

A dynamic uranium-leaching model for process-control studies*

by D.A. VETTER†, I.J. BARKER†, and G.A. TURNER†

SYNOPSIS

The modelling of the uranium-leaching process, and the logging of data from a plant for the evaluation of the model, are reported.

A phenomenological approach was adopted in the development of the model. A set of eight chemical reactions was chosen to represent the complex chemistry of the process, and kinetic expressions for these reactions were incorporated in differential equations representing mass and energy balances. These equations were coded in FORTRAN to form a program that simulated the process, and that allowed averaged and continuous data from the plant to be compared with the model. This allowed the model to be 'tuned', and to reveal a number of minor problems with the control infrastructure on the plant.

SAMEVATTING

Daar word verslag gedoen oor die modellering van die uraanloogproses en die registrering van data afkomstig van 'n aanleg met die oog op die evaluering van die model.

Daar is 'n fenomenologiese benadering by die ontwikkeling van die model gevolg. 'n Stel van agt chemiese reaksies is gekies om die ingewikkelde chemie van die proses te verteenwoordig, en kinetiese uitdrukkings vir hierdie reaksies is ingesluit by verskillende differensiaalvergelykings wat massa- en energiebalanse voorstel. Hierdie vergelyking is in FORTRAN gekodeer om 'n program te vorm wat die proses naboots en dit moontlik maak om deurlopende data en data waarvan die gemiddeld bereken is, wat van die aanleg verkry is, met die model te vergelyk. Hierdeur was dit moontlik om die model 'in te stem' en 'n paar minder belangrike probleme met die beheerinfrastruktuur by die aanleg uit te wys.

Introduction

In an effort to improve metallurgical efficiency and process operability, many uranium-extraction facilities have installed computer-based systems for the monitoring of instruments, basic stabilizing control, and data logging and reporting. Although the benefits of such systems are well known, industry as a whole has realized that it is necessary to include on-line optimization and comprehensive control systems in the control infrastructure¹. Classical single input-single output control systems are severely limited in that they do not compensate for interactions between the controllers and are unable to indicate the optimum process conditions for a given set of process inputs.

In the past, effort was devoted to the development of suitable process sensors for the uranium-leaching process². Once that measurement infrastructure had been established, it became possible to address the question of how a comprehensive control system for uranium leaching could be developed. The objective of that work was a detailed and methodical study of the control of a uranium-leaching plant. Experience gained in other control projects at Mintek has shown that such an investigation usually involves the following stages:

- (a) dynamic characterization of the process in the form of a mathematical model,
- (b) the capture of plant data,

- (c) the testing and development of models by the use of plant data,
- (d) the designing of a controller, and
- (e) the development of a prototype controller.

The mathematical modelling and the logging of plant data reported in this paper represent stages (a), (b) and, to some extent (c), in a generalized study of process control for uranium leaching. Unfortunately, while the study was in progress, the market for uranium declined and, with it, the support for the present work. As a result, further testing and fitting of the model had to be suspended, and the design and development of the prototype controller had to be shelved. Nevertheless, even the limited study that was carried out revealed a number of points that warrant further attention and, should a demand arise for such a study in the future, work could be resumed.

The uranium-leaching model described was developed specifically for purposes of process control, and was not intended to be a comprehensive model, but rather one that described the main dynamic effects of the process so that it could be used in the evaluation of a suitable control system.

The Process Model

A simplified layout of a uranium-leaching plant is shown in Fig. 1. The uranium-leaching model described in this paper is essentially phenomenological in that it describes the process in terms of equations that express the conservation of mass and energy in the system. The nomenclature used for the model is detailed at the end of the paper.

It was decided to develop a phenomenological model

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† Mintek, Private Bag X3015, Randburg, 2125 Transvaal.

