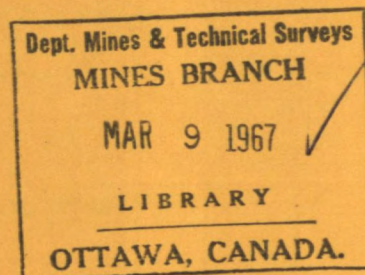




CANADA

DEPARTMENT OF
ENERGY, MINES AND RESOURCES
MINES BRANCH
OTTAWA



*FACTORS INFLUENCING THE
APPLICATION OF BACTERIAL
LEACHING TO A CANADIAN
URANIUM ORE*

V. F. HARRISON, W. A. GOW AND M. R. HUGHSON

EXTRACTION METALLURGY DIVISION

DECEMBER 1966



© Crown Copyrights reserved

Available by mail from the Queen's Printer, Ottawa,
and at the following Canadian Government bookshops:

OTTAWA

Daly Building, Corner Mackenzie and Rideau

TORONTO

Mackenzie Building, 36 Adelaide St. East

MONTREAL

Aeterna-Vie Building, 1182 St. Catherine St. West

or through your bookseller

A deposit copy of this publication is also available
for reference in public libraries across Canada

Price 50 ¢ Catalogue No. M34-20/85

Price subject to change without notice

ROGER DUHAMEL, F.R.S.C.

Queen's Printer and Controller of Stationery
Ottawa, Canada

1966

Mines Branch Technical Bulletin TB 85

FACTORS INFLUENCING THE APPLICATION OF
BACTERIAL LEACHING TO A CANADIAN URANIUM ORE

by

V.F. Harrison*, W.A. Gow** and M.R. Hughson***

ABSTRACT

Laboratory tests were conducted to investigate the effects of particle size and composition of the leaching solution on the extraction of uranium from an Elliot Lake ore in the presence of bacteria. It was shown that the leaching of particles coarser than 4 mesh in the presence of the Ferrobacillus-Thiobacillus group of bacteria was a very slow process, with only 70% of the uranium being extracted in thirty-two weeks. Uranium extractions of 90% were obtained when leaching particles finer than 4 mesh, in periods ranging from five to twenty weeks. The work showed that for ores coarser than 8 mesh it was necessary to add initially to the leach solution 1.0 g Fe⁺⁺/l and nutrient salts containing nitrogen, phosphorus and potassium. Extraction rate of uranium increased with initial iron concentrations of up to 1.0 g Fe⁺⁺/l. The addition of iron and nutrient was not required for ores finer than 8 mesh. The work also indicated that the extraction rate is determined by the rate at which the leaching solutions can penetrate the particles to depth. Further tests showed that bacterial leaching could be used to recover most of the uranium from reject mill products such as flotation tailings. Mineralogical studies were done to determine how bacterial leaching affected the minerals present in the Elliot Lake ore.

*Senior Scientific Officer, **Head, Hydrometallurgy Section, ***Scientific Officer, Extraction Metallurgy Division, Mines Branch, Department of Energy, Mines and Resources, Ottawa, Canada.

Direction des mines

Bulletin technique TB 85

FACTEURS INFLUENÇANT L'APPLICATION DU LESSIVAGE
BACTÉRIEN À UN MINÉRAI D'URANIUM CANADIEN

par

V. F. Harrison*, W. A. Gow**, et M. R. Hughson***

RÉSUMÉ

Les auteurs ont effectué des essais de laboratoire pour déterminer les effets de la granulométrie et de la composition de la solution de lessivage sur l'extraction de l'uranium d'un minerai d'Elliot Lake en présence de bactéries. Ils ont démontré que le lessivage des particules ne traversant pas le tamis de 4 mailles, en présence du groupe de bactéries Ferrobacillus-Thiobacillus, était un procédé très lent, ne permettant d'extraire que 70 p. 100 de l'uranium en 32 semaines. On a extrait 90 p. 100 de l'uranium en lessivant des particules traversant le tamis de 4 mailles, en des périodes de cinq à vingt semaines. L'étude a montré que, pour du minerai tamisé à plus de 8 mailles, il était nécessaire d'ajouter au préalable à la solution de lessivage 1.0 g de Fe^{++}/l et des sels nutritifs contenant de l'azote, du phosphore et du potassium. La vitesse d'extraction de l'uranium a augmenté avec des concentrations initiales de fer allant jusqu'à 1.0 Fe^{++}/l . L'addition de fer et de matières nutritives ne s'est pas avéré nécessaire pour du minerai traversant le tamis de 8 mailles. L'étude indique aussi que la vitesse d'extraction est déterminée par la vitesse à laquelle les solutions de lessivage peuvent s'infiltrer à l'intérieur des particules. D'autres essais ont démontré que le lessivage bactérien pourrait être utilisé pour récupérer la plus grande partie de l'uranium des rebuts de l'usine de traitement, comme les résidus de la flottation. Des études minéralogiques ont été effectuées pour déterminer comment le lessivage bactérien affecte les minéraux présents dans le minerai d'Elliot Lake.

*Agent scientifique sénior, **Chef, Section de l'hydrométallurgie, ***Agent scientifique, Division de la métallurgie extractive, ministère de l'Energie, des Mines et des Ressources, Ottawa, Canada.

CONTENTS

	<u>Page</u>
Abstract	i
Resumé	ii
Introduction	1
Procedure	2
Results	5
Effect of Particle Size	5
Effect of Initial Iron Concentration in Leach Liquor	10
Effect of Nutrient Strength	13
Effect of Miscellaneous Variables	13
Bacterial Leaching of Flotation and Sink-Float Products ..	15
Mineralogical Study	16
Discussion	19
Conclusions	21
Acknowledgements	21
References	22

==

FIGURES

<u>No.</u>		<u>Page</u>
1.	Column Leaching Apparatus	3
2.	Effect of Particle Size on Uranium and Iron Extractions, and on pH	7
3.	Effect of Particle Size on Uranium Extraction	8
4.	Effect of Initial Iron on Uranium Extraction, and on Rates of Iron and Acid Formation	11
5.	Relationship of Initial Iron to Selected Uranium Extractions	12
6.	Effect of Nutrient on Uranium Extraction	14
7.	Photomicrographs of Polished Sections From Bacterial Leach Feeds	17
8.	Photomicrographs of Polished Sections From Bacterial Leach Residues	18

TABLES

1.	Analyses of Leach Feed Materials	4
2.	Summary of Leach Test Results	6

INTRODUCTION

Earlier work⁽¹⁾, done with the object of explaining the previously observed leaching of uranium from broken ore in the underground workings of mines in the Elliot Lake area, showed that mine water and stope ore from Denison Mines Limited contained bacteria belonging to the Ferrobacillus-Thiobacillus group. It was also demonstrated by these earlier studies that the bacteria promoted the production of acidic oxidizing solutions, containing ferric iron, which are known to be effective in leaching the uranium minerals present in the Elliot Lake ores. In the absence of bacteria no leaching of the uranium occurred. It was also indicated by the results obtained that the rate at which the uranium was extracted was related to the particle size of the ore and to the iron and acid concentrations of the leaching solutions.

The results of the preliminary work were in agreement with the literature describing the behaviour and characteristics of the Ferrobacillus-Thiobacillus group⁽²⁾. Investigators had shown that these bacteria derive their energy by oxidizing ferrous iron to ferric iron in an oxygen-bearing aqueous medium. It had also been shown that these bacteria have an optimum growth rate in solutions having a pH range of 2.0 to 4.0 and a temperature range of 25 to 35°C, and containing certain concentrations of nitrogen, phosphorus, potassium, magnesium and calcium which are required as nutrients.

In view of the preliminary studies of the effect of bacteria on the leaching of the Denison ore, and the information given in the literature regarding the most desirable environment for the bacteria, it was decided to study in further detail the relationship of particle size, iron concentration, acidity and nutrient strength on the rate at which uranium is extracted from the ore. Since it was known from previous plant experience that sulphuric acid leaching of the Elliot Lake ores produced solutions containing calcium and magnesium in concentrations at least equal to those which were found to be optimum for bacterial growth, it was decided to add only nitrogen, potassium and phosphorus in this work, in the form of ammonium sulphate and potassium orthophosphate.

The ore used in this investigation was fresh unoxidized ore from Denison Mines Limited which was typical of the ore from the Elliot Lake area in that it was a conglomerate composed of quartz pebbles and a quartz-sericite matrix^(3,4). The uranium-bearing minerals, which were mainly brannerite and uraninite, were disseminated throughout the matrix, as were the sulphide minerals pyrite and pyrrhotite.

In this bulletin a description will be given of the work done to resolve the effects of particle size, and of iron, acid and nutrient concentrations, on the leaching rate of uranium from an Elliot Lake area ore in the presence of Ferrobacillus-Thiobacillus bacteria. Test work done to investigate the possibility of applying bacterial leaching to flotation tailings and sink-float rejects from preconcentration tests done on Elliot Lake ores is discussed. Finally, mineralogical studies done to determine what minerals are attacked by the leaching medium, and to determine the depth of penetration of leaching solutions into the ore particles, will also be described.

