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FURTHER STUDIES OF THE LEACHING
OF URANIUM ORES FROM
ELLIOT LAKE, ONTARIO

by

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EXTRACTION METALLURGY DIVISION

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ABSTRACT

The uranium mines of Elliot Lake area in Ontario, Canada, were
brought into production in the mid-1950's. At that time the extraction
plants were designed to utilize an acid leaching process which, although
adequate in that about 95 percent extraction of the uranium was attained,
was not thoroughly understood. Because of this the plant operators found
it necessary to make trial-and-error adjustments to the operating condi-
tions, one of the more important of these being to increase the temperature
of the pulp during leaching. It was found that as the leaching temperature
increased the acid addition needed for acceptable extraction decreased, the
combined effect being a reduction in operating costs. A recent laboratory
study has served to clarify further this relationship between leaching
temperature and acid requirement. At the same time the effects of other
operating variables were also studied more thoroughly than had been done
in the past. The results of this investigation, which are reported in this
paper, not only supplied new information related to the extraction of
uranium from the Elliot Lake ores, but also emphasized the fact that
currently used processes may not be understood sufficiently. Consequently,
it may be profitable for operators to study existing processes further before
they embark on major process changes.

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NOUVELLES ÉTUDES SUR LE LESSIVAGE DES MINERAIS D'URANIUM D'ELLIOT LAKE (ONTARIO)

par

H.H. McCreedy*, W.A. Gow**, F.J. Kelly*** et H.W. Smith***

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RÉSUMÉ

Les mines d'uranium de la région d'Elliot Lake, Ontario, sont en exploitation depuis le milieu des années 1950. A cette époque, les usines d'extraction étaient conçues pour utilizer un procédé de lessivage à l'acide qui, tout en permettant l'extraction de l'uranium dans une proportion de 95 p. 100, n'était pas entièrement compris. Pour cette raison, les exploitants d'usines devaient expérimenter constamment avec le procédé de traitement, notamment en haussant la température de la pâte durant le lessivage. Il fut établi que la quantité supplémentaire d'acide nécessaire à l'extraction efficace variait inversement avec l'augmentation de la température pendant le lessivage, le résultat net étant une diminution des frais d'exploitation. Une étude récente en laboratoire a permis de préciser davantage le rapport qui existe entre la température de lessivage et la quantité d'acide requise. A la même occasion, on a étudié plus à fond les effets de certaines autres variables de fonctionnement. Les résultats de cette étude, qui font l'objet du présent rapport, ont non seulement fourni de nouveaux renseignements sur l'extraction de l'uranium des minerais d'Elliot Lake, mais ont aussi fait ressortir le fait que les procédés actuels ne sont peut-être pas assez bien compris. Par conséquent, il serait peut-être à l'avantage des exploitants d'approfondir leur connaissance des procédés existants avant d'entreprendre des modifications importantes.

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INTRODUCTION

The leaching plants built in the mid-1950's to treat the uranium ore at Elliot Lake, Ontario, were designed to acid-leach the ore at a temperature of 45°C with an initial free-acid concentration of 50 to 70 g H_2SO_4 per litre. The plants were also designed to grind the ore to about 60% minus 200 mesh, after which the slurry was thickened to 68% solids before going to the leach circuit. These plants were equipped to feed sodium chlorate to the leach to maintain a minimum ferric-iron concentration of 2 g per litre at the conclusion of leaching[1,2].

The initial operating parameters were based on laboratory and pilot-plant test work. Because the time available for the design work was very limited, there could be no claim that the results had delineated the optimum leaching conditions. It was expected that, as with most ore processing plants, experimentation in the full-scale plant would develop more efficient operating parameters than those derived from the pilot-plant studies. In fact, this turned out to be the case.

In current practice, the operators at Elliot Lake leach in rubber-lined pachucas at temperatures of about 75°C. Pulp densities have been increased to about 75% solids. Under these conditions it has been possible to reduce the acid consumption by 10 to 20 lb/ton and to operate with a lower final ferric-iron concentration without causing a reduction in the uranium extraction. This in turn meant that the sodium chlorate addition could be reduced considerably or eliminated.

Because these economically attractive changes in operating conditions were possible, it was apparent that the interrelated effects of initial acid concentration, leaching temperature and the ferric: ferrous iron ratio in the leaching solution had not been fully explored in the initial testwork. It was evident that these relationships should be studied more fully in case further economies were possible in preparation for the time when increased mill capacity might have to be designed.

This paper describes laboratory studies done to investigate the interactions of temperature, initial acid addition, oxidation potential of the leach solution, and ore grind. The investigations were done using factorially designed experiments, and regression methods were used in analyzing the results.

