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Research on the possibility of concentrating low-grade uranium ores by bacterial leaching, (part of a coordinated programme on the bacterial leaching of uranium ores)

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RESEARCH AND DESIGN CENTRE
FOR RADIOACTIVE METALS
Kögurele - Bucharest - Romania

THE POSSIBILITY OF CONCENTRATING LOW-GRADE
URANIUM ORES BY BACTERIAL LEACHING

(Final Report)

Contract Number : 1205/R3/RB

Chief Scientific Investigator :

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Bucharest, December 1978.

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Institute where is being carried out :

RESEARCH AND DESIGN CENTRE FOR RADIOACTIVE METALS
BUCHAREST - MAGURELE - ROMANIA

Chief scientific investigator :

Dr.TATARU SEVER

Time period covered :

Dec.1977 - Dec.1978

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Research on the possibility of concentrating low-grade uranium ores by bacterial leaching
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Research and Design Centre for Radioactive Metals,
Bucharest, Măgurele, Romania.

Dr.Sever Tătaru

Dec.1977 - Dec.1978.

A great part of all uranium reserves made obvious by geological workings can't be mined in our days because of their low content, because of the mining difficulties or of too costly recovery technologies.

Keeping of some uranium reserves besides the reserves that will be mined in the next years, could be due to one or more of the reasons above-mentioned; to all of these, conjuncture factors or emergency factors could be added, which can change essentially the destination of some uranium ores.

The uranium accumulations are considered to be mineable at the moment when the development technology is quite efficient and uncostly so that permit to mine unexpensively the uranium in poor ores.

The leaching in heap bacterially stimulated is one of the possible ways to turn to account the poor ores, which take much shares to all the resources.

The interest in the bacterial leaching in heap is stimulated by the minimum investments required, low mining expenses and by less trained men being required.

The inconstant production, the influence of weather factors, great surfaces covered, make the procedure be applied only for the residual reserves that were otherwise degraded.

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But taking into account the advantages of bacterial leaching, that proved to be competitive for copper, we hope that by practicing, we will improve our knowledge, so that the bacterial leaching be used for the development of important uranium reserves.

Estimating the importance of the technologies utilisable for the recovery of uranium from poor ores, IAEA co-ordinates and subsidizes a research program aiming at the bacterial leaching.

The researches concerning the bacterial leaching of uranium ores performed in the previous years aimed at the separation of autochthonous bacteria strains from the mine waters simultaneously with the finding out of the multiplying conditions. During the following stages, the efficiency of solubilizing the uranium as a function of a series of factors such as : ore granulation, its mineralogical composition, porosity of lumps, have been studied.

This final report concerns the research-works being carried out in the first stage on:

(a) studies on the effect of solvent extraction reagents on bacterial action directed towards quantification of solvent stability and losses;

(b) study of influence of different eluents on bacterial growth.

1. Development of bacteria population in case of a permanent contact with the eluate compounds and with alamine 336

For carrying out the research-work, three bacterial sources were chosen, because of ecological reasons, proper to each type of ore that has to be submitted to the bacterial leaching, considering that the bacteria resulted from every ore individually are ecologically adapted to the chemico-mineralogical composition of the respective ore.

The substances with which the bacteria were contacted are those directly, technologically involved in the process of recovery of uranium from ores and with which the bacterial leaching is put into contact intentionally or accidentally during the operation of the bacterial leaching equipments.

In the first stage, samples were taken from three mine waters, conventionally called SB1, SB2 and SB3 and were inoculated as follows :

SB1 - in the 9 K Brynner nutrient medium - with 1 g/l Fe^{2+} at a pH = 2.0

SB2 - in the 9K Brynner nutrient medium - with 10 g/l Fe^{2+} at a pH = 1.5

SB3 - in a medium without nutrients, at a pH = 6.5, proper to the mine water which it belongs to.

After a 14 days ripening, samples of these bacteria strains were contacted to :

- 0.1 M nitric acid - 5 per cent to the medium
- 0.9 M ammonium nitrate - 5 per cent to the medium
- 0.1 M nitric acid + 0.9 ammonium nitrate - 5 per cent to the medium.
- kerosene mixture with 5 per cent amine - 100 per cent to the medium
- kerosene - 100 per cent to the medium.

Putting into contact the media with the above reactants lasted for 25 days, while all the samples have been daily stirred by air-bubbling and the evolution of the bacteria density was periodically determined.

The evolution of the population with the time is presented in relative values comparatively to the witness in tables no.1, 2, 3, 4, 5 and for the witnesses in table no.6. To illustrate, data in the tables no.1, 2, 3, 4, 5 are plotted in figures no.1, 2, 3, 4 and 5.

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Table no.1

Evolution of bacteria relative density after t
the contact with 0.1 M nitric acid

Time days	% density to the witness		
	Origin of bacteria		
	SB-1	SB-2	SB-3
0	100	100	100
4	45	87	75
7	46	82	67
15	72	86	76
18	76	90	95
20	75	92	100
22	65	92	100
25	70	105	108

Table no.2

Evolution of bacteria relative density after the
contact with 0.9 M ammonium nitrate

Time days	% density to the witness		
	Origin of bacteria		
	SB-1	SB-2	SB-3
0	100	100	100
4	47	66	38
7	53	71	50
15	50	50	48
18	57	46	64
20	81	56	92
22	99	56	85
25	95	60	92

Table no.3

Evolution of bacteria relative density after the contact with 0.1 M nitric acid and 0.9 M ammonium nitrate

Time days	% density to the witness		
	Origin of bacteria		
	SB-1	SB-2	SB-3
0	100	100	100
4	38	75	50
7	52	88	56
15	48	75	85
18	50	73	81
20	74	76	89
22	81	76	89
25	83	80	107

Table no.4

Evolution of bacteria relative density after the contact with kerosene and alamine 336

Time days	% density to the witness		
	Origin of bacteria		
	SB-1	SB-2	SB-3
0	100	100	100
4	78	55	31
7	53	50	22
15	24	30	19
18	29	37	18
20	40	41	20
22	50	57	23
25	45	50	29

Table no.5

Evolution of bacteria relative density after the contact with kerosene

Time days	% density to the witness		
	Origin of bacteria		
	SB-1	SB-2	SB-3
0	100	100	100
4	100	80	62
7	100	75	62
15	60	75	72
18	48	73	73
20	50	62	91
22	55	56	91
25	53	58	100

Table no.6

Evolution of bacteria density (10^6 /ml) in the witness media

Time days	Bacteria density 10^6 /ml		
	Origin of witness		
	SB-1	SB-2	SB-3
0	20	15	8
4	19	15	8
7	19	16	9
9	19	16	9
11	20	18	10
13	21	20	10
15	21	20	11
18	21	19	11
20	20	20	13
22	20	20	13
25	19	19	12