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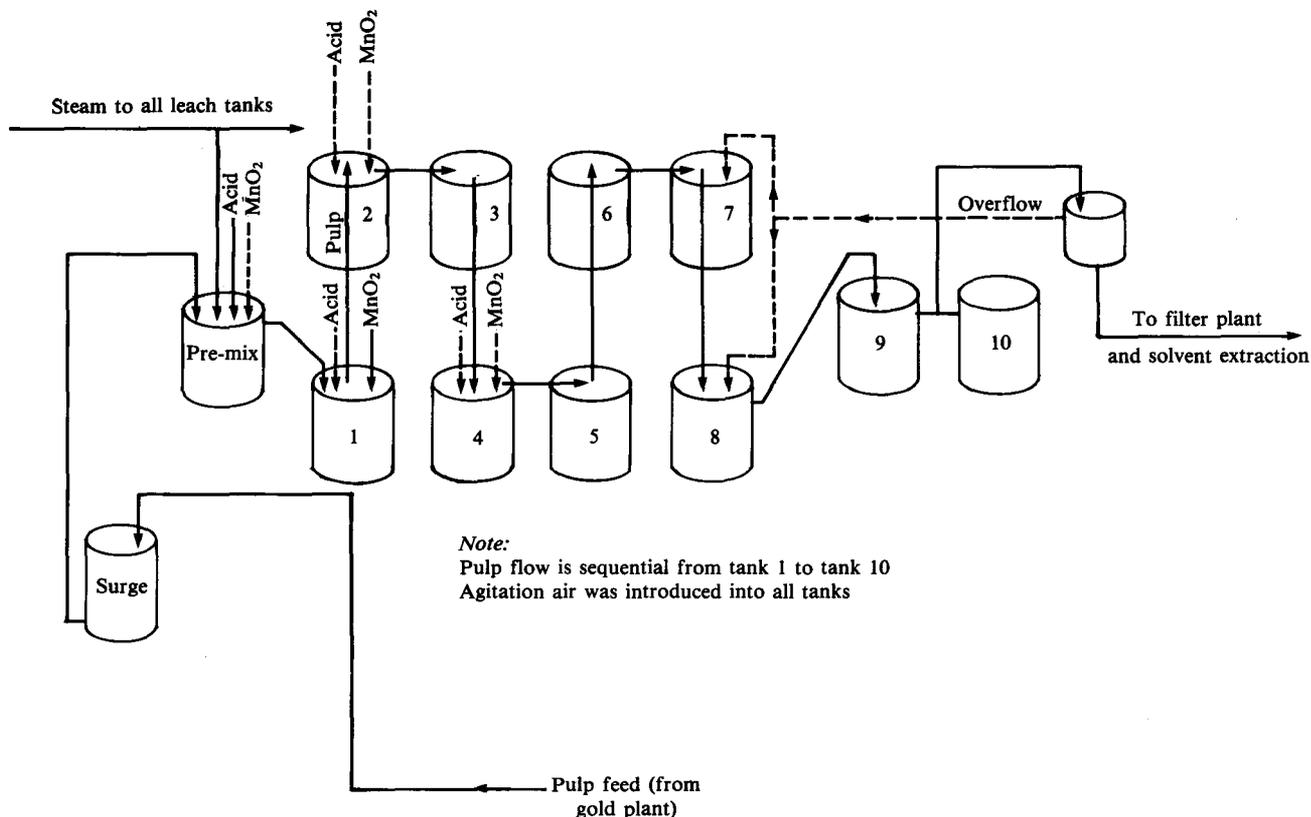


Fig. 1—A typical uranium-leaching cascade

rather than a fundamental mechanistic process model because the latter type of model is extremely difficult to formulate. It contains too many parameters that cannot be determined from process measurements, and the resulting system of differential equations is difficult to solve. A phenomenological model allows most of the important features of the extraction process to be described dynamically, which makes it ideal for use in process-control studies. (In this application, models do not need to be detailed; they need to describe the main dynamic effects adequately.)

For the formation of the leaching model, it was assumed that the reactions take place in only one phase, that the process reactions can be expressed by a fixed scheme of reactions with a given stoichiometry, and that the kinetics of these reactions can be described by rate expressions of a classical form. The individual tanks are idealized as continuously stirred tank reactors configured into a cascade, the ore is taken as being milled to a size at which all the species are liberated, and the leaching tanks are assumed to be effectively agitated throughout.

The mass and energy balances for a typical leaching tank are best interpreted in conjunction with an idealized leaching stage (Fig. 2). Concentration is expressed in terms of the liquid phase of the components. The terms used can be converted to express the concentrations of the solid phase simply by multiplication of the terms by the solid-to-liquid ratio for a given leaching stage. The mass balance for a continuously stirred tank reactor is consequently expressed as

$$V_j \frac{dc_{ij}}{dt} = F_{j-1}c_{i,j-1} - F_j c_{ij} + F_j^{acid} c_j^{acid} + F_j^{MnO_2} c_j^{MnO_2} \\ [\pm] R_{ij}(c_{ij}, T_j), i = 1, \dots, NS, j = 1, \dots, NT, \dots (1)$$

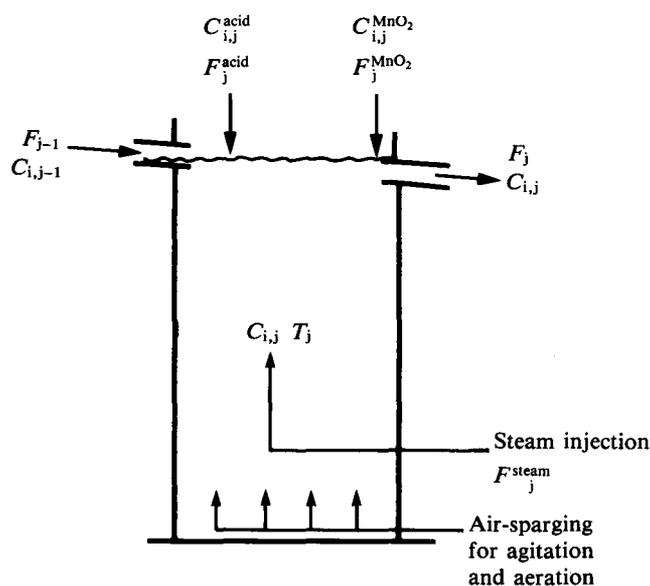


Fig. 2—A diagrammatic representation of a typical leaching tank showing all flows

where
i is the species
j is the tank.