PROCEDURE

The ore used in this work was rod-mill feed from the Algom Nordic mill of Rio Algom Mines Limited at Elliot Lake, Ontario. This ore, which was typical of the Elliot Lake uranium deposits, was quartz conglomerate containing brannerite, uraninite and monazite as the most important uranium-bearing minerals, their relative abundance being in the order listed. The chemical analysis of the sample showed that it contained 0.13% U_3O_8, 4.58% iron, 3.81% sulphur, and 0.1% carbonate. The ore as received was minus 4 mesh, and it was crushed to minus 10 mesh prior to grinding in a porcelain Abbé laboratory mill with steel balls.

The leaching was done in a covered, glass, reaction kettle fitted with a reflux condenser and heated by an automatically controlled, electric heating mantle. The slurries were stirred by a motor-driven glass impeller.

The experimental designs and the statistical analyses of the results were made according to recognized statistical methods. It is beyond the scope of this paper to describe in any detail the way in which the statistical data were handled. (However, an earlier report by the authors has been published in which more details regarding the mathematics have been provided[3]; it provides references dealing with the design of experiments and the mathematical analysis of their results[4-8].) The regression calculations of the present paper were done by computer, and the programs used, written in Fortran II, IV and V, are available on request.

RESULTS

Series One

The first series of tests was done to investigate the effects, on the uranium extraction rate, of changes in the amount of sulphuric acid added to the leach initially and in the temperature of the slurry during the leaching. In these tests, three levels of acid addition and four levels of slurry temperature were investigated, while the fineness of grind, sodium chlorate addition and pulp density were the same for all tests. In this series twenty-four tests were done, i.e. twelve sets of conditions tested in duplicate. The conditions applied in Series One were:

      Acid addition, lb/ton - 40, 60, 80
       Slurry temperature, °C - 65, 75, 85, 95

Fineness of grind, % plus 100 mesh - 5

% minus 200 mesh - 63

Sodium chlorate addition, lb/ton   - 1

Pulp density of leach slurry, % solids - 70

Fig. 1, based on the results of the Series One tests, shows the relationship between initial acid addition, temperature, and 48-hour U_3O_8 extraction. The curves of equal extraction shown in Fig. 1 were calculated from the following equation, that was developed from the test data by standard regression techniques:[4]

48-hr U_3O_8 extraction, % =

$13.18 - 0.16\ X_1 + 1.72\ X_2 + 0.0028\ X_1^2 - 0.0111\ X_2^2 +$

$0.0019\ X_1X_2,\ ..\ (1)$

where X_1 = initial acid addition, lb/ton and
       X_2 = slurry temperature, °C.

As shown by Fig. 1, the 48-hour extraction can be increased by increasing the initial acid concentration and by increasing the leach temperature up to about 82°C. As the temperature is increased above 82°C, the initial acid addition would also have to be increased to maintain a given 48-hour U_3O_8 extraction. This suggests that there would be no advantage in increasing the temperature of the leach slurry above about 80°C.

Other data obtained from the Series One tests are given in Fig. 2, 3 and 4. These graphs show that the rate at which the sulphuric acid is consumed (Fig. 2), the rate of dissolution of iron (Fig. 3), and the rate at which the solution is reduced as indicated by the decrease in oxidation potential (Fig. 4), all increase with increasing temperature when all the other leaching conditions are constant. That is, all the observed reaction rates except the rate of dissolution of uranium increase with increasing temperature, but rate of uranium extraction increases up to 82°C and then decreases at temperatures higher than 82°C.

The empirical mathematical relationships between the oxidation potential of the leach solution, and acid consumption, the initial acid addition and the leaching temperature, as determined

by regression analysis techniques,[3] are shown below (Equations 2 and 3):

48-hr oxidation potential, mv = $458.9 + 0.38 X_2$, .. (2)

48-hr H_2SO_4 consumption, lb/ton = $0.144 + 0.54 X_1 + 0.18 X_2$,..(3)

where X_1 = initial acid addition, lb/ton and

X_2 = slurry temperature, °C.

Series Two

A second series of tests was then done, in which the interactions of initial sulphuric acid addition, temperature, initial sodium chlorate addition and grind were investigated. The levels of these variables investigated, along with the corresponding 48-hour U_3O_8 extraction, the 48-hour acid consumption, and the 48-hour oxidation potential of the solution, are given in Table I. The choice of the levels of variables was based on the central, composite, experimental design. (4-8).