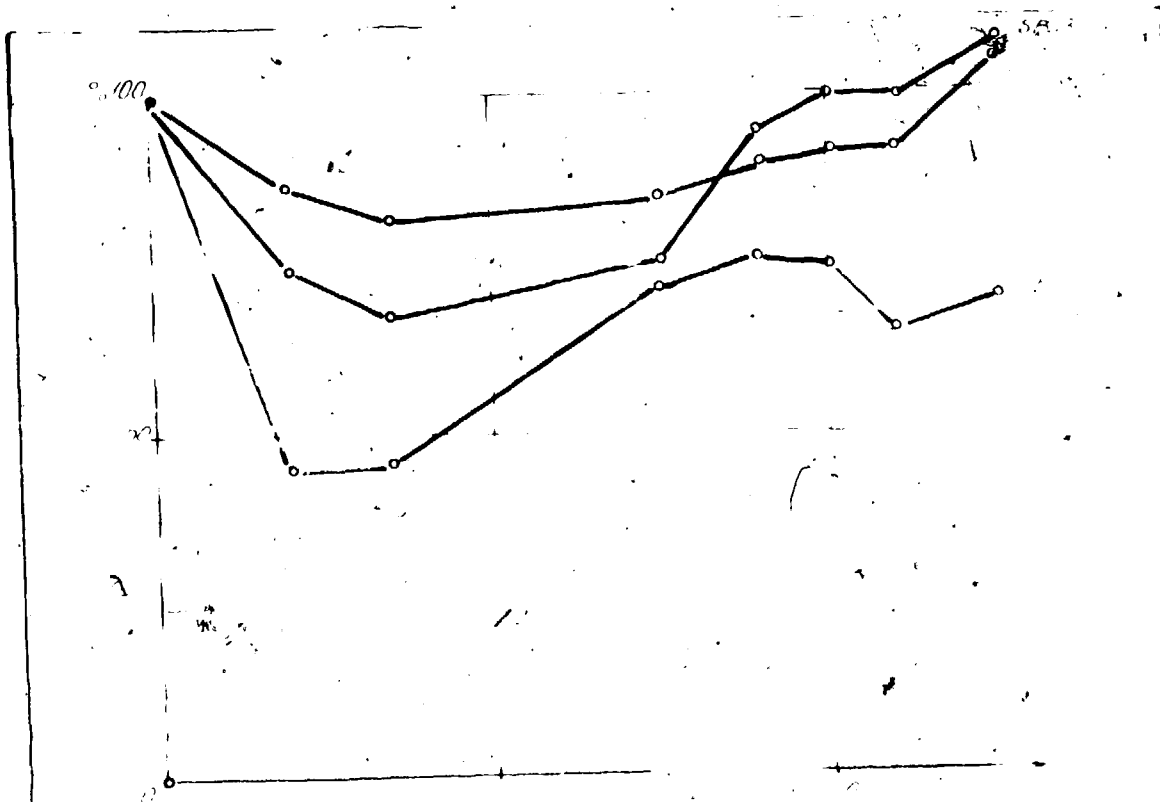


Fig. 1. Evolution of bacteria relative density after contact with 0.9M ammonium nitrate

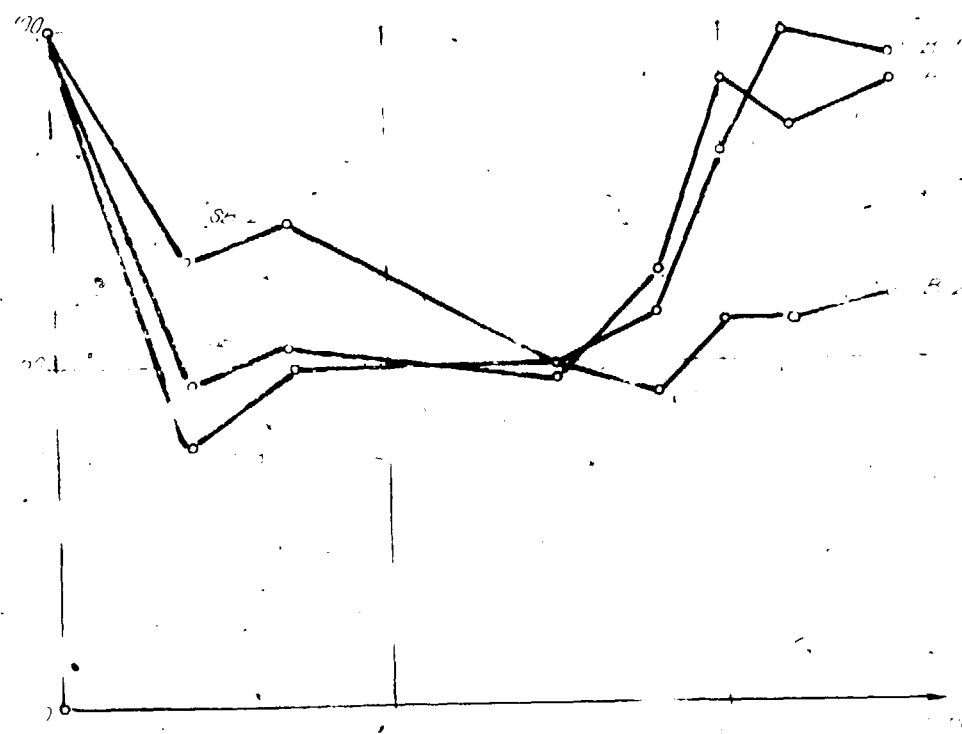


Fig. 2. Evolution of bacteria relative density after contact with 0.9M ammonium nitrate