The $V_j \frac{dc_{ij}}{dt}$ represents the accumulation term and the terms on the right-hand side of the equation represent, from left to right, the mass flow of pulp into the tank, the mass flow of pulp out of the tank, the reagent acid stream entering the tank, the reagent pyrolusite stream entering the tank, and a rate expression (which is

expressed as a function of the set of all concentrations c_{ij} in the tank, and the temperature). This describes the chemical change taking place, and is either positive or negative, depending on whether a species is the product or the reactant.

The heat balance over a given leaching stage is expressed as

$$C_p V_j \frac{dT_j}{dt} = \rho_j F_{j-1} C_p (T_{j-1} - T_j) + \rho^{\text{MnO}_2} F^{\text{MnO}_2} C_p^{\text{MnO}_2} (T_a - T_j) + \rho^{\text{steam}} F^{\text{steam}} C_p^{\text{steam}} (T_s - 100) + \lambda^{\text{water}} F^{\text{steam}} + \rho^{\text{water}} F^{\text{steam}} C_p^{\text{water}} (100 - T_j) + L_j (T_j - T_a),$$

$j = 1, \dots, NT. \dots\dots\dots (2)$

In equation (2), the heat of the reactions and that of the acid dilution have been ignored since these are small compared with the total amount of heat involved.

It was assumed that all the heat-loss terms for a given leaching tank could be lumped together into one loss term, L_j , which represents the heat lost by radiation, convection, and evaporation. It was assumed that all the steam entering a leaching tank condenses, and that this heat is transferred to the leach pulp.

The full set of equations represented by equations (1) and (2) are solved numerically by use of the software model. The initial conditions are pre-specified by the user and, as integration proceeds, the process inputs that describe the changing flow conditions or the set points of the controller, or both, can be read into the model so that it dynamically simulates the evolution of the actual process. The simulator is coded in FORTRAN, and includes standard proportional-integral-derivative (PID) controllers for the addition of acid and pyrolusite, as well as control of the tank temperature.

The numerical integration is carried out by use of a standard fourth-order Adams-Moulton predictor-corrector pair with a fourth-order Runge-Kutta starting method. The integration is stable and acceptably accurate when the step size of the integration is smaller than the shortest reaction time constant; for this system, integration steps

of 60 seconds or less were found to result in acceptable integration characteristics.

Modelling of the Leaching Chemistry

The use of sulphuric acid to dissolve uranium is common practice. Since most uranium minerals can be leached only when they have been oxidized, oxidizing conditions are maintained by ferric ions, which are generated by the oxidation of the ferrous ions present in solution^{3,4}. This reaction is achieved by the addition of pyrolusite (essentially MnO_2) as the oxidizing agent to the leach pulp.

The ore upon which the model is based was derived from a typical Witwatersrand conglomerate. In this ore, the concentration of uranium is low compared with that of the other species taking part in the reaction and, consequently, the influence of the uranium reactions on key variables such as acid concentration and redox potential is negligible. However, it is necessary to obtain the correct acid concentration and redox potential for the efficient leaching of uranium.

The leaching model includes only the reactions that are considered to be significant in the extraction of uranium. The selection of the reaction scheme for the leaching model was based on a study of the literature and consideration of dynamic process-data capture at an extraction facility, and does not include all the possible reactions. Such an approach would be unpractical because of the variability and complexity of the ore deposits. Moreover, the influence of many of the side reactions can be included in the influence of the reactions that are modelled. For example, the main acid-consuming species are accounted for by the reactions with chlorite (slow-reacting) and calcite (fast-reacting).

The eight chemical reactions included in the model are listed in Table I. These reactions and the reasons for their selection are described in detail below.

In consideration of the chemistry of uranium dissolution, it is necessary to take cognizance of the mineralogical characteristics of the uranium in Witwatersrand ores. In these ores, uranium is present as uraninite, in

