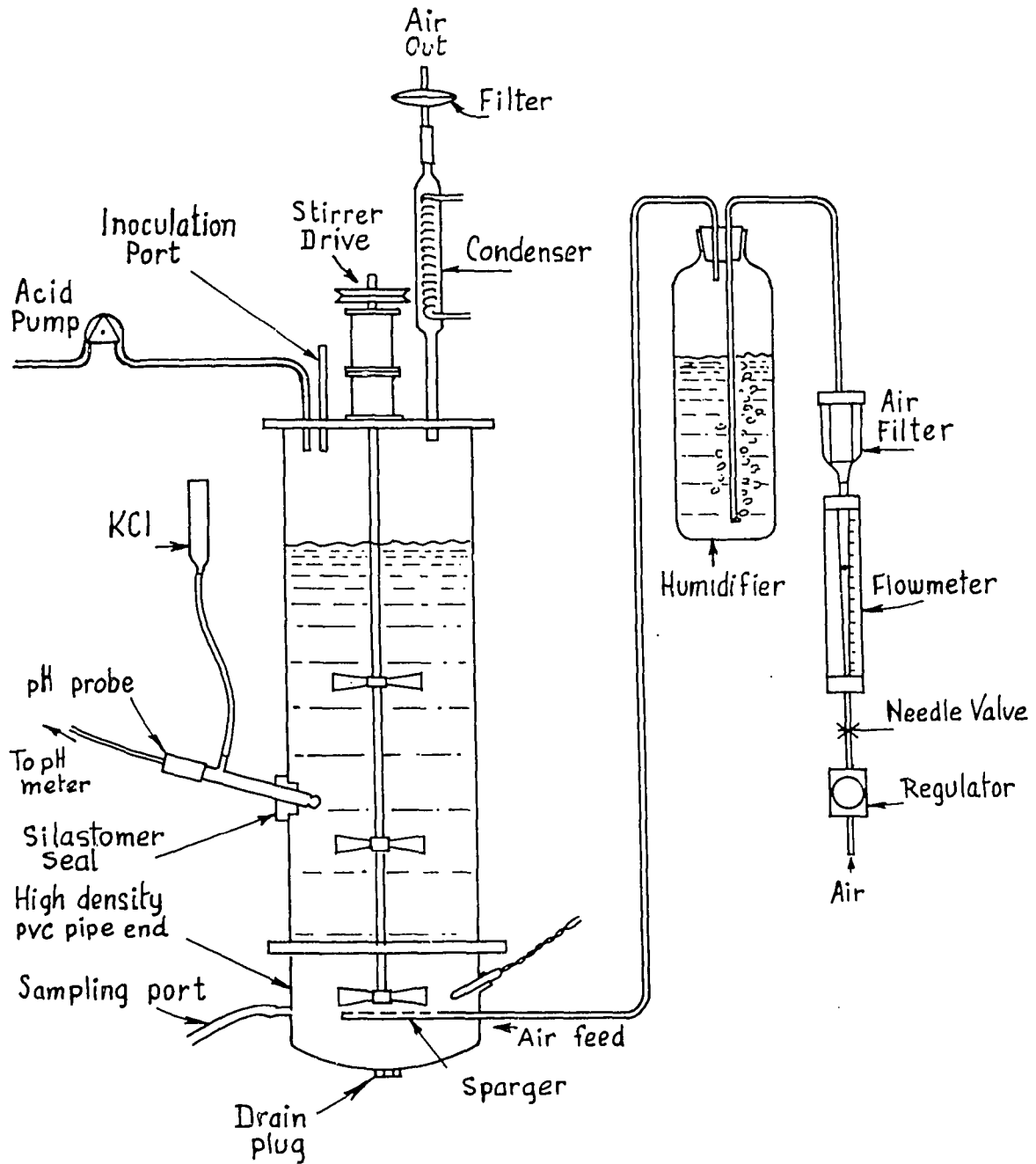


FIGURE 1. SCHEMATIC DIAGRAM OF REACTOR SYSTEM



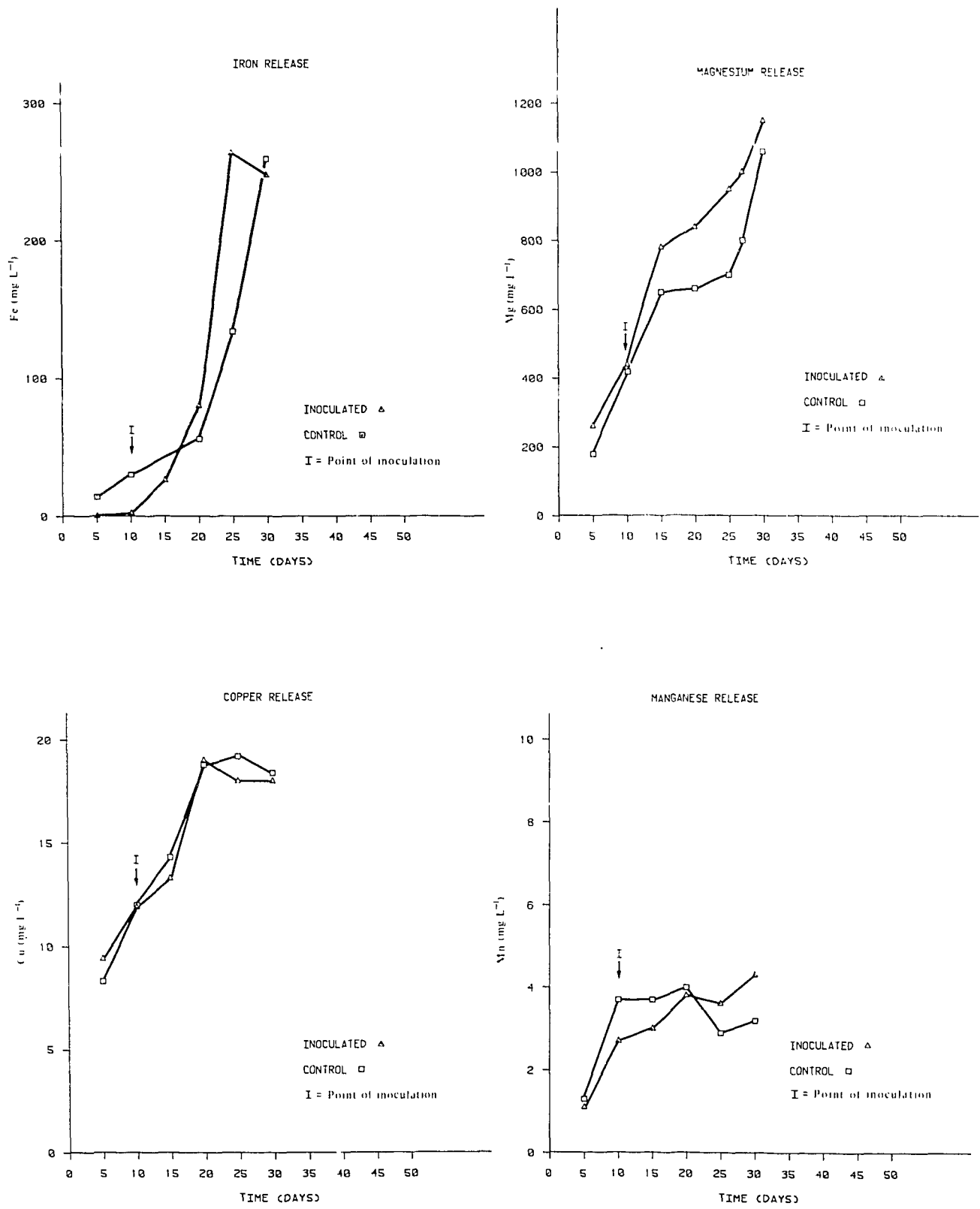