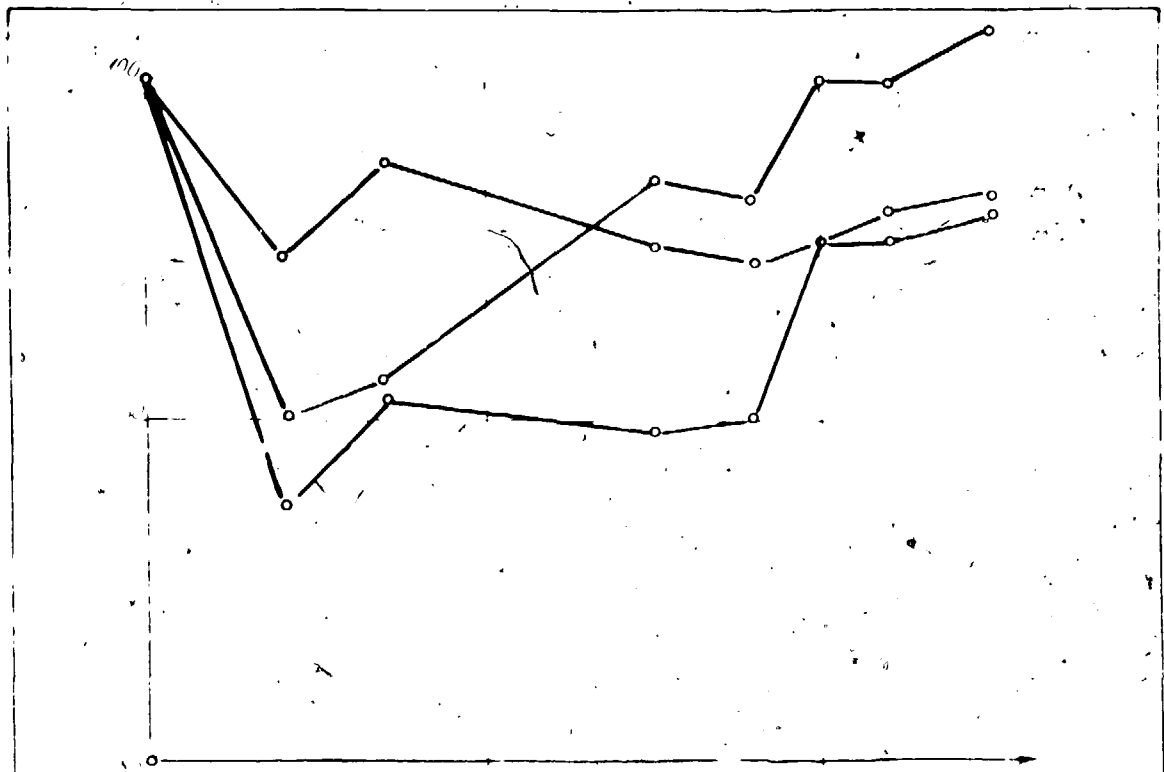


Fig. 3. Evolution of bacterial relative density after contact with Malva acid and ammonium nitrate.



Fig. 4. Evolution of bacterial relative density after contact with kerosene and alamine 336.

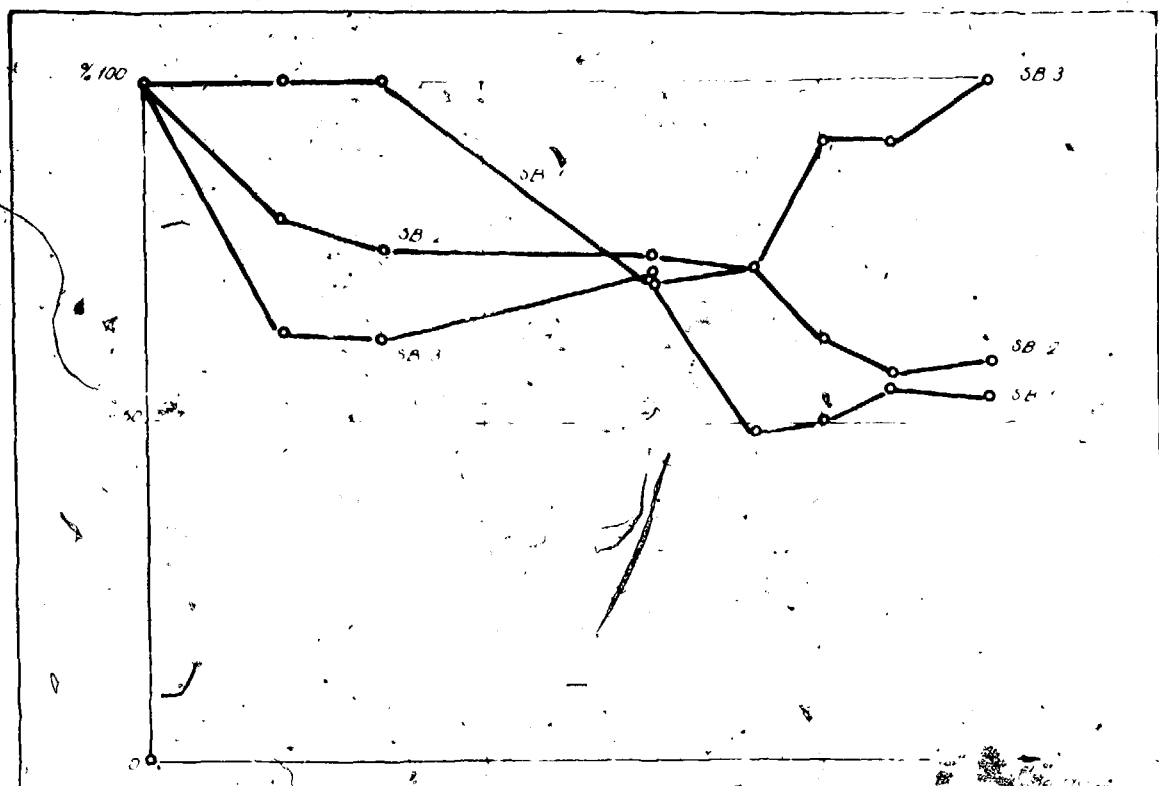


Fig. 5. Evolution of bacterial relative density with respect to time.

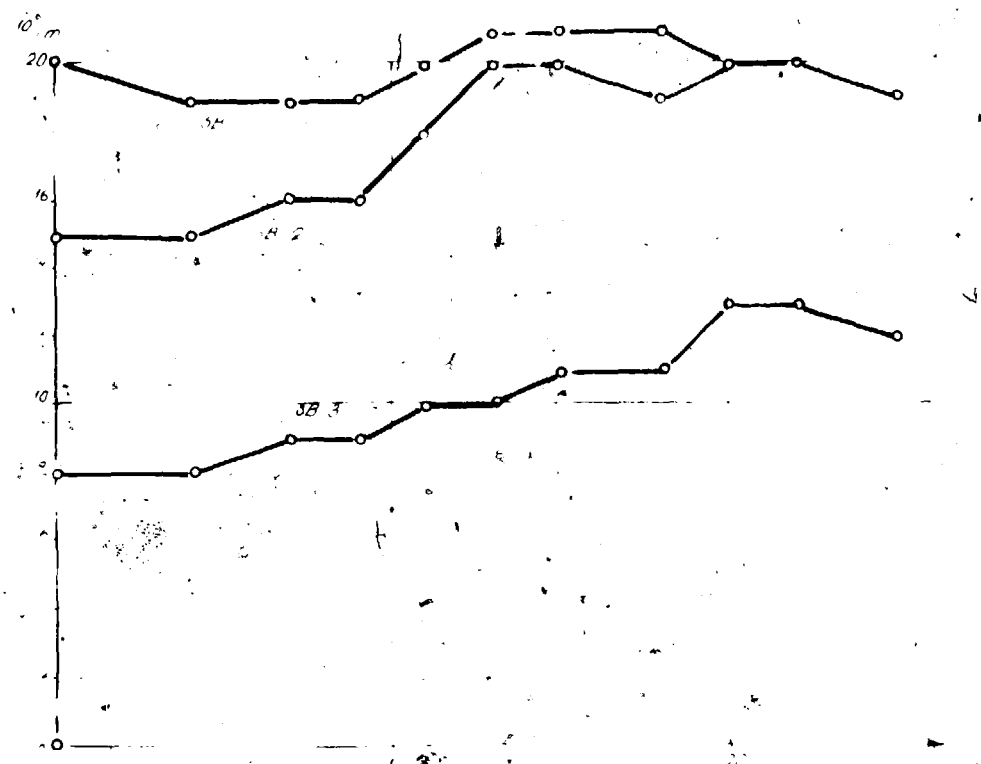


Fig. 6. Evolution of bacterial density (10⁶/ml) with respect to time.