PROCEDURE

A photograph of the equipment used for leaching the ore in this work is shown in Figure 1. When the apparatus was used, 400 g of fresh, unoxidized, sized ore was placed in the glass column and 600 ml of leaching solution was added to the solution reservoir located under the column. The leaching solution was air-lifted at a rate of about 10 ml/min to the top of the column, from where it percolated through the ore and back to the reservoir. This system ensured that the leaching solution was well aerated before being exposed to the ore.

Leach tests were done on samples of mine ore, flotation tailings and sink-float rejects. The ore was crushed to minus 1 inch. For this work, four size ranges were leached: minus 1 inch plus 4 mesh, minus 4 mesh plus 8 mesh, minus 8 mesh plus 14 mesh, and minus 14 mesh plus 20 mesh. The flotation tailings samples were deslimed prior to leaching and the slimes were discarded. The sink-float rejects, which were crushed to minus 1 inch as received, were crushed further to minus 4 mesh prior to leaching. The chemical analyses of all the leach feeds used are shown in Table 1.

The leaching solutions were all aqueous solutions of sulphuric acid having a pH of about 2.0 initially. To these acid solutions were added various amounts of ferrous sulphate, ammonium sulphate and potassium orthophosphate. In most of the tests, Ferrobacillus-Thiobacillus culture grown from a sample of Denison mine water was added to the leach solution. In three tests, used as controls, no bacteria were added.

The tests were continued for periods of up to thirty-three weeks. The progress of the leaching was followed by taking weekly samples of the recycling leach solution for uranium and iron analyses. At the completion of the tests the solids were washed with pH 2.0 sulphuric acid solution and water, and analysed for uranium content.

RESULTS
TABLE I

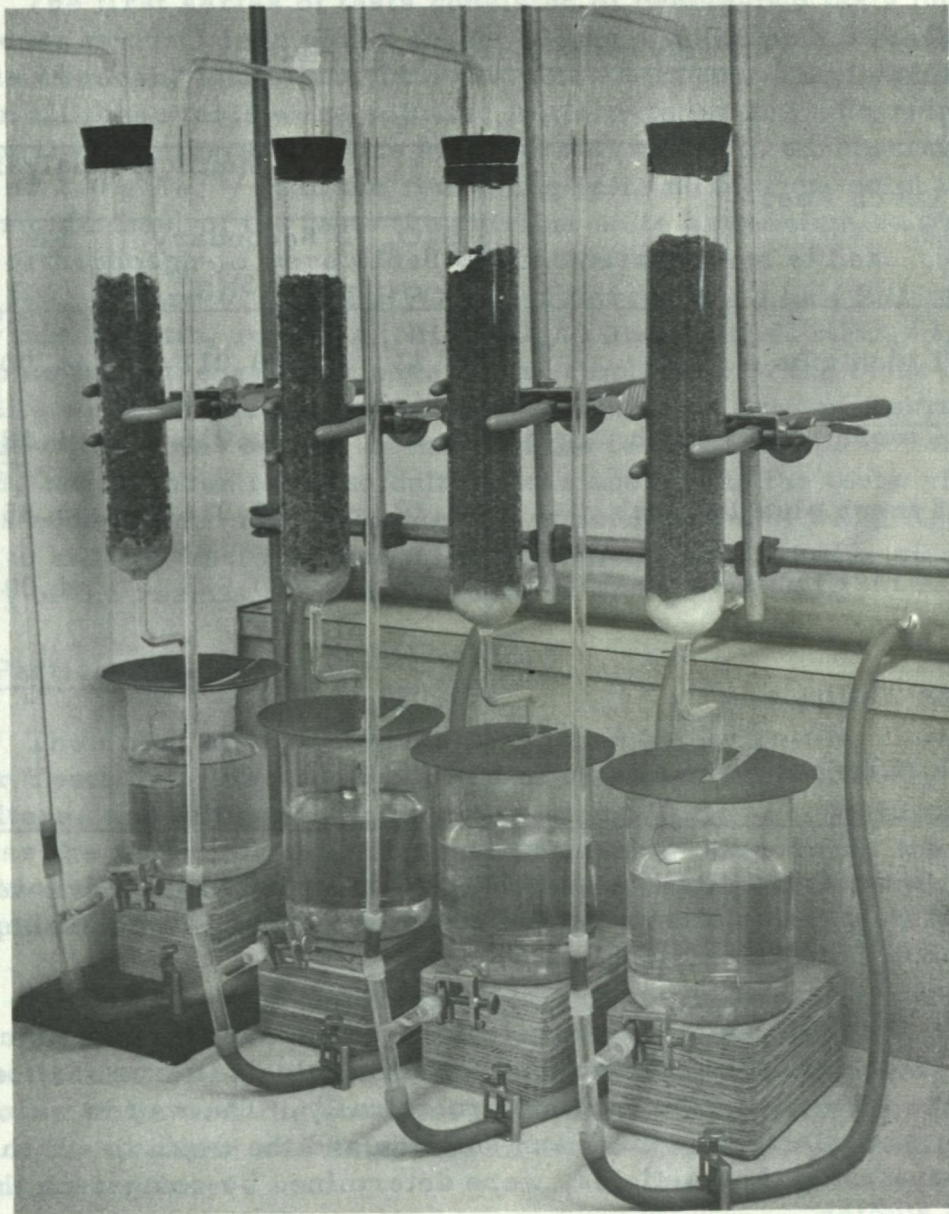


Figure 1. Column Leaching Apparatus

A further analysis of the effect of particle size on uranium extraction was made. The extraction curves for particles of 20, 40, 60, 80, 100, 150, 200, 300, 400, 600, 800, 1000, and 2000 mesh are shown in Figure 2. The curves for particles of 20, 40, 60, 80, 100, 150, and 200 mesh are linear with a slope of about 0.65 to the point of maximum extraction. The curves for particles of 300, 400, 600, 800, and 1000 mesh are linear with a slope of about 0.55 to the point of maximum extraction. The curve for particles of 2000 mesh is linear with a slope of about 0.50 to the point of maximum extraction. With the two intermediate sizes, the extraction curves are made up of two linear sections, one with a slope of about 0.65 up to the fifth week, and one having a slope of about 0.50 from the fifth week to the end of the experiment. The extraction curves for particles of 20, 40, 60, 80, 100, 150, and 200 mesh are linear with a slope of about 0.65 to the point of maximum extraction. The curves for particles of 300, 400, 600, 800, and 1000 mesh are linear with a slope of about 0.55 to the point of maximum extraction. The curve for particles of 2000 mesh is linear with a slope of about 0.50 to the point of maximum extraction.

TABLE 1
Analyses of Leach Feed Materials

Leach Feed	Chemical Analysis, %			
	U ₃ O ₈	Secondary U ₃ O ₈ *	Fe	S
Minus 1 inch plus 4 mesh	0.11	0.011	4.00	2.83
Minus 4 mesh plus 8 mesh	0.11	0.015	4.48	3.17
Minus 8 mesh plus 14 mesh	0.12	0.016	3.85	2.87
Minus 14 mesh plus 20 mesh	0.15	0.023	4.09	3.08
Flotation Tailings	0.027	-	-	0.03
Sink-Float Rejects	0.05	-	-	0.61

* This is the amount of U₃O₈ as hexavalent uranium that dissolved in hot Na₂CO₃ solution, and indicates that about 13% of the contained uranium is in an oxidized state.

Samples of the leach material from two of the tests done on mine ore were made into polished sections for mineralogical examination. Samples of fresh ore were also examined mineralogically. The way in which the minerals were attacked by the leach solution, and the depth to which the solution penetrated the particles, were determined by comparing the results of the two studies.

It is known from the literature that a temperature of 25 to 35°C is the optimum range for bacterial activity⁽²⁾, and that any deviation above or below this level would be detrimental to their growth. For this reason, the test work was carried out at an ambient temperature of 28°C.

RESULTS

The first series of tests consisted of twenty-two tests on the four size fractions, minus 1 inch plus 4 mesh, minus 4 mesh plus 8 mesh, minus 8 mesh plus 14 mesh, and minus 14 mesh plus 20 mesh. The leaching solutions in all these tests were adjusted initially to pH 2.2 with sulphuric acid. In most of the tests, ferrous sulphate was added to concentrations of either 0.5 or 1.0 g Fe^{++}/l ; in the remaining tests no ferrous sulphate was added. Also, in most of the tests the nutrient salts ammonium sulphate and potassium orthophosphate were added to concentrations of either 1.5 g $(\text{NH}_4)_2\text{SO}_4/\text{l}$ and 0.25 g $\text{K}_2\text{HPO}_4/\text{l}$, referred to as half full-strength nutrient in this bulletin, or 3.0 g $(\text{NH}_4)_2\text{SO}_4/\text{l}$ and 0.5 g $\text{K}_2\text{HPO}_4/\text{l}$, referred to as full-strength nutrient. In the remainder of the tests no nutrient salts were added. In sixteen of the tests one drop of Ferrobacillus-Thiobacillus culture was added, in three of the tests six drops of culture were added, and in three tests no culture was added. In the tests where no culture was added, mercuric chloride to a concentration of 0.2 g HgCl_2/l was added to kill any bacteria that might have been introduced accidentally. A summary of the results of all these tests is given in Table 2.