The development, by standard regression techniques, of empirical models describing the relationship between the operating conditions tested and the responses measured gave the following results:

48-hr H_3O_8 extraction, % = $58.8 + 0.274 X_1 + 0.187 X_2 + 2.275 X_3$, (4)

48-hr H_2SO_4 consumtpion, lb/t = $7.65 + 0.40 X_1 + 0.21 X_2 - 0.22 X_3$, (5)

48-hr oxidation potential, mv = $413.6 - 0.7 X_2$,

where X_1 = initial acid addition, lb/ton,

X_2 = slurry temperature, °C, and

X_3 = initial sodium chlorate addition, lb/ton,

As with Series One, the results of Series Two showed that the 48-hour U_3O_8 extraction was affected mainly by the initial acid addition and the slurry temperature. In Series Two, where initial sodium chlorate addition was a variable, the U_3O_8 extraction was also affected by changes in this variable. The fineness of grind within the levels used in this series had no effect on any of the parameters measured (Equations 4, 5 and 6). Fig. 5 shows graphically the interrelated effects of acid and sodium chlorate additions and temperature on the 48-hour U_3O_8 extraction.

The results of the Series Two tests (Fig. 5) show, as did the results of Series One (Fig. 1), that over the range of temperature of 65 to 80°C, for a given extraction, increasing the temperature allows a decrease in acid. Fig. 5 also shows that an increase in sodium chlorate addition allows a decrease in both the required acid and temperature.

Equation 5 shows that the acid consumption increases with increased acid and temperature, indicating that the rates of the chemical reactions involving acid increase with acid concentration and temperature. This was also the case in the Series One tests. Equation 5 also suggests that the acid consumption increases with decreased sodium chlorate but the effect is so small as to be negligible within the levels of sodium chlorate addition investigated.

Equation 6 shows that the 48-hour oxidation potential of the leaching solution is affected only by the temperature: the higher the temperature the lower is the 48-hour oxidation potential. Temperature had a similar effect on the 48-hour oxidation potential in the Series One tests, but in Series One the initial acid concentration also affected the oxidation potential.

DISCUSSION

Obviously the results of this work are of specific interest only to those people concerned with the treatment of the Elliot Lake uranium ores. The results do have a general interest for operators in other areas, in that they demonstrate the effectiveness of two carefully designed series of tests in improving the understanding of hydrometallurgical processes. This in turn can lead quickly to improved operating practices and better plant design. The evidence behind this observation is that this study (which was based on factorially designed experiments, the results of which were analyzed by known regression techniques) demonstrated in a few relatively inexpensive tests what had taken considerable period of in-plant experimentation to learn. In addition, the results provided new information which can be applied in future design and operation of Elliot Lake plants.

For example, the Elliot Lake operators gradually increased the temperature of their leaching from the 45°C, which the leaching plants were designed to handle, to about 75°C which is the current practice. The results of the work reported in this paper show that the effect of temperature as observed in the plants was predictable, and that, for a given acid addition, increasing the

temperature would result in an increased uranium extraction rate, within the limits of past plant practice (< 75°C). New information arising from this study has shown that there is no benefit to be obtained in leaching the Elliot Lake Ore at temperatures above 85°C. In view of this, the designers of future plants need not concern themselves with designing for leach temperatures higher than 85°C.

The oxidation potential of the solution at the end of leaching is used as an operating control in the Elliot Lake plants. The results of this work indicate that the rate of uranium extraction is dependent to some extent on the oxidation potential as measured at the end of the leach. In Series One, for example, it appears that the decrease in extraction rate which occurs as the temperature is increased above 85°C is largely due to the very low oxidation potentials (as low as 310 mv) observed at the higher temperatures. The results of both Series One and Series Two tests show that, as the temperature is increased to 80°C, the effect of temperature outweighs the effect of the lower oxidation potential resulting from the higher temperature. The results suggest that at a temperature of 80°C the oxidation potential should be controlled in the range of 350 to 360 mv, for satisfactory extraction, while at a temperature of 60°C the final oxidation potential would have to be 375 to 390 mv. The oxidation potential could be controlled either by the addition of a chemical oxidant such as sodium chlorate ro by increased aeration.

In Series Two tests, the sodium chlorate had an effect on the U_3O_8 extraction but did not appear to affect the 48-hour oxidation potential of the solution. These results suggest that the need of additional oxidation of the leach solution should not be based on the observed final oxidation potential. Perhaps, measurement of the oxidation potential part way through the leach, rather than at the end of the leach, would be a better means of controlling the addition of oxidant.

The fineness of grind of the Elliot Lake ore was not a significant variable within the range studied in this work, 45 to 85% minus 200 mesh. That the fineness of grind was not critical has been generally accepted by the operators on the basis of plant results. However, this work shows that the grind can vary over a wide range without having a detrimental effect on the leaching operation. The inference from these results is that it is not necessary to equip grinding circuits in the Elliot Lake area with complex automatic controls designed to maintain a consistent grind.