After considering the data in the tables and figures, it follows that in all cases a decrease of the relative number of bacteria will take place, comparatively with the witnesses; this decrease is an aggressive reaction of the respective substances against the bacteria, it leading to an inhibition of the bacteria development.

After an adaptation time, in the final stage of the experiment a growth of the bacteria population density is noticed, as a consequence of some varieties emergence, resistant to the respective agents.

The results of these works certifies on the one hand the harmfulness of the contacting agents being used and on the other hand the outstanding adaptability of the bacteria strains.

2. The influence of the contact with extraction solvents and with the nitric eluent on the biochemical activity of bacteria

Having considered that the evolution of bacteria population is only a partial indication of the effect of solvents and of the nitric eluate, there have been performed determinations in the second part of the research-work, on the evolution with the time of Fe^{2+}/Fe ratio in solutions inoculated with bacteria. In this case, Fe^{2+}/Fe ratio is taken as an indicator with regard to the biochemical activity of bacteria. Bacteria are come from the previous works, being selected by a new inoculation from the solution conventionally called SB-1, that served as inoculum.

The first working conditions are presented as follows :

..//..

	Sample				
	W	A	T	L	E
Culture medium	9K	9K	9K	9K	9K
Inoculum	SB-1	SB-1	SB-1	SB-1	SB-1
Concentration Fe ²⁺ g/l	1.28	1.28	1.28	1.28	1.28
Incubation period days	9	9	9	9	9
Bacteria density after incubation 10 ⁶ /ml	14	14	14	14	14
Fe ²⁺ g/l concentration after incubation	0.05	0.04	0.04	0.05	0.06
Contacting substances	None	Ala mine 336	Trioxyl amine	LIX	Nitric eluant
Time of contacting days	-	7	7	7	continuous

The A, T and L solutions after contacting were separated from the extraction solvents by decantation and an amount of Fe²⁺ (proper to a concentration increase of 1.67 g/l Fe²⁺) was added to all the solutions, including W and E, the concentration becoming 2.85 g/l Fe. Beginning with the fourth day, after Fe²⁺ adding, periodical analyses for Fe²⁺, Fe³⁺ and Fe were performed, simultaneously with the determination of bacteria population density expressed in 10⁶ individuals/ml.

The results of these determinations are presented in table no.7 for the variation of bacteria density as a function of time, and the variation of Fe²⁺/Fe ratio as a function of time is presented in table no.8.

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Table no.7

Evolution of bacteria population after the contact
and Fe^{2+} adding

Time days	Bacteria density (10^6 , individuals/ml)				
	Samples				
	W	A	T	L	E
1	15	10	11	8	9
4	16	9	10	8	8
7	16	6	8	7	7
10	16	6	8	7	7
14	15	6	7	6	7
17	15	5	7	6	6
21	16	5	7	5	5
25	16	4	6	4	5

Table no.8

Evolution of Fe^{2+}/Fe ratio after the contact and
 Fe^{2+} adding

Time days	Fe^{2+}/Fe				
	Samples				
	W	A	T	L	E
1	0.55	0.49	0.50	0.53	0.50
4	0.48	0.52	0.49	0.54	0.51
7	0.41	0.48	0.47	0.50	0.48
10	0.27	0.52	0.50	0.51	0.50
13	0.14	0.47	0.45	0.50	0.46
17	0.08	0.51	0.53	0.50	0.46
21	0.00	0.48	0.51	0.49	0.47
25	0.00	0.49	0.50	0.50	0.46

On the basis of the values in tables no.7 and 8, the following were graphically represented :

- the evolution of bacteria population depending upon the time after contacting for W, L and E samples in figure no.7;
- the evolution of Fe^{2+}/Fe ratio for the W, L and E solutions depending upon the time in figure no.8;
- the evolution of bacteria population depending upon the time after contacting for W, A and T samples in figure no.9;
- the evolution of Fe^{2+}/Fe ratio for W, A, and T solutions depending upon the time in figure no.10.

The following can be noted after investigating the data :

- a continuous decrease of the bacteria population for all the samples contacted, excepting the witness, this confirming the inhibitive action of solvents on the development of bacteria;
- Fe^{2+}/Fe ratio is kept at a relatively constant value of about 0.5 for all the samples contacted, while in the witness sample Fe^{2+} passes entirely to Fe^{3+} .

All these confirms the unfavourable action of extraction solvents on the bacteria development and implicitly on the oxidizing speed of divalent iron to trivalent iron.

In order to avoid these undesirable effects, the bacterial leaching in heap or "in situ" must be carried out on such areas so that after uranium sorption by means of solvents, have much time enough to remake the biochemical activity until the next periodical sorption cycle.