TABLE I
REACTIONS AND RATE LAWS

Reaction no.	Reaction	Rate law	Rate constant	Activation energy	Reference
1	$2\text{Fe}^{2+} + \text{MnO}_2 + 4\text{H}^+ = 2\text{Fe}^{3+} + \text{Mn}^{2+} + 2\text{H}_2\text{O}$	$k[\text{Fe}^{2+}][\text{MnO}_2]$	0,025	0	5,6
2	$\text{UO}_2 + 2\text{Fe}^{3+} = \text{UO}_2^{2+} + 2\text{Fe}^{2+}$	$k \exp\left(\frac{-E}{RT}\right) [\text{UO}_2][\text{Fe}^{3+}]^{1/2} [\text{Fe}^{2+}]^{-1/2}$	180 000	61 300	4,7
3	$\text{UO}_3 + 2\text{H}^+ = \text{UO}_2^{2+} + \text{H}_2\text{O}$	$k \exp\left(\frac{-E}{RT}\right) [\text{UO}_3][\text{H}^+]^2$	$6,0 \times 10^{-8}$	61 300	—
4	$\text{O}_2 + 4\text{H}^+ + 4\text{Fe}^{2+} = 2\text{H}_2\text{O} + 4\text{Fe}^{3+}$	$k[\text{Fe}^{2+}]^2$, where $k = k' P_{\text{O}_2}$	$2,2 \times 10^{-3}$	0	5,8
5	$\text{FeO} + 2\text{H}^+ = \text{Fe}^{2+} + \text{H}_2\text{O}$	$k[\text{FeO}][\text{H}^+]^2$	0,15	0	—
6	$2\text{H}^+ + \text{CaCO}_3 = \text{Ca}^{2+} + \text{H}_2\text{O} + \text{CO}_2$	$k[\text{H}^+][\text{CaCO}_3]$	2,3	0	—
7	$6\text{H}^+ + \text{Mg}_2\text{Fe}(\text{OH})_6 = 2\text{Mg}^{2+} + \text{Fe}^{2+} + 6\text{H}_2\text{O}$	$k[\text{H}^+]^2[\text{Mg}_2\text{Fe}(\text{OH})_6]$	$4,0 \times 10^{-5}$	0	—
8	$\text{FeS}_2 + 14\text{Fe}^{3+} + 8\text{H}_2\text{O} = 15\text{Fe}^{2+} + 2\text{SO}_4^{2-} + 16\text{H}^+$	$k[\text{FeS}_2][\text{Fe}^{3+}]$	$3,8 \times 10^{-5}$	0	9-11

brannerite minerals, in coffinite, and in uraniferous zircon¹². Generally, however, uraninite and brannerite are the principal carriers of the element. In the leaching model, the uranium is therefore assumed to be present as uraninite, in the form of either UO_2 or UO_3 .

The UO_3 is treated as the fast-leaching uranium component. The reaction describing the dissolution of UO_3 in acid solutions^{3,13} is presented in reaction (3) of Table I. The dissolution of UO_2 is assumed to take place more slowly by reaction with ferric ions, as described in reaction (2)¹³.

Ferric ions are produced by the oxidation of ferrous ions with pyrolusite, as described in reaction (1)^{3,13}. The ferrous ions are present in the leach pulp as a result of the dissolution of the iron-bearing minerals, such as chlorite and pyrite, in the ore, and from the iron abraded from steel in the milling circuit^{3,4}.

For the leaching model, it is assumed that the iron that reacts to produce the ferrous ions within the pulp is present as FeO. The basis for this assumption is that, even though the source material is probably fairly diverse, the iron will be converted to the oxide after being exposed in the aerated pulps. The reaction of the iron oxide with hydrogen ions in acid pulps is described by reaction (5). A second mechanism is assumed by which ferrous ions are oxidized direct to ferric ions, namely reaction with dissolved oxygen in acid media. This reaction alone will not produce sufficient ferric ions at atmospheric pressure, but will have some influence on the overall oxidizing potential of the pulp. This ferrous oxidation reaction is described by reaction (4).

Pyrite, which is present in Witwatersrand ores, will react slowly with the ferric ions present in the leach pulp. This will result in the consumption of these ions in the leaching process, and will also add to the level of total iron in solution. This is described by reaction (8)¹⁴.

Various constituents in the gangue will react with the hydrogen ions in the pulp, and can consequently be termed acid-consuming species. However, in the leaching model, it is assumed that the major acid-consuming species are iron oxide—in reaction (5)—calcite, and chlorite¹⁵.

Chlorite is an aluminosilicate that has a double-layered structure incorporating a brucite-like layer¹⁵. The brucite layer accounts for about 34 per cent of the chlorite in Witwatersrand ores that dissolve by acid attack¹⁵. The dissolution of chlorite in uranium leach pulps is assumed to take place according to reaction (7).

The acid will also react with the calcite in the gangue—reaction (6)^{13,15}.

Reaction Kinetics

A summary of the rate expressions used for the leaching reactions is included in Table I. These rate expressions were obtained from the literature^{4,5-11} and, where no rate expression was available, one was proposed, either from a study of the available reaction data or from a consideration of the actual behaviour of the leaching process in question¹⁶. The selection of the appropriate rate expressions was an extremely difficult task in view of the limited process information available in the literature. Some of the rate forms had to be selected by intuition, and should therefore be regarded as conjectural.

The leaching kinetics of uranium are highly non-linear because the reaction rate, which is very rapid initially, becomes retarded as high conversions are approached. Such non-linear kinetics can be described by a number of approaches^{17,18}. In the uranium-leaching model, a distributed rate-expression approach was adopted. It was assumed that some of the uranium was fast-leaching while the rest was slow-leaching, and that both these fractions could be described by separate reactions and kinetic expressions. A typical dissolution plot for uranium ore is illustrated in Fig. 3.