CONCLUSIONS

The purpose of this paper is not just to present data
relevant to the treatment of Elliot Lake ore. It is hoped, also,
that this work will demonstrate the value of well-designed
laboratory experiments for assisting the operators and designers
of hydrometallurgical processes. It might well be that, in the
designing of new plants to handle the predicted increase in
demand for uranium, it will be more profitable to spend some
time working towards a better understanding of known processes
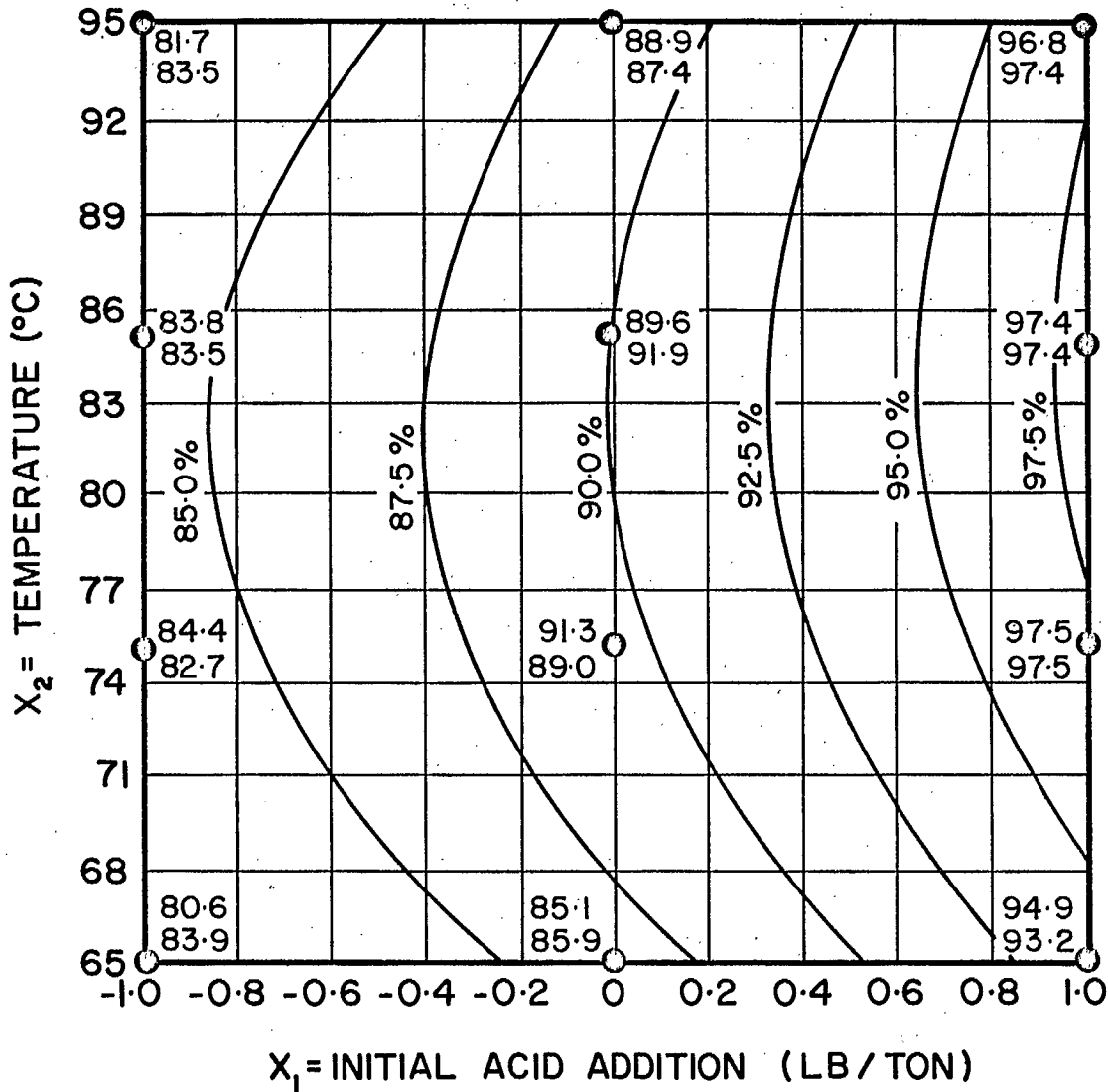than to devote all of our efforts towards new and untested
processes.