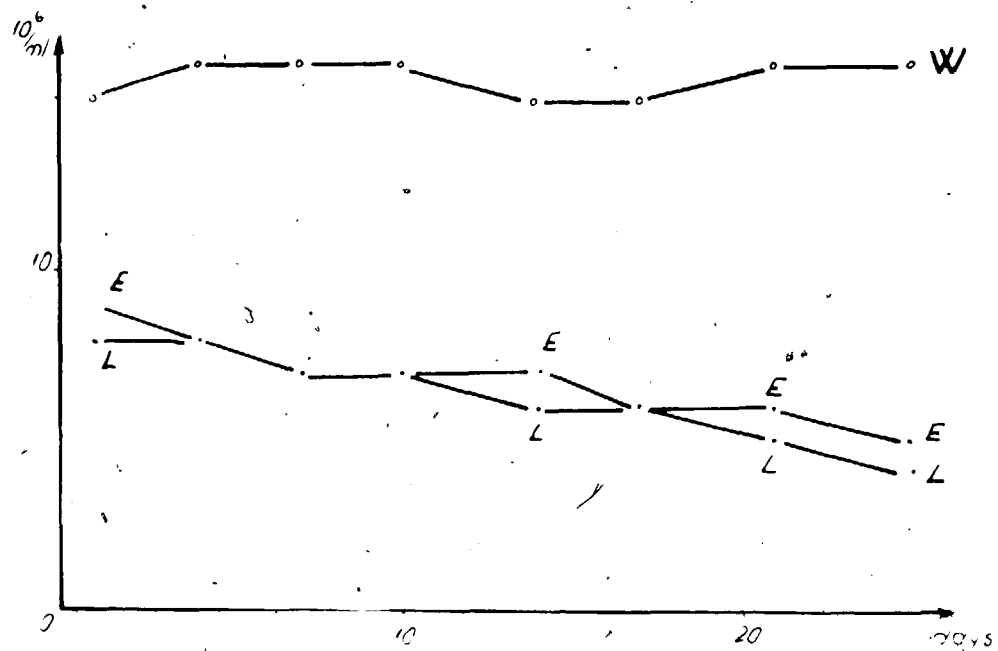


Fig 7
EVOLUTION OF BACTERIA POPULATION

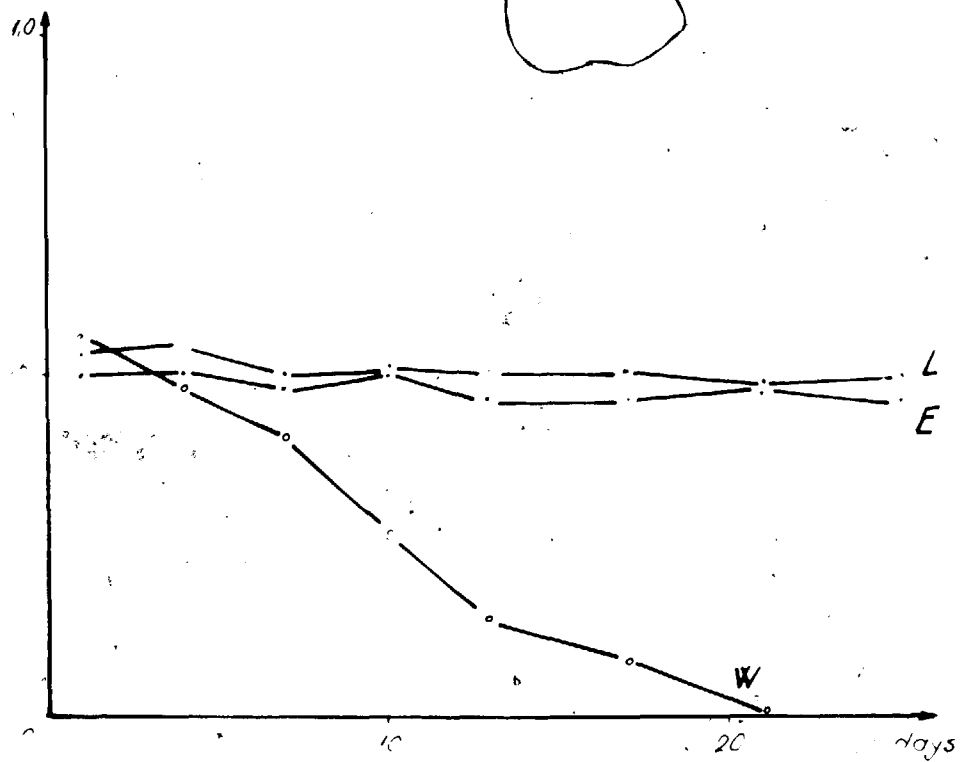


Fig 8.
EVOLUTION OF Fe^{2+}/Fe RATIO

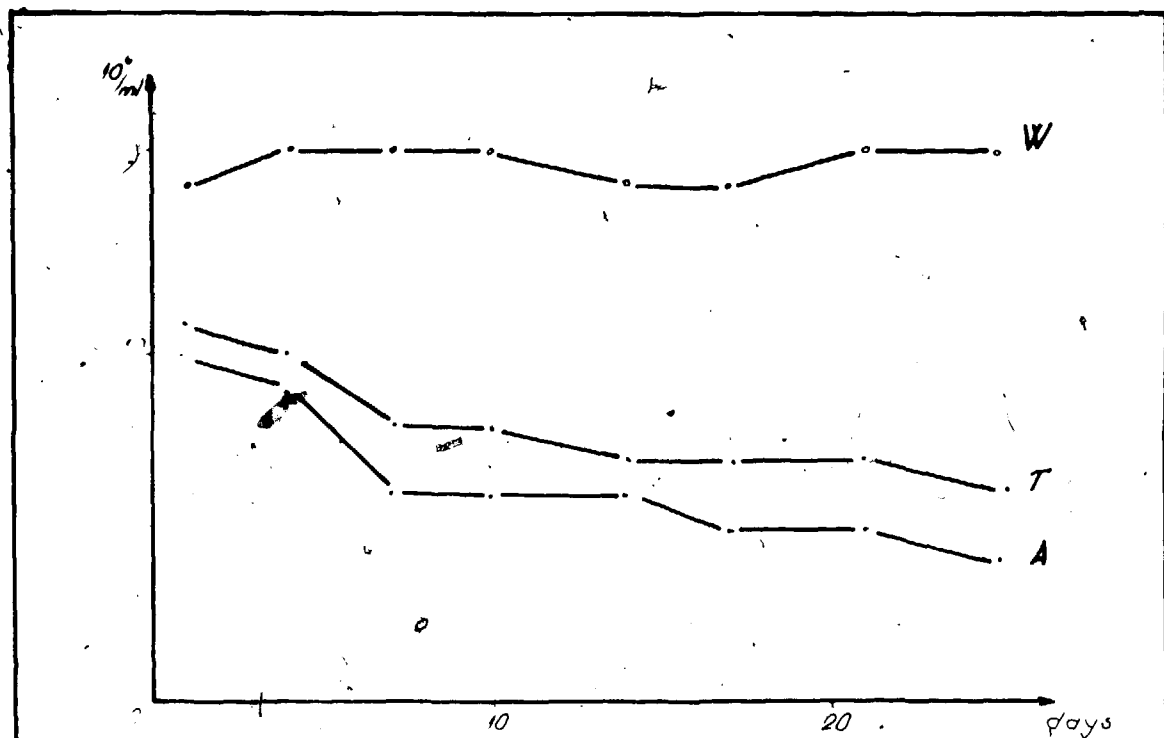


Fig. 9
EVOLUTION OF BACTERIA POPULATION

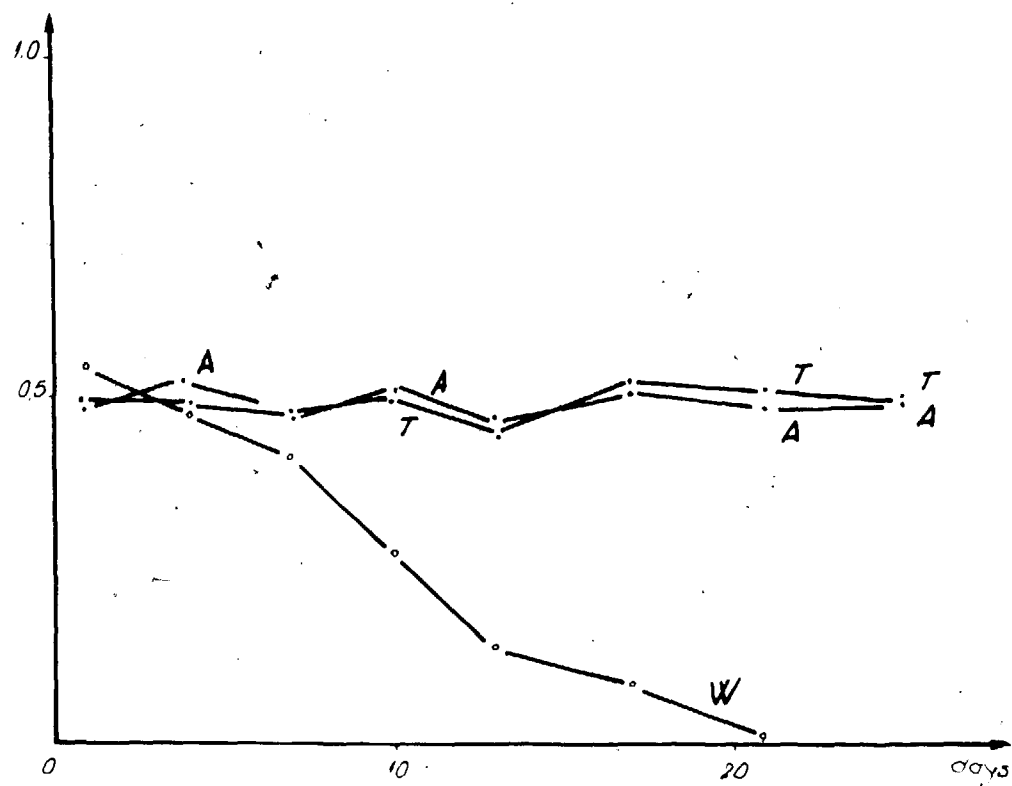


Fig. 10.
EVOLUTION OF Fe^{2+}/Fe RATIO

3. Influence of solvent and of nitric eluant on the bacteria population, biochemical activity and bacteria morphology

In order to study the morphological changes as a result of contacting the bacteria with the extraction solvents, respectively with the nitric eluant, a series of experimental determinations were initiated starting from a bacterial strain selected by repeated inoculations from SB-1 solution come from the washing of ore. This series of determinations was performed under conditions similar with those of section 2, above presented, with the difference that the contacting time was of only 48 hours and the initial Fe^{2+} content was 1.36 g/l Fe^{2+} . The bacteria population after incubation was of $10 \cdot 10^6$ individuals/ml in comparison with $14 \cdot 10^6$ individuals/ml in the previous range.