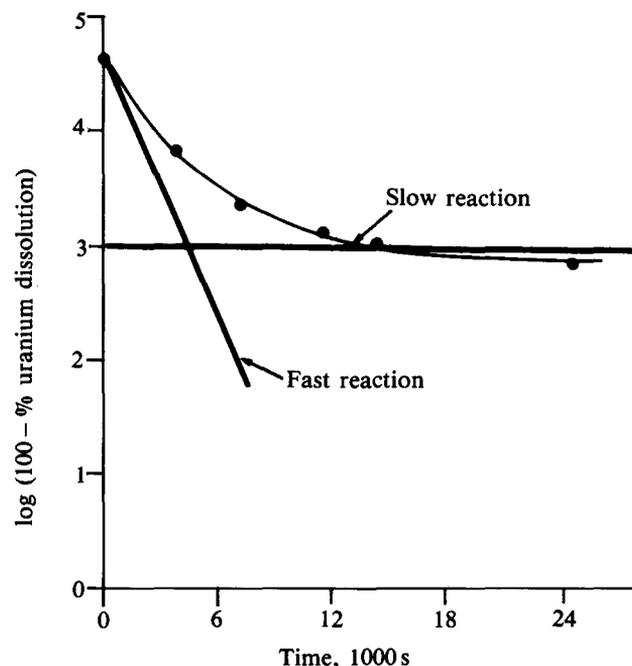


Fig. 3—A first-order plot of uranium dissolution against time¹⁹

The UO_3 was taken to represent the fast-leaching uranium species, the dissolution reaction being represented by reaction (1). The dissolution of UO_2 by reaction (2) was assumed to be slower.

Simulation and Results

The uranium-leaching model must behave like an actual processing plant if it is to be of use in process-control studies. As mentioned previously, the model must be able to simulate the dynamic behaviour of the plant in terms of the key process variables. For an adequate description of the process, it is necessary to compare the simulations with the actual dynamic plant data, and to adjust the model so that the correlation between the simulated measurements and the actual process measurements will be reasonably good. The model parameters that can be adjusted are the feed concentration, reaction rate constants and activation energies, reagent concentrations, pulp and reagent flows, residence times, and steam flows and temperatures.

Two sources of data were used for the validation and fitting of the process model: averaged steady-state data obtained from plant samples that had been collected during leaching studies on a plant¹⁹, and dynamic plant data that were logged for use in the validation of the model²⁰.

The plant data obtained from the leaching studies¹⁹ were used as fixed averaged inputs to the model. In this way, a simulation could be run to steady state, and the outputs compared with the averaged process measurements. The simulations reached steady state after 50 000 seconds at an integration time interval of 50 seconds¹⁷. The variables that were considered to be significant when the behaviour of the model was matched to the plant data were the concentrations of ferric and ferrous ions, the acid concentrations, and the percentage extraction of uranium. The simulated concentration profiles are presented in Figs. 4 and 5. In Fig. 4, the simulation represents the plant being run under a 'decay' leaching strategy that involves the addition of all the reagents to the first leaching tank. In Fig. 5, the simulation represents the plant being run under 'controlled' leaching conditions that involve the addition of acid and pyrolusite to the first, second, and fourth leaching tanks.

For the dynamic simulations, the pulp flow, pulp density, reagent flows, and tank temperatures were read as inputs to the model from logged plant data²⁰. Wherever