Effect of Particle Size

The effect of particle size is typified by the results shown in Table 2 for Tests 3, 8, 13 and 18. In all of these tests 1.0 g Fe^{++}/l , one drop of culture, and full-strength nutrient were added, so that the only variable was the particle size. The rate of uranium extraction, the rate of change of iron concentration, and the pH of the leaching solution are all shown graphically in Figure 2.

It can be seen from Figure 2 that the overall rate of uranium extraction increases as the particle size decreases. For the finest size investigated (minus 14 mesh plus 20 mesh), 90% of the uranium was extracted in four weeks, while with the coarsest size (minus 1 inch plus 4 mesh), only about 70% of the uranium was extracted in thirty-two weeks, at which time the test was discontinued.

A further analysis of the effect of particle size on uranium extraction is shown in Figure 3, where the extraction is plotted against the logarithm of the leaching time. With minus 1 inch plus 4 mesh ore, the extraction-log t curve is linear with a slope of 0.36, showing that the extraction rate was constant throughout the leaching period of from one to thirty-two weeks. With the two intermediate sizes, the extraction-log t curves are made up of two linear sections, one with a slope of about 0.64 up to the fifth week, and one having a slope of about 0.30 beginning at the fifth week. With the finest size (minus 14 mesh plus 20 mesh), the extraction-log t curve is linear with a slope of 0.65 to the point of maximum extraction

TABLE 2
Summary of Leach Test Results

Test No.	Leach Feed Size (anal.)	Initial Leach Conditions			* Intermediate Results												* Final Results		
		Culture	g Fe/l	Nutrient	2nd Week			5th Week			10th Week			15th Week			Week	Residue, % U ₃ O ₈	U ₃ O ₈ Ext. %
					pH	** g Fe/l	U ₃ O ₈ Ext. %	pH	** g Fe/l	U ₃ O ₈ Ext. %	pH	** g Fe/l	U ₃ O ₈ Ext. %	pH	** g Fe/l	U ₃ O ₈ Ext. %			
1		1 drop	0.0	F.S.	2.50	0.04	13.7	2.30	0.10	19.6	2.58	0.24	24.9	1.78	0.64	34.0	21		
2	-1"+4m (0.11% U ₃ O ₈)	"	0.5	F.S.	2.42	0.63	35.7	2.15	0.78	45.2	2.28	1.02	51.4	1.80	1.31	54.6	23	0.038	65.4
3		"	1.0	F.S.	2.65	1.28	37.1	2.30	1.28	52.0	2.20	1.04	54.5	1.80	1.01	57.8	32	0.028	74.6
4		"	0.5	1/2F.S.	2.40	1.15	28.8	2.20	0.87	35.3	2.25	0.64	35.6	2.20	0.60	47.9	17	0.054	50.9
5		6 drops	0.5	F.S.	2.20	0.64	24.7	2.18	0.70	32.4	2.30	0.69	40.7	--	--	--	11	0.055	50.0
6			1 drop	0.0	F.S.	2.70	0.06	19.3	2.80	0.12	30.9	2.50	0.45	49.2	1.80	1.36	66.2	25	0.005
7	-4+8m (0.11% U ₃ O ₈)	"	0.5	F.S.	2.80	0.69	38.6	2.50	0.79	50.8	2.15	1.21	71.4	1.80	2.00	80.7	19	0.005	95.6
8		"	1.0	F.S.	2.95	1.05	42.7	2.48	0.85	69.2	2.28	0.98	81.4	1.70	1.42	84.4	18	0.003	97.3
9		"	1.0	none	2.50	0.87	52.2	2.32	0.92	66.2	2.30	0.93	68.0	2.15	1.21	72.6	17	0.015	86.4
10		6 drops	1.0	none	2.45	1.02	53.6	2.18	1.30	67.8	2.00	2.26	73.7	1.75	4.40	81.8	17	0.006	94.6
11			1 drop	0.0	F.S.	3.30	0.04	23.9	3.00	0.50	44.9	2.12	1.03	67.3	1.80	3.10	85.2	21	0.003
12	-8+14m (0.12% U ₃ O ₈)	"	0.5	F.S.	2.70	0.60	49.1	2.15	1.30	88.3	1.62	3.89	91.6	1.58	5.40	94.2	33	0.003	97.5
13		"	1.0	F.S.	2.52	0.98	52.9	2.08	1.50	78.2	1.55	4.18	82.8	1.45	5.90	88.6	31	0.003	97.5
14		"	0.5	1/2F.S.	2.65	0.64	61.9	2.80	0.86	89.6	1.98	1.70	87.8	1.67	3.44	92.5	16	0.004	96.7
15		6 drops	0.5	1/2F.S.	2.60	0.54	55.2	1.95	1.80	81.8	1.55	4.90	91.4	1.47	6.60	92.6	17	0.003	97.5
16			1 drop	0.0	F.S.	2.75	0.73	51.3	2.10	1.57	93.2	1.40	5.65	95.9	1.32	8.30	95.6	16	0.005
17	-14+20m (0.15% U ₃ O ₈)	"	0.5	F.S.	2.60	0.66	61.4	2.00	1.65	90.5	1.40	6.02	99.1	1.30	8.60	99.1	15	0.004	97.4
18		"	1.0	F.S.	2.40	0.80	74.6	1.80	2.26	96.0	1.45	5.84	99.0	1.32	8.00	99.0	15	0.002	98.7
19		"	0.5	none	2.45	0.77	85.7	2.18	0.88	90.4	2.03	1.40	95.8	1.85	2.49	97.2	15	0.003	98.0
20	-1"+4m	none	0.0	F.S.	2.30	0.07	4.50	2.30	0.05	3.04	2.50	0.07	2.26	2.50	0.06	2.00	15	0.11	
21	-4+8m	"	0.0	F.S.	2.80	0.03	0.64	2.30	0.08	3.10	2.55	0.09	2.61	2.70	0.08	2.52	15	0.11	
22	-8+14m	"	0.0	F.S.	2.50	0.06	1.39	2.45	0.08	4.42	2.72	0.09	5.62	2.90	0.09	8.58	15	0.11	8.34

* Extractions under Intermediate Results were based on total uranium content of solutions and leach feed. Extractions under Final Results were based on uranium analyses of leach feed and washed residue.

** In tests with culture, iron was 95% ferric; and in tests with no culture, iron was more than 70% ferrous.

F.S.- full strength.