FIGURE 2. METAL RELEASED FROM A SAMPLE OF WHITE'S OVERBURDEN DUMP LEACHED AT pH 2.5

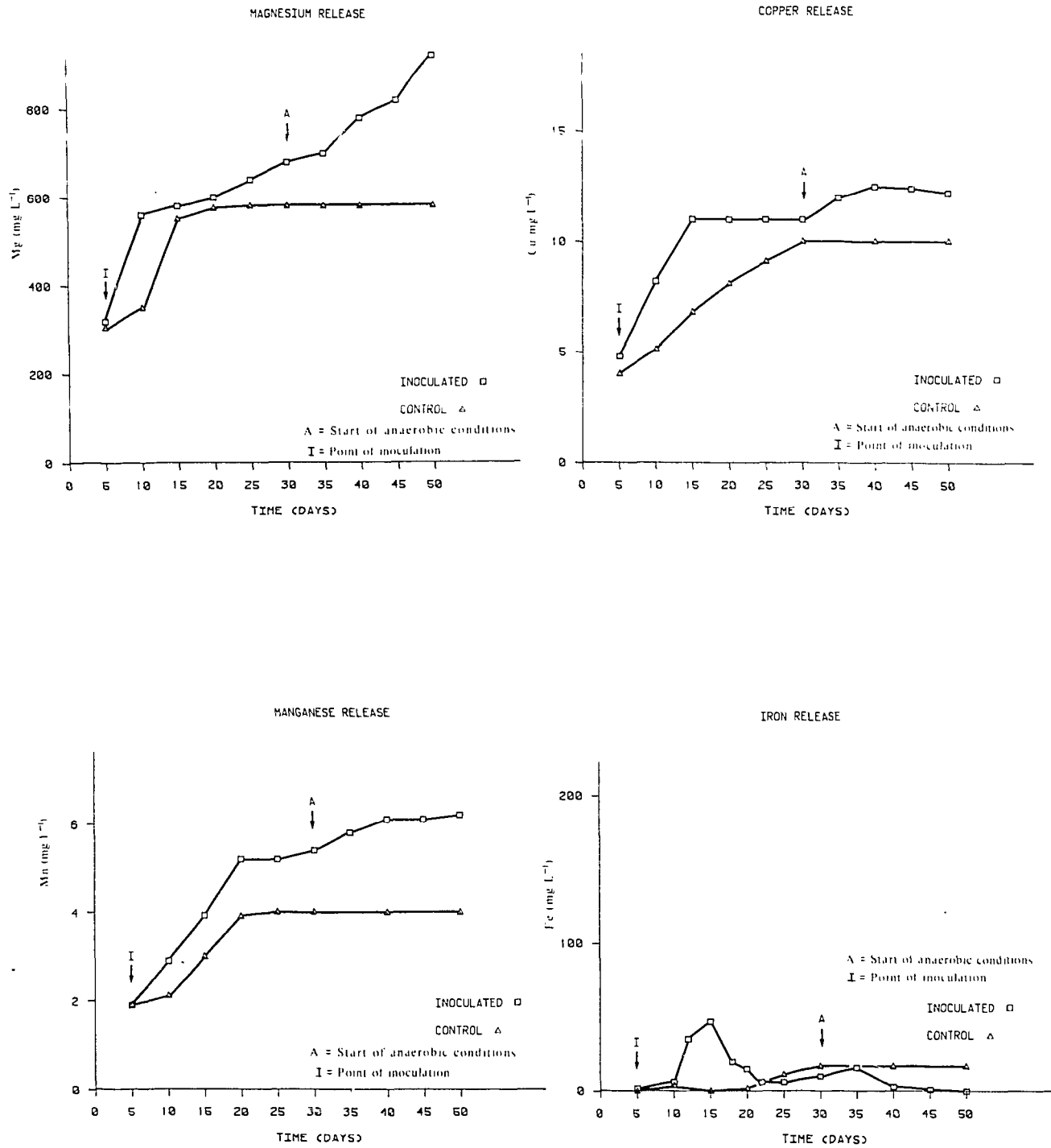