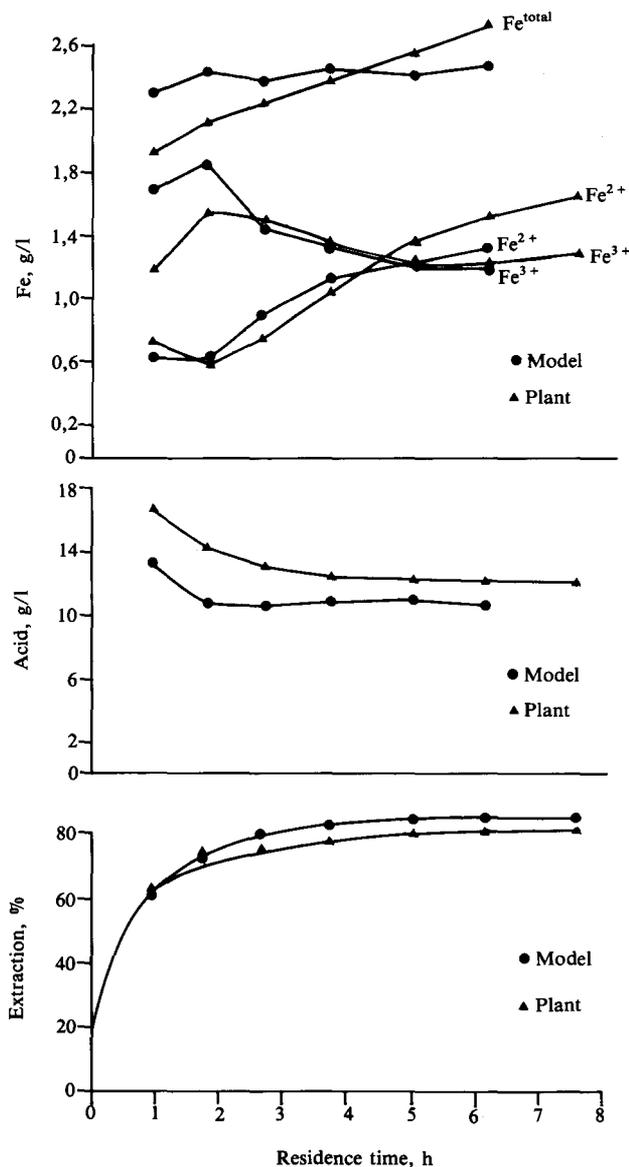


Fig. 4—Regime 1: Decay leach

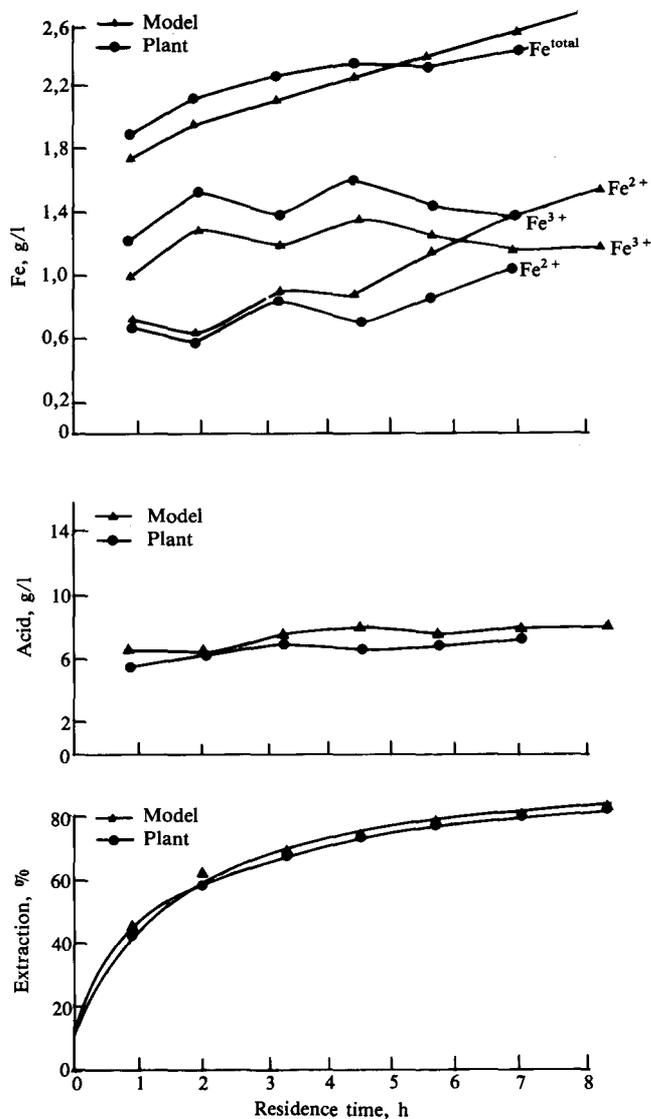


Fig. 5—Regime 2: 'Controlled' leach in which acid and MnO₂ were added to tanks 1, 2, and 4

input data were not given explicitly, they were estimated from averages of the data that were available from plant log sheets. The active tank volumes, rate data, concentrations of feed species, and the concentration of pyrolusite were taken to be the same as those used during the simulations that were run to steady state, as described above.

A comparison of the measured, titrated, and predicted values of the redox potential in the first leaching tank over a period of two days is shown in Fig. 6. It should be noted that the predicted and the titrated values follow a similar trend, but that the trend is totally different from that of the measured values. The predicted values also have a noticeable offset. Besides emphasizing the difficulty incurred in the determination of the absolute 'correct' values, this shows that the predictions of the model do indeed track the process, even if this is not what the redox instrument reports. The offset is caused by inaccuracies in the calibration of the various instruments around the plant, which influence this particular predicted variable. Inaccuracies were a major problem throughout the present investigation.

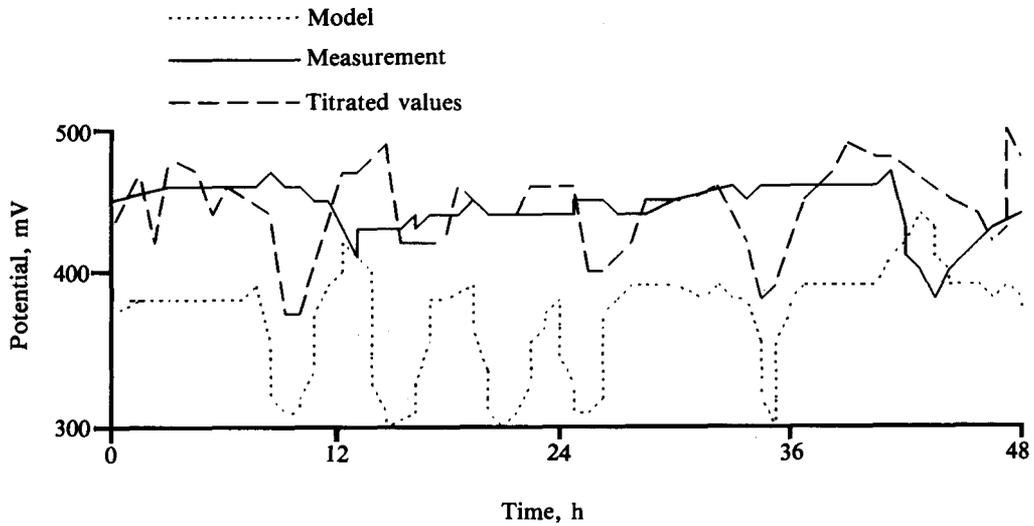


Fig. 6—Redox potential in tank 1

Fig. 7 compares the measured, titrated, and predicted values of the acid concentration and of the redox potential in the first tank over another period—three days on this occasion—of particularly poor agreement. It is apparent that there are some major discrepancies between the plant data and the simulations in this case. Of particular interest are the periods 12 to 18 hours and 34 to