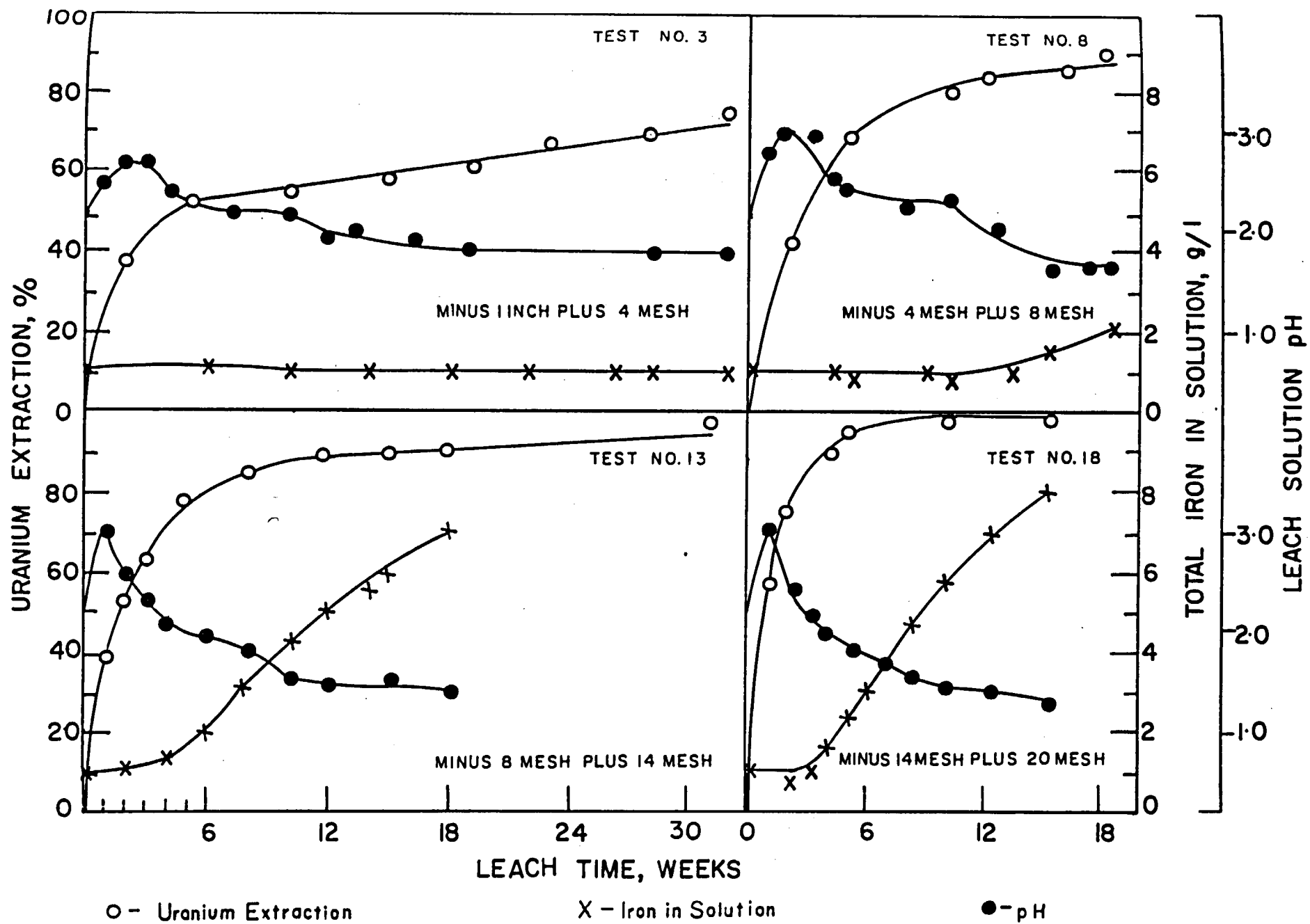


Figure 2. Effect of Particle Size on Uranium and Iron Extractions, and on pH

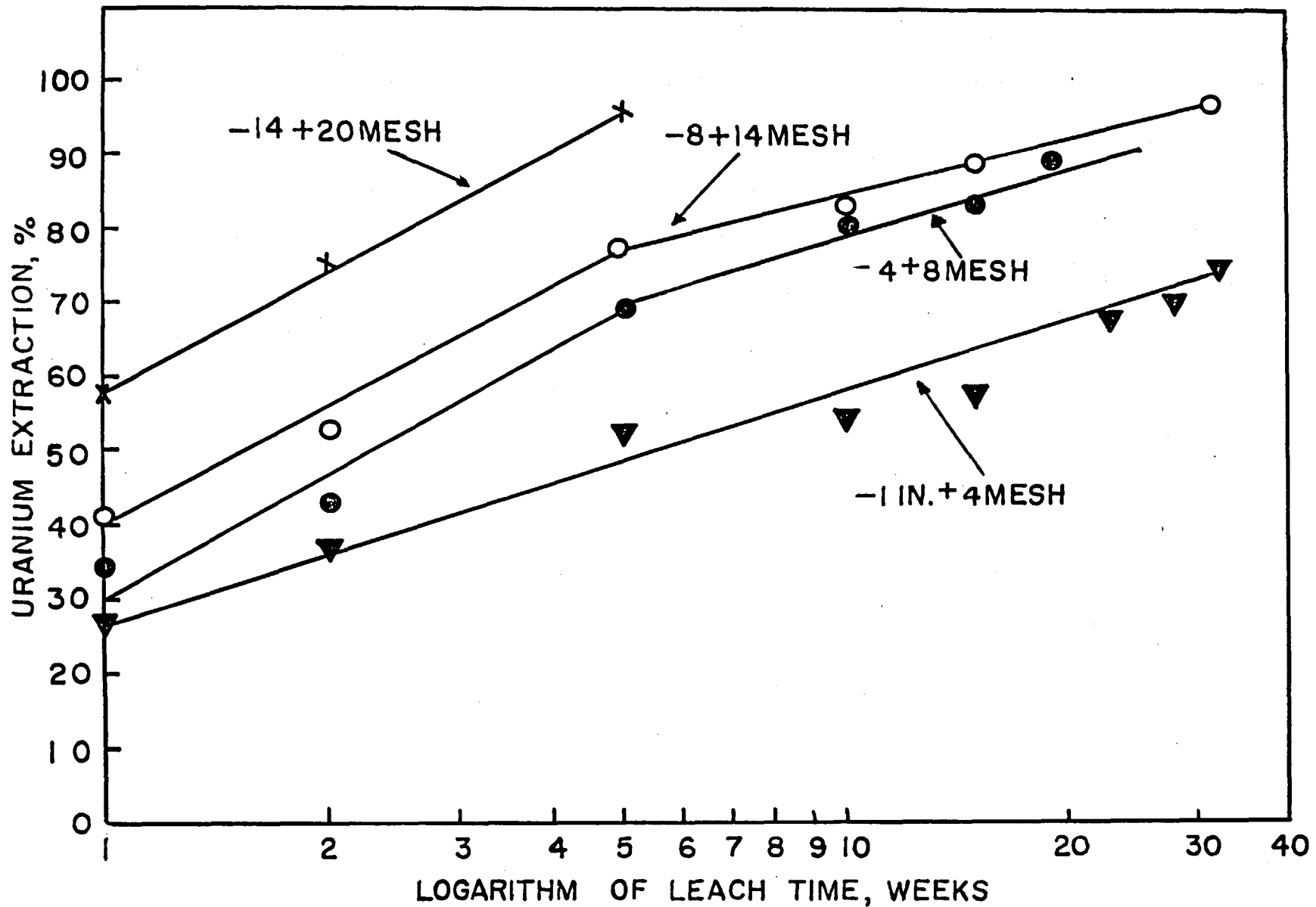


Figure 3. Effect of Particle Size on Uranium Extraction

after five weeks of leaching. These results show that, depending on the particle size and elapsed leaching time considered, one of two extraction rates is observed. It is suggested that the higher of these two rates is determined mainly by the rate at which uranium is being leached from the surface (or close to the surface) of the ore particles, while the slower of these two rates is related to the rate at which the solutions can penetrate the particles to depth. If this assumption is valid, it is indicated that, with particles coarser than 4 mesh, the rate of solution penetration controls the uranium extraction rate almost from the start of leaching, whereas the penetration rate has a minor effect on the extraction rate with particles less than 14 mesh. With sizes between 4 and 14 mesh, surface leaching is the controlling rate initially and the penetration effect assumes control after five weeks. The effect of particle size is to determine which of the two leaching rates predominates. In the sizes coarser than 4 mesh, where the slower rate predominates almost from the start of leaching, the time required to reach 90% extraction would be measured in years.

It can also be seen from Figure 2 that the rate of acid production, as indicated by the pH curves, and the rate of iron dissolution, as indicated by the iron concentration curves, are higher in the tests done on the finer particles. The relationship between rate of iron dissolution, acid formation and particle size is probably also due to the surface-penetration effects previously discussed with regard to uranium extraction rates. The pH values of the leaching solutions increased during the first one to three weeks of the tests and then decreased to a minimum of pH 1.5 to 1.8 at about the fifteenth week. This indicates that the acid is being used up faster than it can be produced at the beginning of the test, possibly due to the reaction of the acid with carbonates and other acid-consuming minerals in the ore, but an equilibrium condition between acid formation and consumption is eventually reached.

In the tests shown in Figure 2, the uranium concentrations of the leach liquors increased with time to values ranging from about 0.5 to 1.0 g U_3O_8/l , and the pH values dropped to as low as pH 1.4 in the tests on the fine sizes. It is interesting to note that the bacteria were able to survive in this environment, since there had been some doubts that the bacteria would live in a solution having a pH value of less than 2.0 and containing an appreciable heavy-metal concentration. The presence of bacteria in these solutions is indicated by the fact that the iron in the solution remained in the ferric state, and also by the fact that uranium extraction continued to take place, conditions that would not exist if bacterial action had ceased⁽¹⁾.

Although Tests 3, 8, 13 and 18 were used in compiling Figure 2, the effect of particle size could have been demonstrated equally well by the results of Tests 2, 7, 12 and 17 or Tests 1, 6, 11 and 16. In these last two groups of tests, the initial iron concentration of the first group was 0.5 g $\text{Fe}^{++}/1$, and in the second group no Fe^{++} was added initially. Full-strength nutrient was used in all eight tests. The only variable within each of these groups was the particle size. The results obtained in Tests 3, 8, 13 and 18 were used for Figure 2, since this group of tests produced the highest extraction rates for any given size, presumably due to the initial iron concentrations used in these tests.

Effect of Initial Iron Concentration in Leach Liquor

The effect of the initial iron concentration on the uranium extraction rate, and on the rates of acid formation and iron dissolution for the ore, is shown in Figures 4 and 5, in which the data from Tests 6, 7 and 8 of Table 2 are plotted. In all of these three tests, an ore particle size range of minus 4 mesh plus 8 mesh, one drop of culture, and full-strength nutrient were used. The only variable within the test was the initial iron concentration, which ranged from 0 to 1.0 g $\text{Fe}^{++}/1$.

Figure 4(c) shows that as the initial iron concentration was increased from 0 to 0.5 to 1.0 g $\text{Fe}^{++}/1$, the overall uranium extraction rate was increased proportionally. The proportionality of this relationship is shown in Figure 5, where the times taken to reach 50, 70 and 90% uranium extractions are plotted against the initial iron concentration. Since all of these graphs are roughly linear and of similar slope, it appears that the time required to reach a given uranium extraction is directly proportional to the initial iron concentration, and the effect of the initial iron concentration persists throughout the test. Since the bacteria require the presence of soluble iron as an energy source, and since some of the chemical reactions involved in the decomposition of pyrite and the dissolution of uranium minerals depend on the presence of ferric iron, it is reasonable to conclude that the effect of higher initial iron concentrations is to increase bacterial growth and chemical reaction rates initially. The importance of the high initial extraction rate is shown by the fact that the gain made initially, where 1.0 g $\text{Fe}^{++}/1$ was added, was maintained to 90% extraction; this in spite of the fact that the iron and acid concentrations in all three tests were approximately 0.8 to 1.5 g $\text{Fe}/1$ and pH 2.0 by about the twelfth week. The evidence of the above results suggests that initial iron concentrations of more than 1.0 g $\text{Fe}^{++}/1$ would result in still greater overall rates of extraction.

