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Effect of Microorganisms on In Situ Uranium Mining

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The extraction of some metal values, e.g., uranium or copper, may be accomplished by using solutions to remove metals from ore bodies without practicing conventional mining. This process is referred to as in situ leaching and has been used industrially to recover uranium. The growth of microbial populations during in situ leaching is believed to be one of the causes of flow path plugging in the ore body, which results in decreased uranium production. Leach solution and solid samples from well casings and submersible pumps were collected from an in situ mining operation experiencing plugging problems. *Bacillus* sp., *Micrococcus* sp., pseudomonads, and xanthomonads were isolated from these samples in concentrations of 10^5 CFU ml^{-1}. A mixed culture of these organisms was inoculated into a uranium core specimen in the laboratory to assess the role of microbes in the plugging problem. A one-third decrease in permeability was effected in 16 days. Hydrogen peroxide (0.2 g $liter^{-1}$) killed the microorganisms in the core and alleviated the plugging problem. Periodically injecting hydrogen peroxide into the ore body through the production wells may reduce microbial plugging problems.

In situ mining has become an attractive alternative to conventional open pit and underground mining owing to the lower cost, reduced surface disturbance, and suitability of this method for low-grade deposits. This process has been used commercially by the mining industry to extract uranium. The amount of uranium produced by in situ mining in the United States has increased from 0.6% of the total uranium production in 1975 to 7.76% in 1979 (13). Many problems encountered with conventional mining techniques such as exposure to radon gas and mine roof failures are avoided with in situ mining. However, other problems are created by the need to inject solutions into shallow aquifers. Restoration of the aquifer and ore body must be undertaken after the mining operation has ceased; injected chemicals must be removed and metals that have been solubilized must be immobilized in the ore body. Specific requirements for restoration of the ore body have been established by both federal and state agencies (4, 10). To avoid possible contamination of adjacent aquifers, monitoring wells are required so that any lixiviant escaping from the region designated for leaching can be quickly detected. Bacteria, ubiquitous in the overlying soil, can also present problems. They can be introduced into the ore body when wells are drilled or solutions are injected. The addition of some oxidants to solubilize uranium can act as a source of electron acceptors, and the organic matter associated with most uranium deposits of the western and southwestern United States can be a source of carbon. These conditions provide an environment in which aerobic, heterotrophic organisms can flourish. The increased solution flow rates associated with in situ mining could be responsible for desorbing microorganisms that readsorb to soil particles under conditions of natural groundwater flow. These microbes would be transported with the leach solution until they became trapped in pores of fine-grained materials or on well screens.

Several in situ mining operations have experienced decreased production due to heavy accumulations of inorganic particles and bacteria on well screens, casings, and submersible pumps. Similar problems are encountered in both oil wells and water wells. Unfortunately, none of the well treatments developed for those problems are particularly suitable for in situ uranium leaching. The petroleum industry uses antibiotics and organic biocides such as formaldehyde

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(1) to remedy plugging problems. The significantly shallower depths at which in situ leaching is conducted (50 to 300 m) precludes the injection of such toxic agents since domestic aquifers are relatively nearby. An encrustation or microbial fouling problem in a water well is generally remedied by acidifying or chlorinating the well. Neither method is desirable for in situ leaching. Acidification is inappropriate since the correct pH must be maintained throughout the formation to obtain good uranium recovery. The use of chloride is to be avoided since that agent can significantly reduce the loading capacity of the ion-exchange resins used to recover uranium from the leach solution.

The goals of this investigation were to characterize the microbial flora of the in situ mining environment, to ascertain whether these microorganisms were capable of inducing permeability losses (causing plugging problems) in uranium ore under laboratory conditions, and to find an environmentally safe means by which microbially induced plugging problems could be alleviated.

MATERIALS AND METHODS

Sample collection. Samples from a submersible pump and well casing and water samples from several wells (monitor, production, and injection wells) were obtained from a south Texas in situ uranium-leaching operation. The lixiviant in use at this site was ammonium carbonate-bicarbonate and the oxidant, sparged into the lixiviant at the injection wells, was O_2 gas. More detailed physical and chemical data on the wells have been presented elsewhere (M. V. Yates, M.S. thesis, New Mexico Institute of Mining and Technology, Socorro, 1982). Samples were collected aseptically and stored in sterile containers at 4°C until processed in the laboratory.