FIGURE 3. METAL RELEASED FROM A SAMPLE OF WHITE'S OVERBURDEN DUMP LEACHED AT pH 3.5

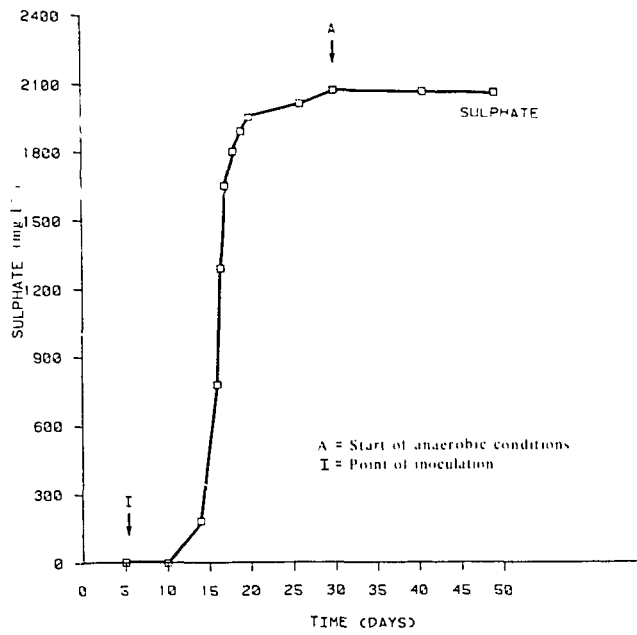


FIGURE 4.