The importance of having some soluble iron present in the leach liquor at the start of leaching is also shown by a comparison of Tests 1, 2 and 3 of Table 2, done on the minus 1 inch plus 4 mesh fraction. In the tests on the two finest sizes the effect of the initial iron concentration on the

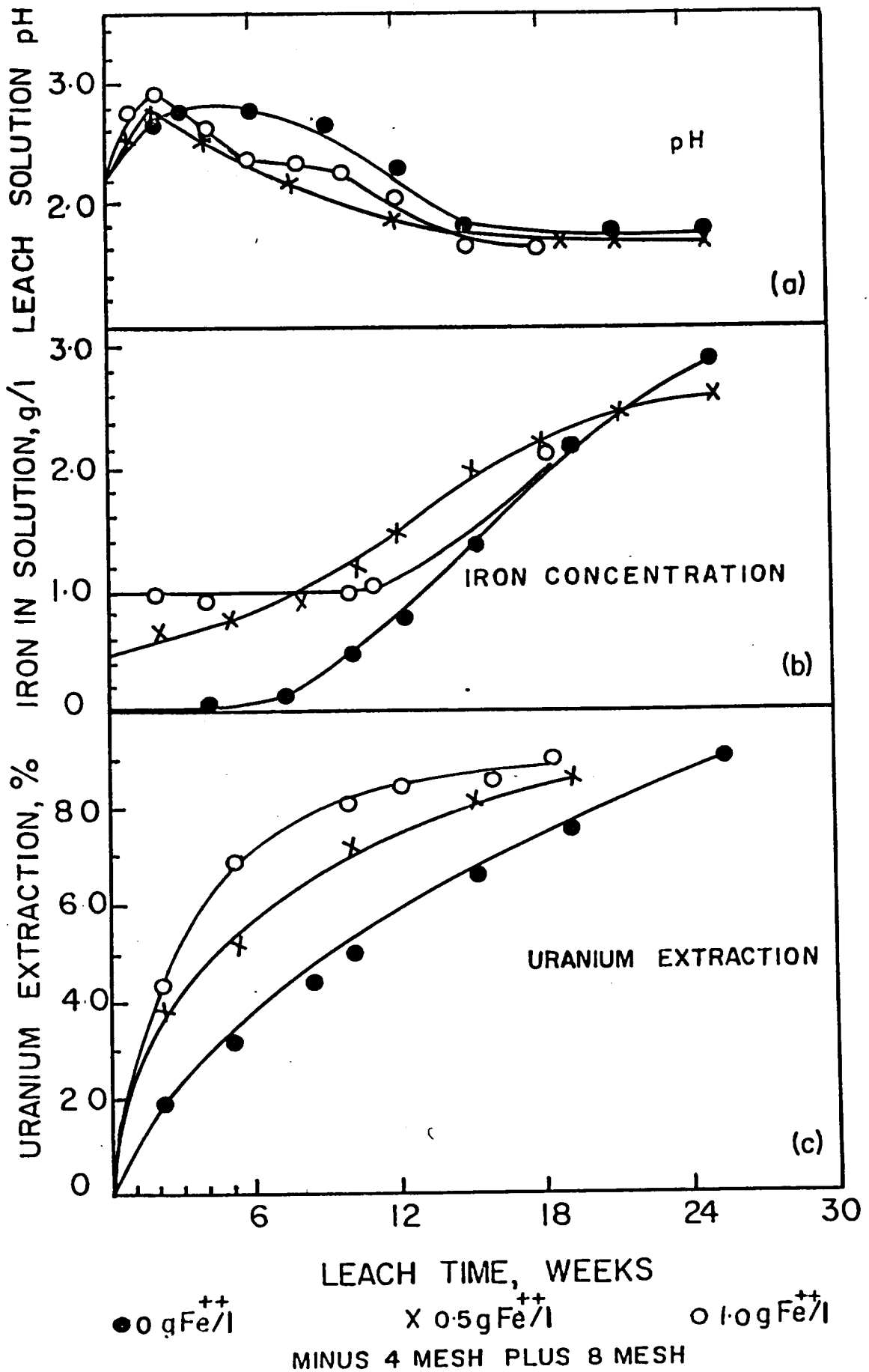


Figure 4. Effect of Initial Iron on Uranium Extraction, and on Rates of Iron and Acid Formation

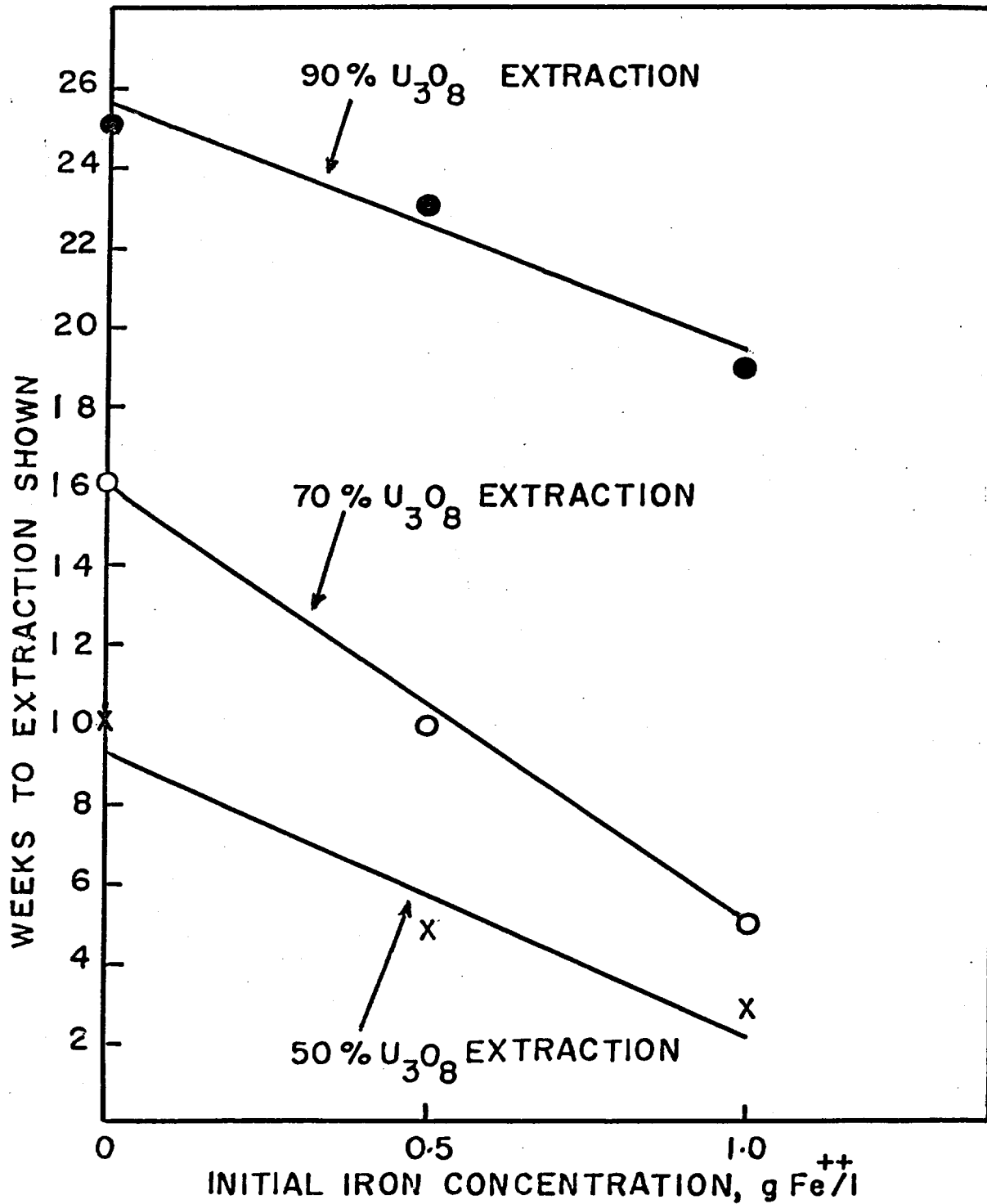


Figure 5. Relationship of Initial Iron to Selected Uranium Extractions

uranium extraction rate was less pronounced (Tests 11, 12 and 13, and Tests 16, 17 and 18), presumably because the rapid dissolution of iron which occurs when the finer sizes are leached obviates the need to add iron initially to the leach.

Effect of Nutrient Strength

The effect of adding nitrogen, phosphorus and potassium as $(\text{NH}_4)_2\text{SO}_4$ and K_2HPO_4 as nutrients for the bacteria is shown by the graphs in Figure 6. With the sizes coarser than 8 mesh, the addition of full-strength nutrient increased the overall extraction rate in comparison with the rate obtained when half full-strength nutrient or no nutrient was added. On sizes finer than 8 mesh, the addition of nutrients did not appear to have any significant effect on the overall uranium extraction rate. These results suggest that only when bacterial leaching is applied to ore crushed to coarser than 8 mesh would there be any advantage in adding nutrient salts.