Simultaneously with the population determinations and with Fe^{2+} analyses, there have been carried out periodical smears with the bacteria gathered from W, A, T, L, E samples in order to investigate microscopically the morphology of bacteria and the eventual evolution of it.

The evolution of bacteria population after contacting and Fe^{2+} adding is presented in table no.9 and the evolution depending upon the time of Fe^{2+}/Fe ratio is presented in table no.10.

The plotting of bacteria of bacteria population evolution for W, L and E samples is presented in figure 11 and the evolution of Fe^{2+}/Fe ratio depending upon the time is presented in figure no.12.

The plotting of evolution of bacteria population for W, A and T samples is presented in figure 13 and the evolution of Fe^{2+}/Fe ratio depending upon the time is presented in figure 14.

Table no.9

Evolution of bacteria population after the contact and Fe²⁺ adding

Time days	Bacteria density (10 ⁶ individuals/ml)				
	W	A	T	L	E
1	16	7	11	10	9
4	16	7	11	9	8
7	17	3	8	7	4
10	15	2	7	4	3
14	15	2	6	3	4
17	16	3	5	3	3

Table no.10

Evolution of Fe²⁺/Fe ratio after the contact and Fe²⁺ adding

Time days	Fe ²⁺ /Fe				
	Samples				
	W	A	T	L	E
1	0.44	0.48	0.48	0.52	0.53
4	0.19	0.48	0.52	0.56	0.53
7	0.08	0.50	0.56	0.55	0.50
10	0.00	0.55	0.55	0.59	0.50
14	0.00	0.50	0.50	0.57	0.52
17	0.00	0.45	0.51	0.58	0.60