Microbiological assay. Isolation and enumeration of microorganisms were performed with duplicate pour plates of iron-peptone agar (5). All CFU counts were made after incubation of the plates for 5 days at 25°C. Identification of the microorganisms was based on criteria in *Bergey's Manual of Determinative Bacteriology* (3) and that given by Gilardi (7).

The microorganisms that were isolated from the solid material obtained from the submersible pump were used as inocula in the field simulation. Microbes were inoculated to achieve a concentration of 10^5 CFU ml of pore space of ore^{-1} (M. V. Yates, M.S. thesis).

Chemical analyses. Determination of hydrogen peroxide concentration was accomplished by titration with potassium permanganate (6). Dissolved oxygen concentrations were measured in the laboratory with a dissolved oxygen analyzer (New Brunswick Scientific Co.).

Leach solution. The aqueous leach solution (pH 10.8) contained 3.0 g of carbonate as potassium carbonate liter of distilled water^{-1}; the solution was buffered to pH 7.0 with concentrated HCl for selected experiments.

Ore. The uranium ore body core was obtained from a Wyoming mining operation located in the Wasatch geological formation. The core was characterized as fine- to medium-grained arkosic sandstone, moderately sorted, medium gray, and containing a trace of calcite. The organic content, determined by loss of weight upon combustion (11), was 3.4%.

Core preparation. The uranium core was cut into two blocks, 5 by 5 by 6 cm and 5.9 by 5.3 by 4.6 cm. Lucite plates with a 1-cm hole tapped into the center and a gridwork etched onto one face were glued onto opposing ends of each block, with the gridded face adjacent to the ore. The etched plates assured uniform distribution of solution flow across the face of the ore. These were placed so that the solution flow was directed horizontally across the blocks with respect to their original orientation in the formation. After covering the holes in the plates with tape, the blocks were placed in disposable plastic containers and covered with epoxy resin (Devcon WR; Devcon Corp.). The containers were cut away after 48 h, and threaded metal fittings were screwed into the holes in the plates (Fig. 1).

Field simulation. Leach solution (nonsterile), contained in a 12-liter reservoir, was pumped into the cores by means of a flow inducer (model MHRE2; New Brunswick Scientific Co.). A free-standing head of solution was maintained at the influent to ensure that the flow of solution through the cores was governed only by the difference in hydraulic head across the sample and its hydraulic conductivity. A constant head of leach solution was maintained by adjusting the speed of the pump. Leach solution was directed into the cores through silicon tubing attached to the metal fittings at the ends of the cores (Fig. 2). All simulations were performed in the same laboratory, which had its temperature maintained between 20 and 25°C.

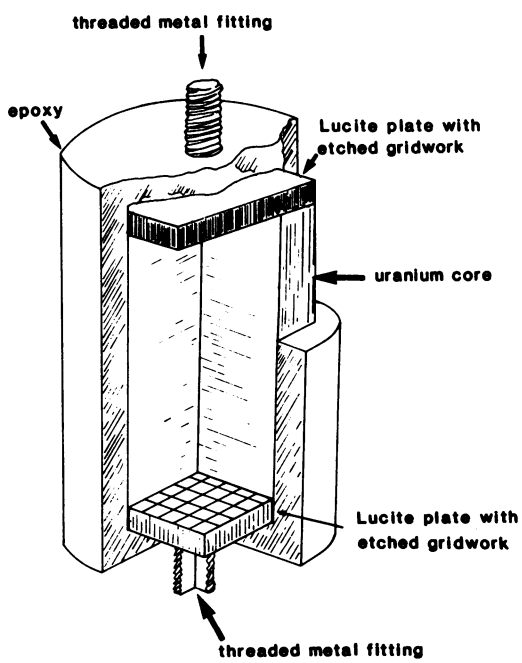

FIG. 1. Cutaway schematic of epoxy resin-covered core from a uranium ore body.