Effect of Miscellaneous Variables

Culture of the Ferrobacillus-Thiobacillus group was added to the leach solutions at the start of each of nineteen tests: one drop of the culture was added to sixteen tests, and six drops were added to three tests. Only in one instance, where minus 4 mesh plus 8 mesh ore was leached in a solution with no nutrient salts added but having an initial iron concentration of $1.0 \text{ g Fe}^{++}/\text{l}$, did the addition of six drops of culture appear to result in an improved uranium extraction over that obtained when one drop was added (Tests 9 and 10 of Table 2). The other tests in which six drops of culture were added did not result in significantly higher uranium extractions (Tests 4 and 5, and Tests 14 and 15, Table 2). On the basis of these results, there is no conclusive evidence that the amount of bacteria added at the beginning of a test has any significant effect on the uranium extraction.

In three of the tests (Tests 20, 21 and 22, Table 2), no bacteria culture was added and $0.2 \text{ g HgCl}_2/\text{l}$ was added to kill any bacteria which might have been present. In these tests the uranium extractions were all less than 9.0% after fifteen weeks of leaching. The concentration of iron in the liquor in these control tests was less than $0.15 \text{ g Fe}/\text{l}$, almost entirely in the ferrous state. These results confirm those obtained in the earlier work⁽¹⁾, where it was shown that the presence of bacteria was essential for uranium and iron leaching to take place and for the iron to be oxidized rapidly to the ferric state.

In four tests, not shown in Table 2, $0.12 \text{ g}/\text{l}$ of the wetting agent, Tween 21, manufactured by the Atlas Powder Company, Brantford, Canada, was added at the beginning of the test to determine whether its presence would increase the leaching rate by making it easier for the solution to penetrate the rock. In none of these tests was the Tween addition beneficial.

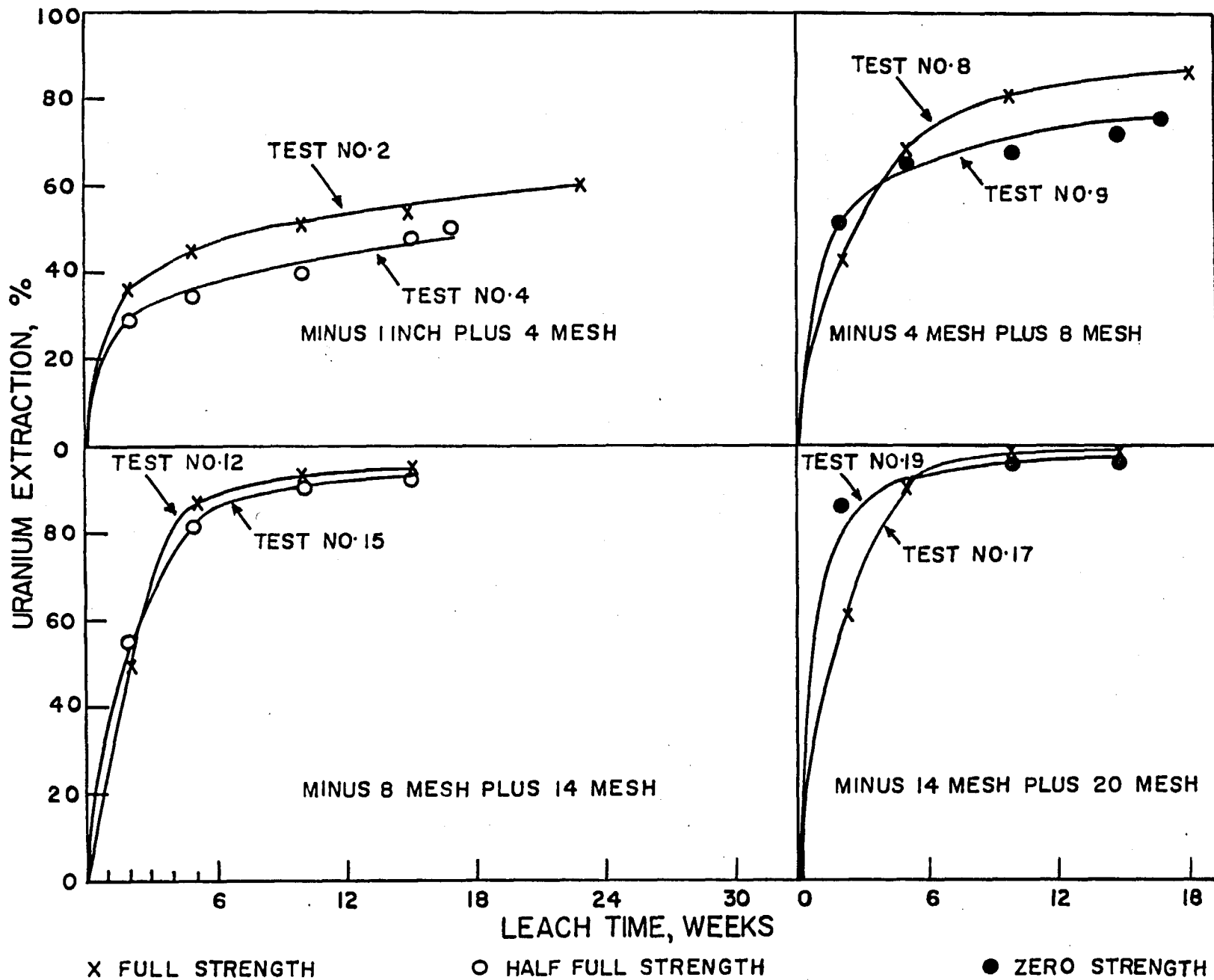


Figure 6. Effect of Nutrient on Uranium Extraction

In several of the tests the leaching solution was removed after eighteen to twenty weeks of leaching and replaced with a fresh solution and fresh culture, to see whether this procedure would increase the uranium extraction rate, which slowed down after eighteen weeks. This procedure was not effective, showing that the reduced extraction rate was not due to impurity build-up or to lack of bacteria in the original solution.

Bacterial Leaching of Flotation and Sink-Float Products

In studying methods for treating the Elliot Lake ores, one of the processes that has been considered involves the preconcentration of the ores by either flotation or sink-float procedures prior to acid leaching of the concentrate. The main disadvantage of this approach has been that the tailings losses have been too high for the method to be seriously considered. Consequently, some bacterial leach tests were done on flotation tailings and sink-float rejects to determine whether or not bacterial leaching could be used to recover the uranium lost in the preconcentration step.

The leach feeds for this part of the study were flotation tailings from a previous pilot plant flotation investigation done at the Mines Branch⁽³⁾, and float products from bench scale sink-float tests. In the flotation work, the concentrate contained 88.1% of the uranium in the ore in about 40% of the weight, leaving 11.9% of the uranium in the tailings used for bacterial leach feed. In the sink-float concentrate 76.4% of the uranium was recovered in about 35% of the weight, leaving 23.6% of the uranium in 65% of the weight for bacterial leach feed. It was found necessary, in treating the flotation tailings, to remove the slimes in order to obtain a satisfactory percolation rate in the columns; this step resulted in a loss of 26% of the uranium in the tailings, equivalent to 3.2% of the overall uranium contained in the preconcentration feed. In all of the bacterial leach tests done on these products, an initial iron concentration of 1.0 g Fe^{++} /l was used and full-strength nutrient was added. In the tests on the sink-float rejects, the ore was crushed to minus 4 mesh.

Since the flotation tailings contained only 0.03% S, bacterial leach tests were conducted on these tailings, both as received and with 6% pyrite added. The results of these tests showed that, when pyrite was not added, 70% of the uranium was extracted in twenty weeks; while, in the test where pyrite was added, 90% of the uranium was extracted in eleven weeks. The results of a test in which 12% pyrite was added showed that neither the uranium extraction rate nor the overall extraction was increased over those obtained when 6% pyrite was added. Assuming that conventional acid leaching of the flotation concentrate would recover 95% of the uranium contained in it, the overall recovery of uranium by preconcentration, acid leaching of the concentrate and bacterial leaching of the flotation tailings would be about 92%.

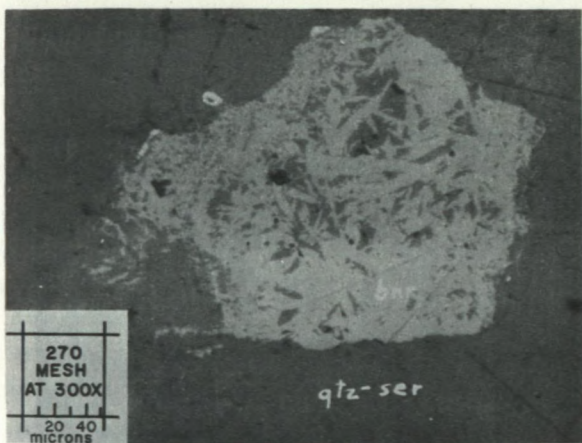
Since the float rejects from the sink-float test contained 0.6% S, no pyrite was added to the bacterial leach tests on this product. The results showed that only 42% of the uranium had been extracted in 29 weeks of bacterial leaching. Since only 75% of the uranium was recovered in the sink fraction, the bacterial leach extraction would have to be about 80% for the overall recovery of the combined methods to be attractive. Although the addition of pyrite to the bacterial leach feed might improve the extraction obtained in bacterial leaching, it is doubtful that the extraction could be increased to the required 80%, since the low extraction is probably largely due to the coarseness of the leach feed (minus 4 mesh). This latter observation is based on the results obtained in leaching the coarser fractions of the whole ore (Figure 2).