41 hours, during which the model predicts that the acid concentration should rise and the redox potential should drop. Instead, a relatively steady acid concentration and redox potential is observed. This deviation occurs as the result of fluctuations in the pyrolusite activity during these periods, since unexpected increases in the pyrolusite activity cause extra acid to be consumed and increase the

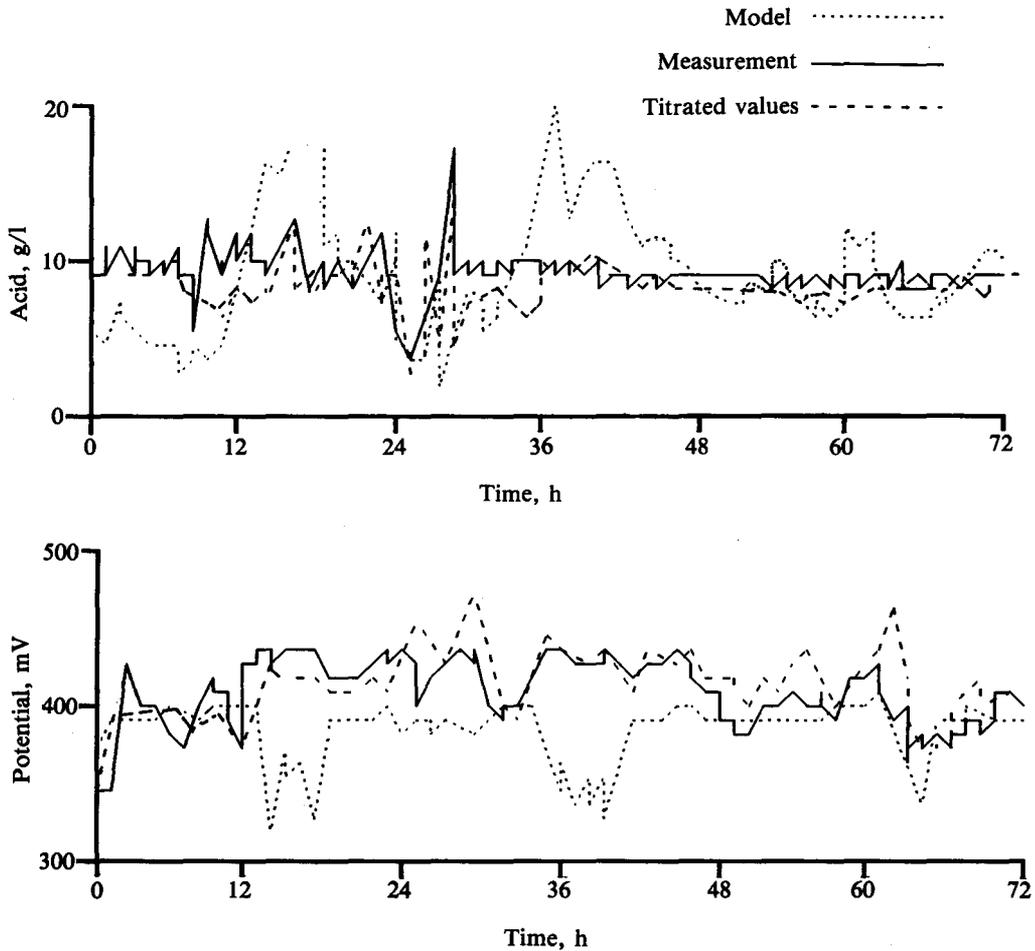


Fig. 7—Example of predicted acid concentrations and redox potentials in tank 1 when the pyrolusite feed is noisy

redox potential. As such changes in pyrolusite activity are not input to the model, it has no direct way of predicting these changes. This emphasizes the multivariable nature of the problem in that it is necessary to take more than one variable into account before the source of the problem can be identified.

Discussion

The uranium-leaching model was found to agree adequately with the averaged plant data, but not with the continuous plant data. This was attributed to the following causes.

- (a) There were unmonitored variations in the pyrolusite fed to the plant. These variations appeared to occur as a result of variations in the feed materials and in the reagent concentration in the slurry.
- (b) Some of the plant instruments were not properly calibrated, while others were non-operational. It was found that the model could be used as an aid identification of faulty or wrongly calibrated instruments, but this aspect would need further investigation.
- (c) The residence times were uncertain as a result of silting in the tanks, and the bypassing of tanks without similar modification of the model.