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TABLE 1

Test Conditions and Results, Series Two

Run No.	CONDITIONS				RESULTS		
	X1 Acid (lb/t)	X2 Temp. (°C)	X3 Oxidant (lb/t)	X4 Grind (%-200M)	48-hr U_3O_8 extraction (%)	H_2SO_4 consumption (lb/t)	48-hr oxidation potential (mv)
1	50.0	55.0	1.0	50.5	87.1	39.9	370
2	50.0	55.0	2.0	71.1	89.1	40.3	374
3	70.0	55.0	1.0	49.5	89.8	43.6	375
4	70.0	55.0	2.0	71.2	90.8	49.3	382
5	50.0	75.0	1.0	71.9	87.7	42.8	378
6	50.0	75.0	2.0	49.6	93.9	43.0	364
7	70.0	75.0	1.0	72.0	97.6	51.7	362
8	70.0	75.0	2.0	54.3	97.5	51.9	369
9*	60.0	65.0	1.5	62.9	89.6	45.4	364
10*	60.0	65.0	1.5	60.4	90.1	48.6	365
11*	60.0	65.0	1.5	59.1	89.9	47.9	376
12	70.0	75.0	2.0	71.8	97.5	52.7	361
13	70.0	75.0	1.0	52.1	96.8	52.3	373
14	50.0	75.0	2.0	72.3	92.0	40.4	361
15	50.0	75.0	1.0	54.3	88.7	43.7	360
16	70.0	55.0	2.0	52.6	95.9	40.6	397
17	70.0	55.0	1.0	73.0	90.7	47.5	375
18	50.0	55.0	2.0	53.7	89.1	38.6	379
19	50.0	55.0	1.0	73.0	84.3	39.6	366
20	60.0	65.0	0.5	65.3	88.6	44.6	357
21	60.0	65.0	2.5	62.1	90.7	45.4	365
22	40.0	65.0	1.5	61.5	87.1	35.3	366
23	80.0	65.0	1.5	62.1	97.7	53.1	373
24	60.0	45.0	1.5	61.7	91.0	40.9	398
25	60.0	85.0	1.5	62.5	96.0	47.5	359
26	60.0	65.0	1.5	54.6	88.8	47.4	374
27	60.0	65.0	1.5	86.9	89.0	45.8	366
28*	60.0	65.0	1.5	62.9	89.2	49.8	353
29*	60.0	65.0	1.5	61.6	89.2	48.4	360
30*	60.0	65.0	1.5	61.0	87.7	47.6	352
31*	60.0	65.0	1.5	61.8	87.3	48.4	351
32*	60.0	65.0	1.5	61.8	87.3	48.5	354

* Duplicate runs.

Fig. I. Interrelation of leach temperature, initial acid addition, and uranium extraction (Series One).

Note: Extraction curves are based on the following regression equation:

$$\text{Ext'n } \% = 13.18 - 0.16X_1 + 1.72X_2 + 0.0028X_1^2 - 0.0111X_2^2 + 0.0019X_1X_2.$$
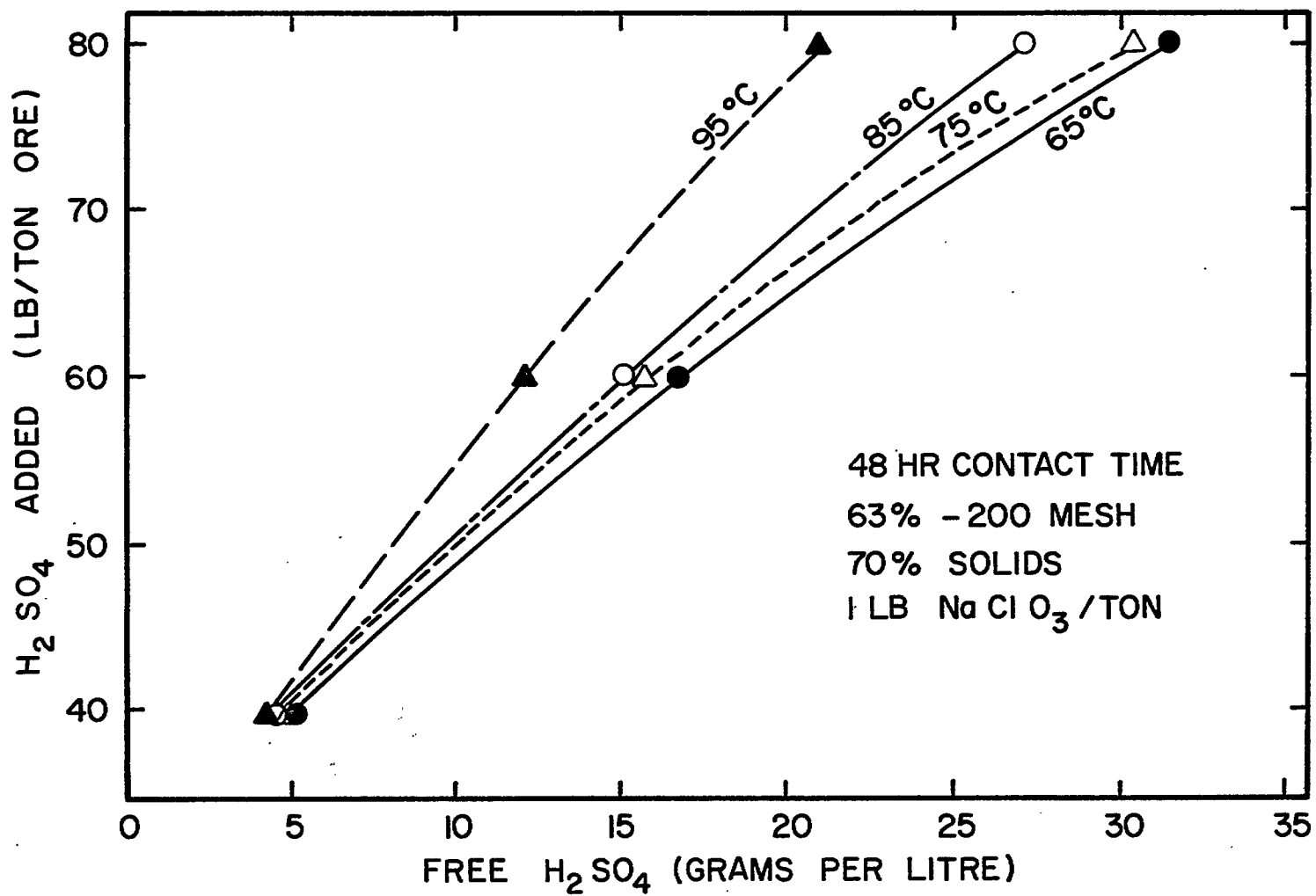
● Test results.