Mineralogical Study

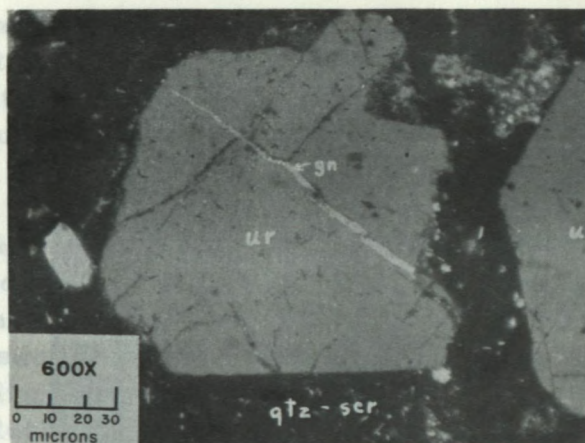
Samples of the residues for Tests 7 and 12 of Table 2, along with the samples of the feed material used in these tests, were examined mineralogically to determine how the minerals were affected by the leaching⁽⁴⁾. The sample of residue from Test 7 was taken after nineteen weeks when the uranium extraction was about 85%, while the sample from Test 12 was taken after thirty-three weeks when the uranium extraction was 94%. Particle size range in Test 7 was minus 4 mesh plus 8 mesh, and that of Test 12 was minus 8 mesh plus 14 mesh. Photomicrographs of polished sections of the feed samples are shown in Figure 7, and of the residues are shown in Figure 8.

Microscopic study of the feed materials showed the sample to be typical of the Elliot Lake ores^(3,4) containing the radioactive minerals uraninite, brannerite, monazite and coffinite, and the sulphide minerals pyrite, pyrrhotite, chalcopyrite and galena (Figure 7). Of the radioactive minerals identified in the feed samples, uraninite and brannerite appear to be completely altered, while monazite and coffinite remain unchanged in both residues. In no section was there any indication of a differential leaching effect, regardless of the position of the radioactive particle in the grain of ore. It appeared, therefore, that the leaching solution had completely penetrated the grains of ore in both residues.

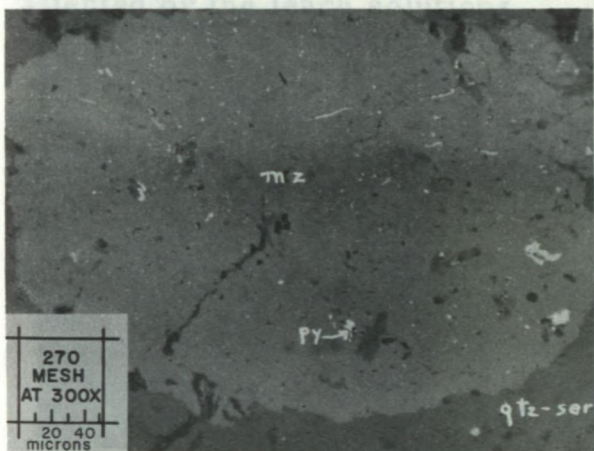
The apparent absence of uraninite in the residues may be due to (a) complete dissolution or to (b) dissolution of most of the exposed uraninite and the oxidation of the remainder to uranic oxide, UO_3 , which is amorphous. Brannerite, too, has apparently dissolved or decomposed, since its presence could not be confirmed after nineteen weeks. Since brannerite is commonly intergrown with anatase or rutile in the ore conglomerate, the occurrence of radioactivity in leach residue grains which contain anatase or rutile strongly suggests that these grains originally contained brannerite (Figure 8 (a)). Although X-ray diffraction patterns of these grains do not show brannerite lines, it is possible that either too small a proportion of residual brannerite remains therein to permit its detection, or the grains contain an amorphous uranium compound derived from brannerite.



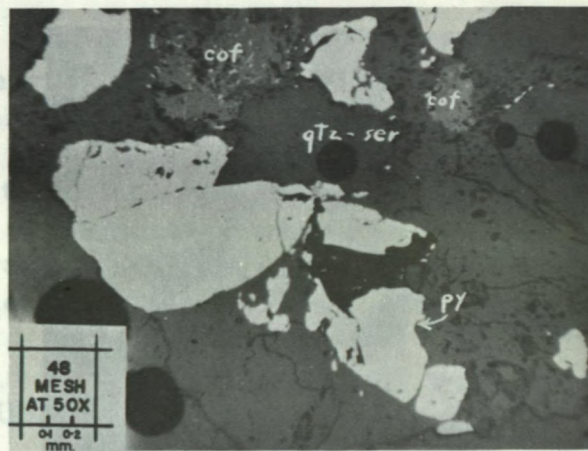
(a)



(b)



(c)



(d)

Figure 7. Photomicrographs of Polished Sections From Bacterial Leach Feeds

- (a) A dense aggregate of lath-like crystals of brannerite (bnr) in a grain of quartz-sericite (qtz-ser).
- (b) Crystals of uraninite (ur) in a grain of quartz-sericite. Galena (gn) fills a fracture in one of the uraninite crystals.
- (c) A rounded particle of monazite (mz) in a quartz-sericite grain contains minute particles of pyrite (py).
- (d) Irregular particles of coffinite (cof) in a quartz-sericite grain with minute white specks of pyrite. Coarse particles of pyrite also occur in the quartz-sericite.

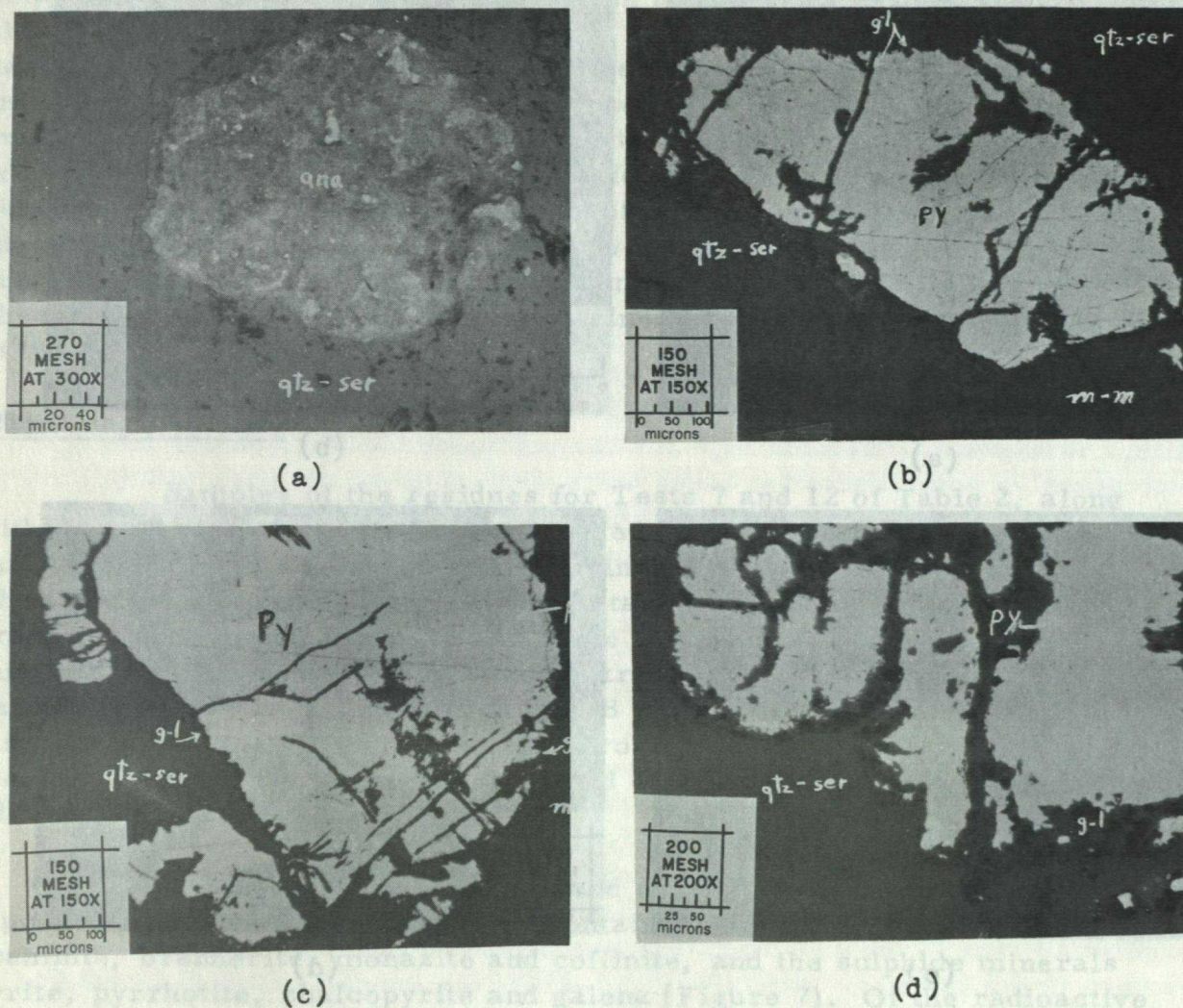


Figure 8. Photomicrographs of Polished Sections From Bacterial Leach Residues
19 Week Residue (Test 7)

- (a) A radioactive particle of anatase (ana) in a grain of quartz-sericite (qtz-ser).
- (b) A particle of pyrite (py) surrounded and penetrated by a grey layer (g-l) in a quartz-sericite grain. Mounting medium (m-m) is shown.
- (c) A particle of pyrite occurring at the edge of a quartz-sericite grain and showing more intensive alteration on exposed side of grain.