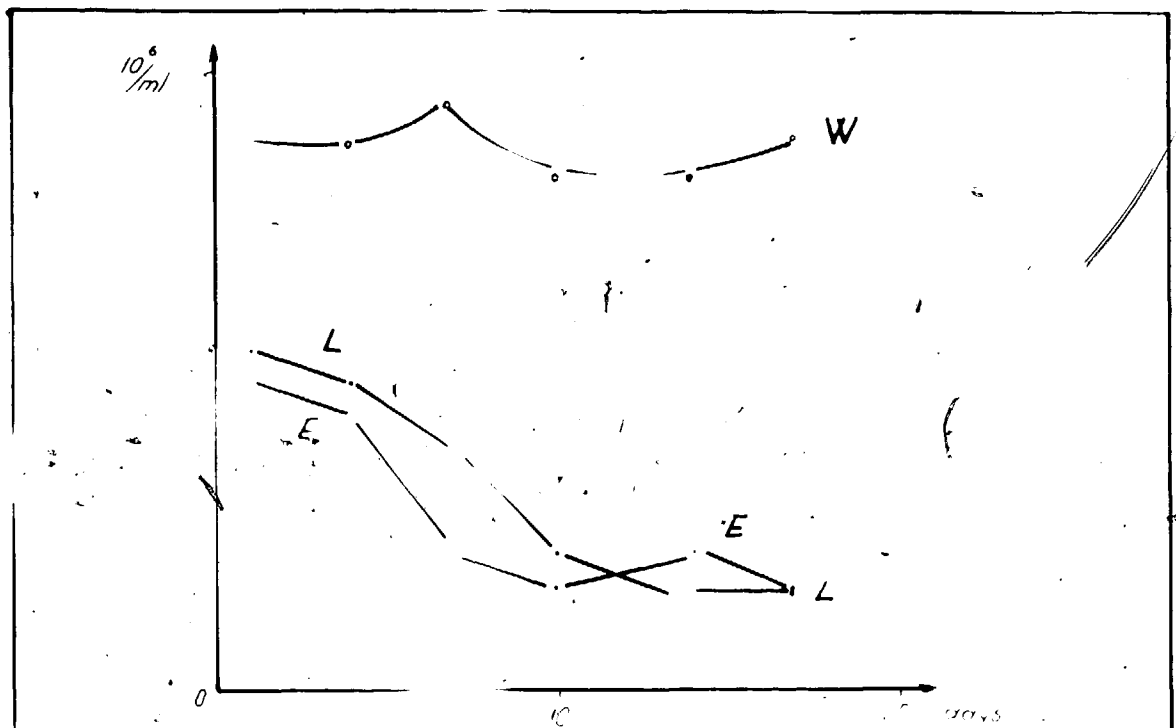


Fig. 11
EVOLUTION OF BACTERIA POPULATION

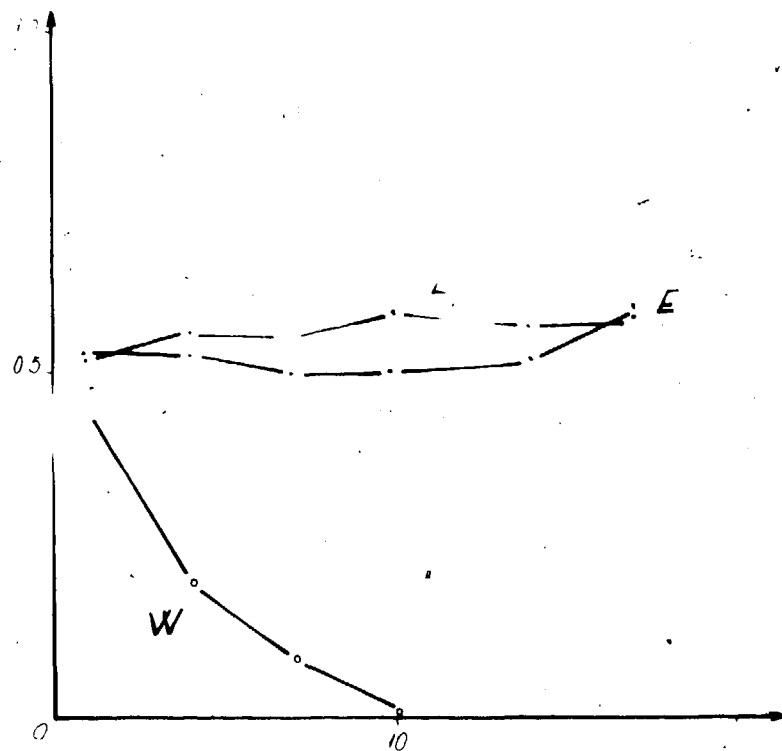


Fig. 12.
EVOLUTION OF Fe^{2+}/Fe RATIO

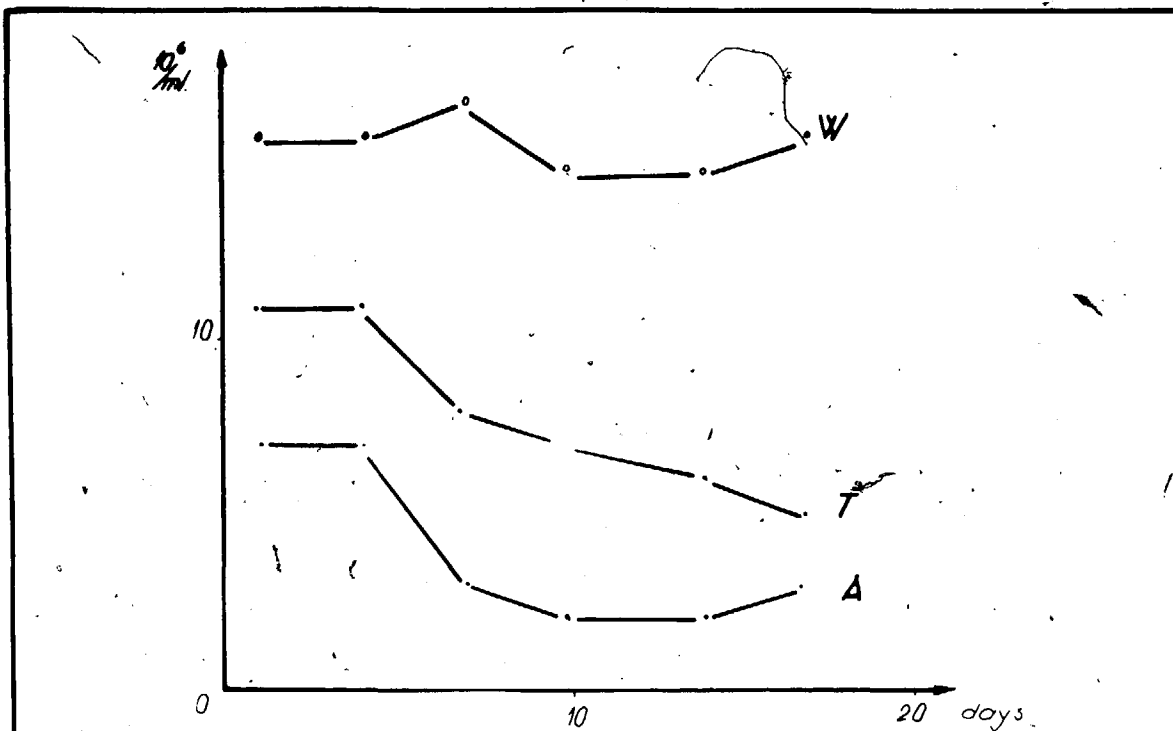


Fig. 13
EVOLUTION OF BACTERIA POPULATION

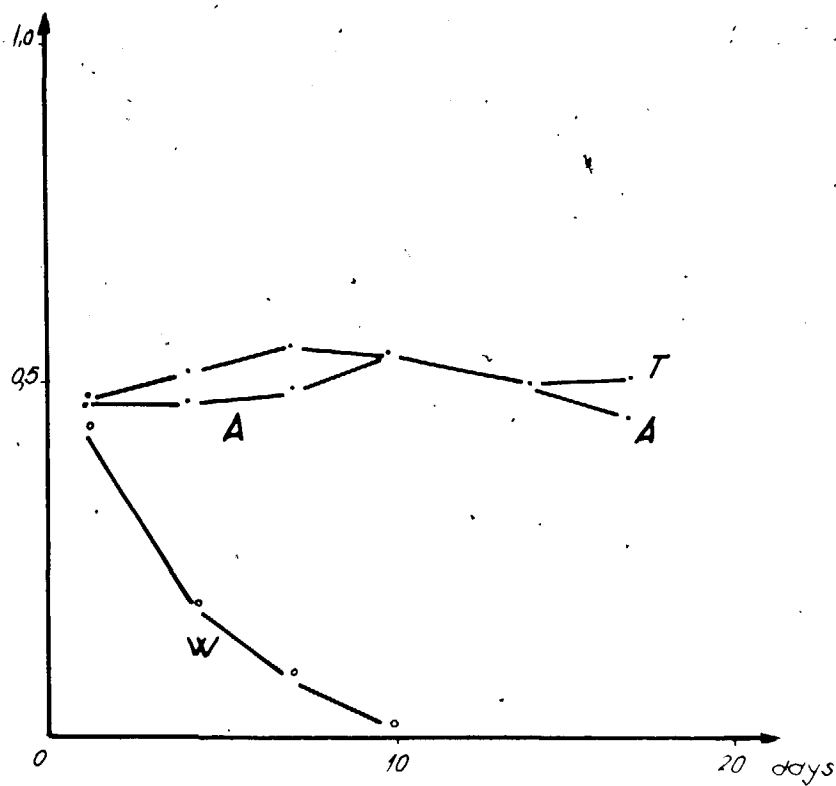


Fig. 14.
EVOLUTION OF Fe^{2+}/Fe RATIO

Similarly with the previous determinations, the inhibitive effect of the contact (even a short one) of the solvents and of the nitric eluate is noticed, both on the development of population and on the oxidizing ability of Fe^{2+} to Fe^{3+} .