Fig. 2. Effect of acid added and temperature on free acid remaining in leach liquor (Series One).
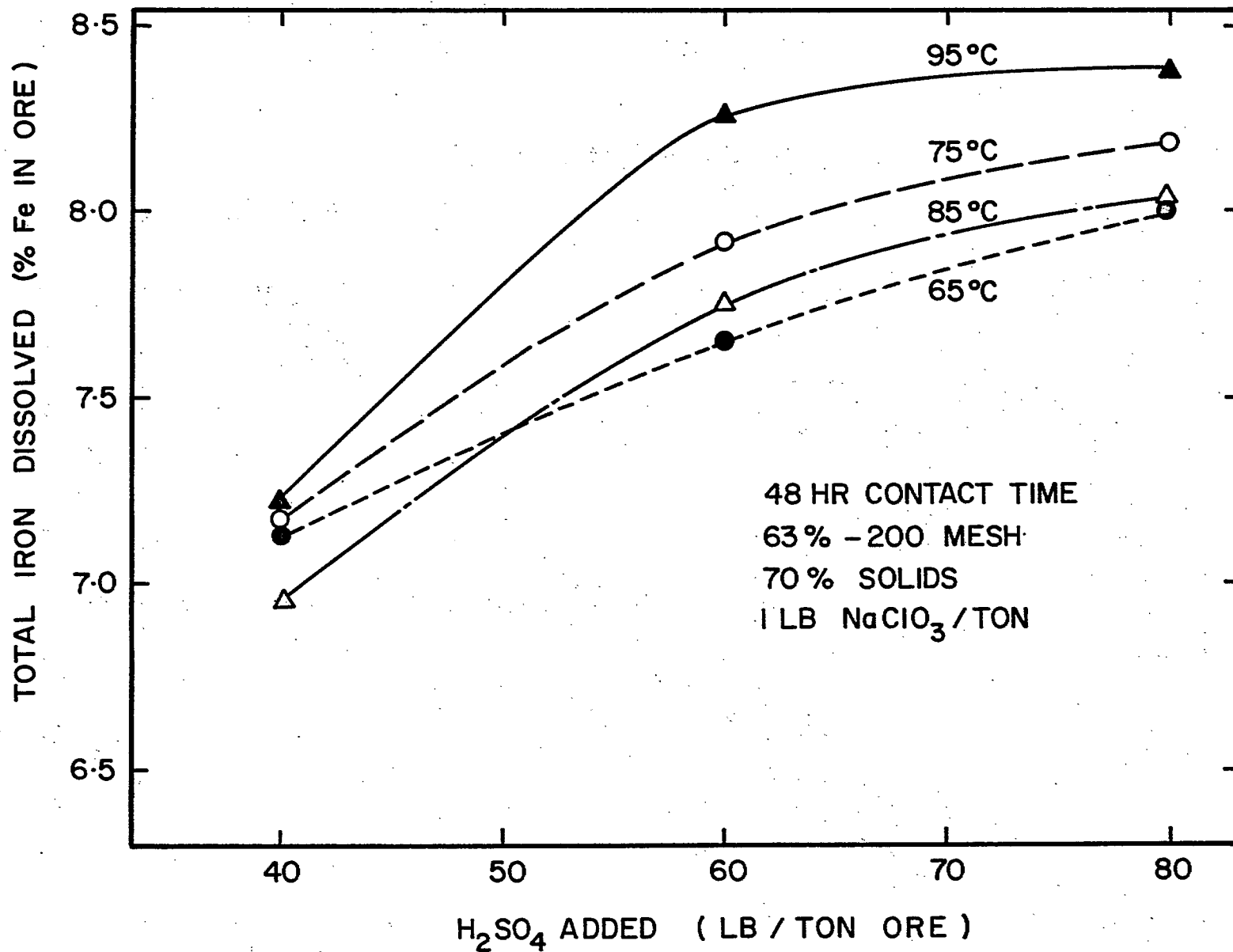
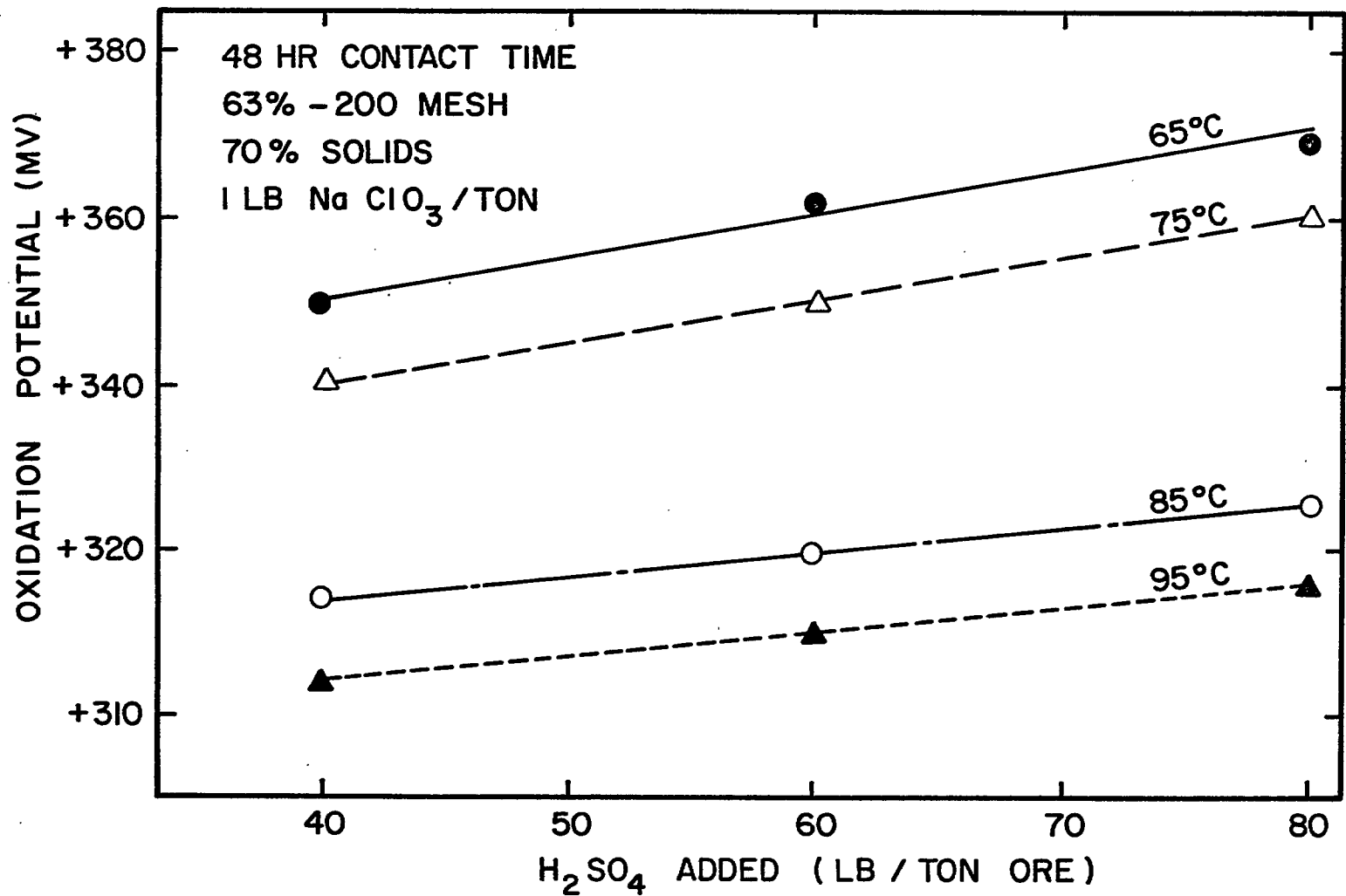Fig. 3. Effect of acid added and temperature on total iron dissolved (Series One).