33 Week Residue (Test 12)

- (d) Alteration product (g-l) surrounds and penetrates a fractured crystal of pyrite in a quartz-sericite grain.

Monazite appears to be unaffected by the leach solutions. Whether the uranium silicate, coffinite, has been affected is doubtful, but it is so scarce in the leach feed that its apparent absence in the residues leaves this question open. The uranium appears to have been removed from the hydrocarbon thucholite.

Pyrite is the main sulphide mineral present in the leach feed and appears to have been the most affected by the leach solutions (Figure 8 (b), (c) and (d)). The few pyrrhotite grains observed in the residues show evidence of pitting. Jagged edges of such grains indicate that particles of pyrrhotite may have broken away during the polishing procedure. If the pyrrhotite had been leached by the solution, the edges of the pyrrhotite grains would have appeared rounded. There are no residual rims or pitted zones around grains of chalcopyrite or galena, showing that they were not affected by the leach solutions.

The leaching solution appears to have attacked the outer surfaces of all pyrite particles, entered and enlarged fractures and cavities, and left behind a layer of soft grey material. This layer is several times thicker in the thirty-three-week residue than in the nineteen-week residue, indicating that the leaching solution continues to decompose the pyrite for a period of up to thirty-three weeks. X-ray diffraction analysis indicates that the soft grey residue around the pyrite particles is amorphous.

DISCUSSION

From the results of this test work it is observed that, although initial iron concentration and nutrient strength are important in determining the overall uranium extraction rate obtained in leaching the plus 8 mesh sizes, the major factor influencing the efficiency of bacterial leaching is the ore particle size. In the tests on plus 4 mesh material, only 72% extraction of the uranium was obtained after thirty-two weeks of leaching. Although it is not possible to predict, with any degree of certainty, the time that would be required to reach an extraction of 90% on this coarse ore, the indications from the slope of the curve in Test 3 of Figure 2 are that a leaching period of at least one year would be needed. Even with ore crushed to minus 4 mesh, up to about twenty weeks would be needed to effect 90% extraction. It is probable that the relatively long periods required to obtain an economically high uranium extraction by bacterial leaching, even with relatively finer particles, will make unattractive the use of bacterial leaching as the primary method for recovering uranium from the Elliot Lake ores.

On the other hand, it is quite reasonable to consider using bacterial leach techniques to supplement more efficient ore treatment methods, or to recover uranium from sources that could not be treated otherwise. An example of this latter use is the present application of bacterial leaching in the Elliot Lake mines to recover uranium from broken ore left in the stopes on the completion of mining operations⁽⁵⁾. Although this use of bacterial leaching is resulting in the economical recovery of an appreciable amount of uranium, the results of the test work suggest that the efficiency of the operation might be improved by adding iron and nutrient salts to the stope ore.

Examples of the use of bacterial leaching along with more efficient ore treatment methods would be the bacterial leaching of flotation or gravity tailings from a preconcentration operation, where the preconcentrate would be leached by conventional acid leach methods. It has been shown, in this work, that flotation tailings can be leached efficiently by bacterial action, provided pyrite is added to the tailings. The need to add pyrite is not considered to be a disadvantage, since the pyrite could be obtained by combining the tailings from the preconcentrate leach step with the flotation tailings. This is possible because the pyrite is not significantly affected by acid leaching. The combination of the acid leach tailings and the flotation tailings in preparation for bacterial leaching would have the added advantage that some of the uranium not recovered by acid leaching might be recovered by bacterial leaching. On the basis of the test work, it is indicated that, where a concentrate for acid leaching is made by flotation and where the tailings are leached by bacterial action, the overall recovery of uranium would be as good as that obtained by acid leaching the whole ore.

The above comments would apply equally well to an operation in which gravity preconcentration and bacterial leaching were used. On the basis of the results obtained in this work, from leaching sink-float tailings in the presence of bacteria, it would appear that in order to get 90% extraction of the uranium from this product it would be necessary to crush it considerably finer than the sizes normally used in sink-float operations. If equipment such as tables or Humphreys spirals were used for the preconcentration step, where the ore would be crushed to about 48 mesh for the concentration operation, the tailings could probably be leached by bacterial action without further crushing being necessary.

It has been shown in this test work that as the initial iron concentration increased from 0 to 0.5 to 1.0 g Fe^{++}/l , the uranium extraction rate increased proportionately. The leaching rate curves suggest that initial iron concentrations of more than 1.0 g Fe^{++}/l might possibly yield higher rates of extraction than those already established. When leaching ore coarser than 8 mesh, it was shown that the addition of full-strength nutrient resulted in an increased extraction rate as compared to tests where less nutrient was added. In any large-scale bacterial leaching operation, particularly

where some or all of the ore is coarser than 8 mesh, most of the leaching solution would have to be recycled to maintain a suitable concentration of soluble iron and nutrient salts. If recycling were not used, the concentration of iron and nutrient salts would have to be maintained by the addition of suitable chemicals. It has been demonstrated by the test work (page 15), where solution that had been recycled for eighteen weeks was replaced by fresh solution with no resulting improvement in extraction rate, that recycling of solution is not detrimental.

CONCLUSIONS

In the bacterial leaching of Elliot Lake ore, the overall extraction rate that can be obtained is determined mainly by the particle size of the leach feed. On sizes coarser than 4 mesh, where thirty-two weeks or more is required to reach a uranium extraction of 70%, the extraction rate is related mainly to the rate at which the solutions penetrate the particle to depth. Since this relationship is logarithmic in nature, the time taken to extract appreciable amounts of uranium is increased greatly for a moderate increase in particle size. Regardless of the particle size, the overall uranium extraction rate is increased when the iron concentration in the leach solution increases. The addition of nutrient improves the leaching rate for particles coarser than 8 mesh.

The relatively slow leaching rates indicated by this work would preclude the general use of bacterial leaching as a primary method of treating the Elliot Lake ores. However, it might have an application in the treatment of ores which could not be treated economically in any other way.

Bacterial leaching will extract 70-80% of the uranium contained in the tailings from the treatment of Elliot Lake ore by flotation. The use of bacterial leaching would appear to be of most interest in applications, such as this, where it is employed to supplement more efficient recovery methods.

ACKNOWLEDGEMENTS

The contribution of other members of the Extraction Metallurgy Division to this work is acknowledged. The authors wish particularly to thank those of the Chemical Analysis Section of the Extraction Metallurgy Division under the supervision of Mr. J. C. Ingles, Section Head, and especially Messrs. J. B. Zimmerman and E. D. Kornelsen. Acknowledgement is also made of the assistance given by Mr. S. Kaiman and his staff of the Mineralogy Section for their work on the mineralogical studies. The authors are grateful to Mr. K. T. Price for conducting the experiments and compiling the test data. Thanks are expressed to Dr. K. C. Ivarson of the Department of Agriculture for preparing the bacterial culture used in the leach tests,

and for his advice obtained during the test work. The authors are also indebted to the staff of Denison Mines Limited who supplied the mine samples used in the investigation, and to Mr. H. W. Smith, Divisional Planning Officer, for his helpful advice.

REFERENCES

1. V. F. Harrison, W. A. Gow and K. C. Ivarson, "Leaching of Uranium from Elliot Lake Ore in the presence of Bacteria", Extraction Metallurgy Division Internal Report EMA 65-13, Department of Mines and Technical Surveys, Ottawa, July 26, 1965.
2. M. P. Silverman and D. C. Lundgren, "Studies on the Chemoautotrophic Iron Bacterium Ferrobacillus ferrooxidans". An improved medium and a harvesting procedure for securing high cell yield. J. Bacteriol., 77, 642-647 (1959).
3. W. R. Honeywell and S. Kaiman, "Flotation of Uranium from Elliot Lake Ores", C.I.M. Transactions, 69, 99-107 (1966).
4. M. R. Hughson and S. Kaiman, "Mineralogical Investigation of Bacterial Leach Residues of Ore from Denison Mines Limited, Elliot Lake, Ontario", Mines Branch Investigation Report IR 65-64, Department of Mines and Technical Surveys, Ottawa, Canada, September 7, 1965.
5. M. R. Hughson and S. Kaiman, "A Further Report on the Mineralogy of Bacterial Leach Residues, Denison Mines Limited, Elliot Lake, Ontario". Mines Branch Investigation Report IR 66-40, Department of Mines and Technical Surveys, Ottawa, Canada, May 29, 1966.
6. J. R. Fisher, "Bacterial Leaching of Elliot Lake Uranium Ore". C.I.M. Trans., 69, 167-171 (1966).
7. R. A. MacGregor, "Recovery of U_3O_8 by Underground Leaching", C.I.M. Trans., 69, 162-166 (1966).