Among the three ranges of determinations starting from the same bacterial source but ever better selected, it can be noticed that the inhibitive effect of solvents increases as the bacteria population is more homogeneous. This is explained by the firstly unhomogeneous bacteria populations having greater chances of having more resistant bacteria populations than the homogeneous population.

The smears taken for the microscopical photographs of bacteria in A, T, L and E samples were carried out from samples taken in the first day after Fe^{2+} adding and the 7th and the 16th days after the first sampling.

The smears for W witness sample were carried out from samples taken in the 1st, 4th, 7th, 10th, 13th and 16th day after divalent iron adding.

The aspect of the smears being microscopically photographed and 4400 times multiplied is seen in the photographs no.1, 2, 3 and 4 for A, T, L and E samples and in the photography no.5 for W samples. In these photographs the figures written by the pictures represents the day of sampling beginning with the moment of Fe^{2+} adding to the samples.

After examining the aspect of the microscopic field it can be noticed that bacteria in all the contacted samples were altered.

In case of Alamine 336 contacting, the alterations are like breakings up for the beginning and in the final stage some shorter and thicker bacteria species appear, probably because of the subsequent contamination with some other bacteria.

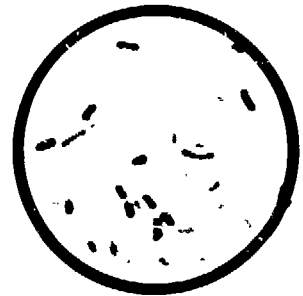
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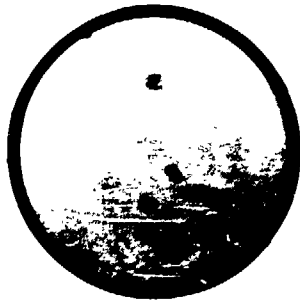
16. days

ALAMINE 336 RANGE

Foto 1.



1.



7.



16. days

TRIOCTYLAMINE RANGE

Foto 2.



1.



7.



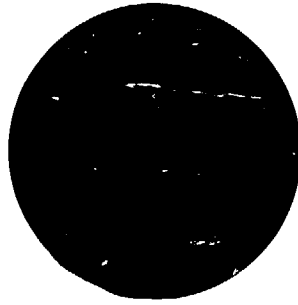
16. days

LIX RANGE

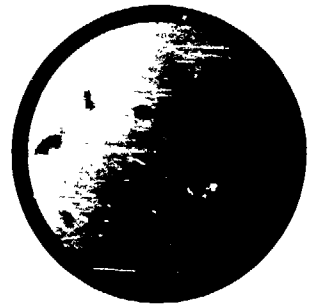
Foto 3.



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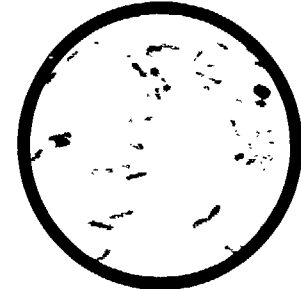
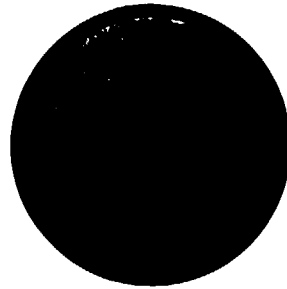
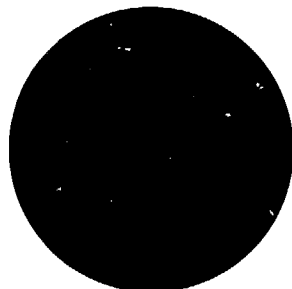
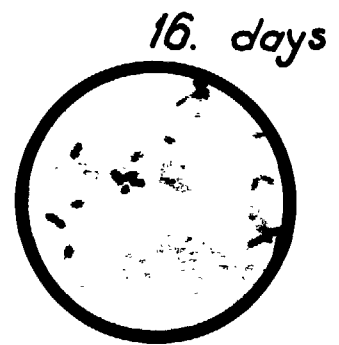


16.

NITRIC ELUENT RANGE

Foto 4.

WITNESS RANGE



4.

10.

13. days

In case of trioctyl amine, the alterations are analogous to those in case of Alamine 336.

In case of LIX contacting, the alterations are the most marked, only some fragments appearing at last.

In case of nitric eluant adding, the destruction of bacteria is obvious from the beginning.

The microscopical images of the witness sample show that the bacteria species are the same until the end.

The morphologically pointed out alterations and a decrease of the number of bacteria explain the decrease of biochemical activity pointed out by the Fe^{2+}/Fe ratio being quite constant in the contacted solutions. Before the contact, the divalent iron changed to trivalent iron in all the samples, in a ratio of 90-95 per cent in only 9 days after the bacteria inoculation; after the contact, the ratio of divalent iron changing to trivalent iron is about 50 per cent after 17-25 days.

CONCLUSIONS

The autochthonous bacteria adapted to a pH=1.5-2.0 have a better resistance on the permanent contact with solvents and with the nitric eluate. Because of very many bacteria varieties being present, some of which having a better resistance, a regeneration capacity can be noticed.

The autochthonous bacteria selected by repeated inoculations have a weaker resistance on the contact with solvents, their bacteria population density and their biochemical activity being decreased. After the contact, Fe^{2+}/Fe ratio is practically the same (about 0.5) while in the witness cultures the total oxidation of Fe^{2+} takes place after about 20 days.

The autochthonous bacteria - Thiobacillus ferrooxidans and thiooxidans very well selected have a weaker resistance in comparison with an even short contact with the extraction sol-

vents and also have a decreased biochemical activity after the contact.

In the witness cultures the total oxidation of Fe^{2+} takes place in less than 10 days.

From a technological point of view, the sorption from bacterial leaching solutions in heap must be made periodically, at time intervals that permit to remake the bacteria population or to make repeated programmed inoculations of the leaching solutions.

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