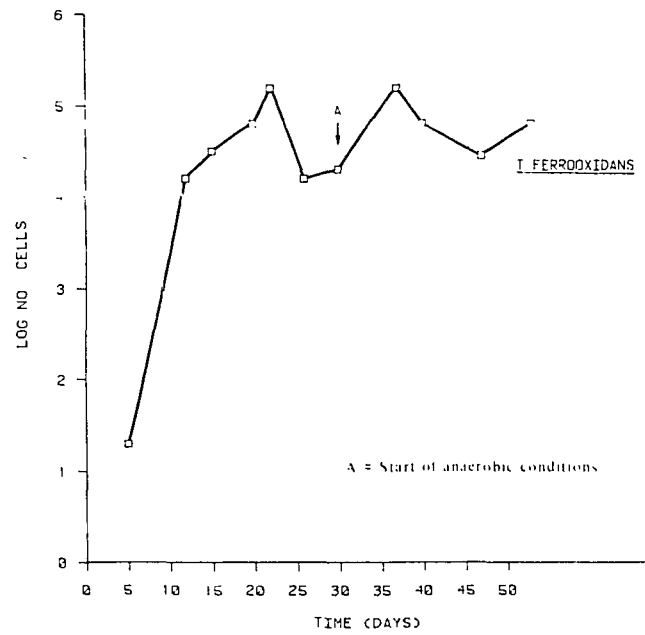


FIGURE 5.

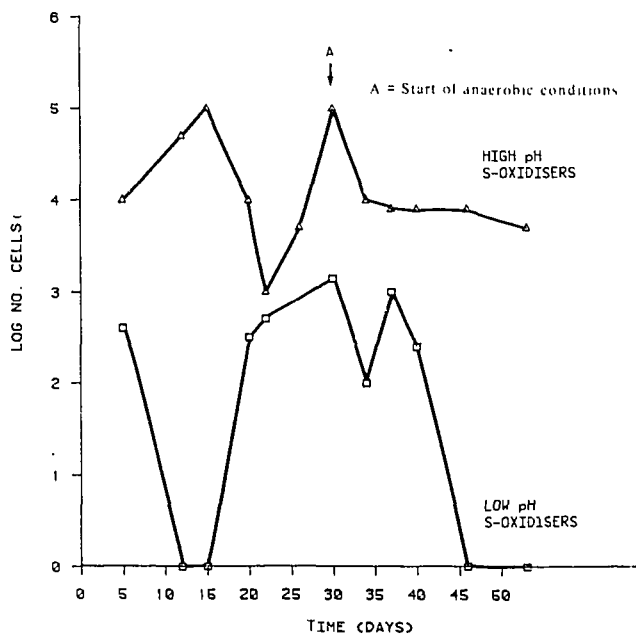


FIGURE 6.

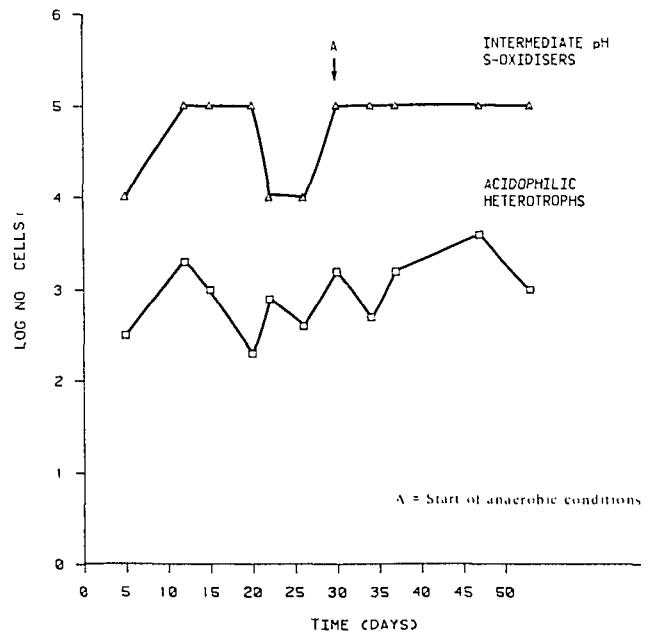


FIGURE 7.

FIGURES 4-7. SULPHATE AND BACTERIAL LEVELS UNDER AEROBIC AND MICROAEROPHILIC CONDITIONS IN AN INOCULATED REACTOR CONTAINING A SAMPLE OF WHITE'S OVERBURDEN DUMP; pH 3.5