There is no evidence to indicate that the model itself is the source of any major discrepancies. It can therefore be concluded that the model would be adequate to support a process-control investigation.

From the results obtained from plant studies, it is clear that the addition of pyrolusite has a major influence on the process as a result of variations in the feed material and the reagent concentration in the slurry. This factor is not evident unless the process is studied dynamically and globally.

As a result of the closure of this project, it was not possible to obtain more-comprehensive and more-consistent process data for validation of the model, or to determine which control structure would be most suitable for the South African uranium industry. A number of control options are available, ranging from model-based decision-support systems to optimal controllers. Before the work on uranium-leaching control was suspended at Mintek, some work was done in which the leaching model was incorporated in a recursive estimator that uses a trajectory filter²¹. In such a structure, the model can be used as a decision-support structure. However, no work was done on the user interface of such a decision-support system, or on the hardware infrastructure necessary for the implementation of such a system on a plant.

Conclusions

The model that was developed describes the process adequately in terms of the main variables of concern. The results are in good agreement with the averaged plant data, but difficulties were encountered when recorded plant data were used in an attempt to predict the dynamic evolution of the plant. This shortcoming was caused by several factors, the main one being the uncertainty in the pyrolusite feed system.

The present work was undertaken as part of an investigation on the control of a uranium-leaching plant. The results as summarized in this paper are therefore not conclusive. Further work is needed in which the control

of the process would be investigated more fully, instruments would be evaluated and, possibly, a comprehensive control system or a decision-support system would be developed for such processes. Unfortunately, further work has now been suspended because of the poor market for uranium, but it could be resumed if the need arose.

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Nomenclature

c	Species concentration, $\text{kg}\cdot\text{m}^{-3}$
F	Flow, $\text{m}^3\cdot\text{s}^{-1}$
C_p	Heat capacity, $\text{J}\cdot\text{kg}^{-1}\cdot\text{C}^{-1}$
L	Lumped heat-loss term, $\text{J}\cdot\text{C}^{-1}$
$R()$	Rate expression
T	Temperature, $^{\circ}\text{C}$
V	Tank volume, m^3
t	Time, s
ρ	Density, $\text{kg}\cdot\text{m}^{-3}$
λ	Latent heat $\text{J}\cdot\text{m}^{-3}$
NS	Number of species

NT Number of tanks

Subscripts and superscripts

i	Species index ($i = 1, \text{NS}$)
j	Tank index ($j = 1, \text{NT}$)
a	Ambient conditions
s	Steam
acid	Reagent acid stream
MnO_2	Reagent pyrolusite stream
Steam	Process steam
Water	Condensed steam

Geostatistics at Wits*

One of the world's leading experts in the evaluation of ore reserves and author of the book *Practical Geostatistics*, mining engineer Dr Isobel Clark, has joined the Department of Mining Engineering at the University of the Witwatersrand.

On her recent arrival from London to take up her post, Dr Clark explained that she had taken advantage of the offer of an academic position at Wits because of her close ties with the South African mining industry.

She is to take over the responsibilities of well-known expert in geostatistics Professor Danie Krige, who retired at the end of 1988, and will also be involved in the development of new courses to expand the scope of the undergraduate mining-engineering degree.

Dr Clark is the founder and managing director of London-based consultancy Geostokos Limited, which specializes in the interpretation and valuation of ore deposits worldwide.

'I have consulted for South African mining houses on many occasions in the past and will continue to act on behalf of Geostokos in South Africa when my academic obligations permit,' says Dr Clark.

Dr Clark was born in London and brought up in Scotland, obtaining her first degree at Strathclyde University, Glasgow. She studied for her Masters Degree at Reading



Dr Isobel Clark

University in the south of England, and then spent a year in the civil service as a statistician for the Department of Education and Science in London.

A conviction that she could do more with her life and an inclination towards engineering took her in a teaching capacity to the Department of Mining and Mineral Technology at the London Royal School of Mines, where she remained for almost 12 years.

During this period she obtained her Ph.D., specializing in reserve estimation. Her book published in 1979, *Practical Geostatistics*, has been described as the most readable account of the topic ever written—not surprisingly as her first love is the essentially practical production side of mining.

'The mining industry has plenty of high-quality technical expertise. What it needs is an interface between the mathematicians and the mines themselves,' she says.

She points out that only five years ago geostatistics as a routine method of ore evaluation was virtually unknown in South Africa. 'Today all the major mining houses have dedicated computerized geostatistics departments.' She likes to think that her close involvement with the local mining industry has made some contribution to this progress.

She stresses, however, that her work involves applying a synthesis of the most appropriate estimating techniques to the case in hand. 'Geostatistics may be the most modern method of evaluation, but there is nothing wrong with traditional techniques.'

Her responsibilities in the Department of Mining Engineering at Wits will include the teaching and supervision of four post-graduate mining courses, the development of a new undergraduate course in reserve valuation, and the development and expansion of the undergraduate mine-surveyors course.

* Press release from Lynne Hancock Communications (P.O. Box 1564, Parklands 2121).