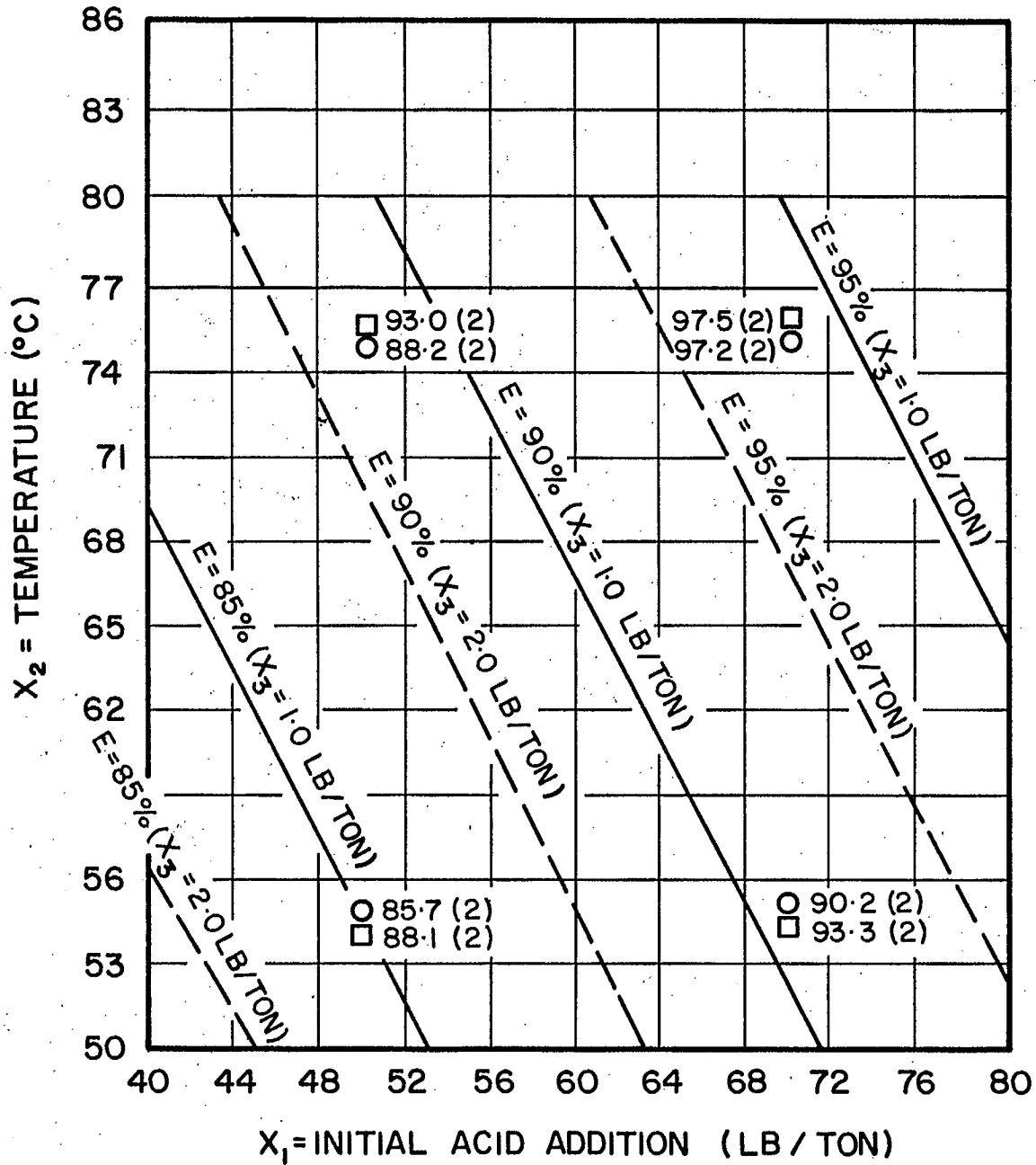
Fig. 4. Effect of acid added and temperature on oxidation potential of final leach solution (Series One).

Fig. 5. Interrelation of temperature, initial acid addition, sodium chlorate addition, and uranium extraction (Series Two).

Note: Extraction lines based on regression equation:
Ext'n,% = 58.8 + 0.274X_1 + 0.187X_2 + 2.275X_3,

Where X_1 = initial acid addition, lb/t;
X_2 = slurry temperature, °C;
X_3 = sodium chlorate addition, O - 1 lb/t,
□ - 2 lb/t.