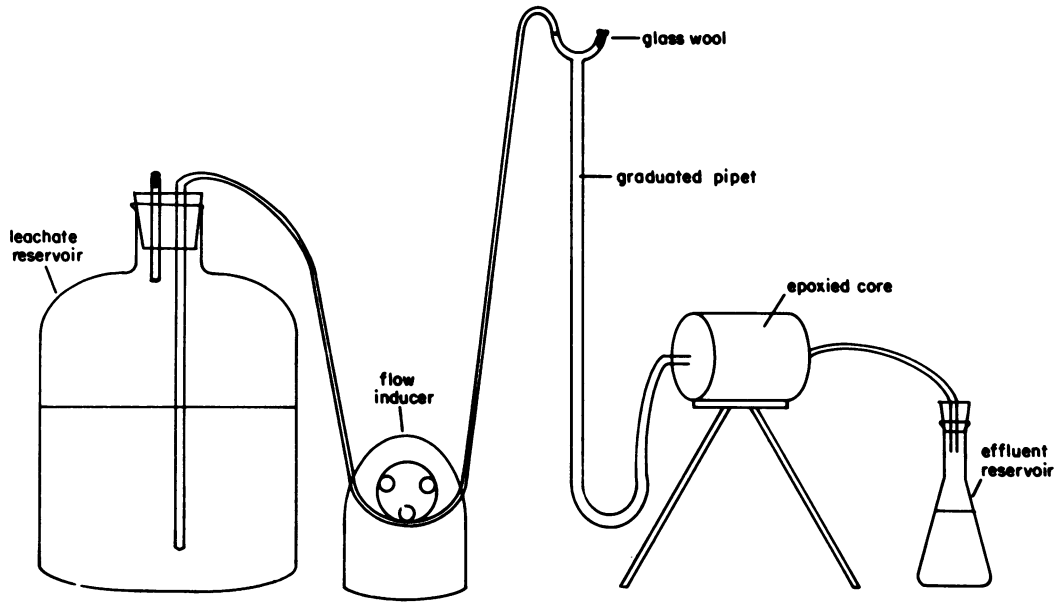

FIG. 2. Diagram of system used for laboratory simulation of an in situ uranium-leaching process.

The hydraulic conductivity, in centimeters per second, was determined by using Darcy's law:

$$K = Q\,[A\,(\Delta h/\Delta x)] \quad (1)$$

where Q is the flow rate (milliliters per second), Δh is the difference in head across the sample (centimeters), Δx is the length of the sample (centimeters), and A is the cross-sectional area of the sample (square centimeters). Δh was kept constant throughout the test, and Q was determined by measuring the volume of effluent produced in a fixed period of time.

The hydraulic conductivity (a porous medium-fluid property) is related to the permeability (a property of the medium only), k in square centimeters, by

$$k = K\nu/g \quad (2)$$

where ν is kinematic viscosity (square centimeters per second) and g is gravitational acceleration (centimeters per second squared). The temperature, pressure, and fluid density were not varied during these experiments so the viscosity remained constant. Hence, k was proportional to K so that, for example, a reduction of K by one-third meant that the permeability was reduced by one-third.

The validity of Darcy's law under these experimental conditions was established based on the calculation of the Reynold's number (R):

$$R = qd_x/\nu \quad (3)$$

where q is the Darcian flux (Q/A) and d_x is the grain size at x percent of the sample (x is usually taken to be either 10 or 50). The Reynold's number was calculated to be 1.00×10^{-5} for this experiment. Since Darcy's law is valid for $R < 1$ (14), the use of equation 1 is justified.

Sterile tap water was passed through the cores until steady flow rates were obtained. Leaching with potassium carbonate solution at pH 7.0 was initiated on day 10. Microorganisms were inoculated into core 1 on day 28 whereas core 2 served as a control. On day 44 the pH of the leach solution was changed from 7.0 to 10.8, and on day 137 hydrogen peroxide (0.2 g liter^{-1}) was added to the leachate reservoir. This sequence of parameter changes is shown in Fig. 3. Enumeration of microorganisms in core 1 effluent was performed on days 73, 110, and 18 additional times between days 137 and 160. Core 2 effluent was analyzed periodically for microbial contamination. After day 137, the leachate solution was analyzed periodically to ascertain that the hydrogen peroxide concentration was unchanged.

RESULTS

The microorganisms isolated from the samples obtained at the in situ mining operation were all common soil microbes: pseudomonads, xanthomonads, *Bacillus* sp., and *Micrococcus* sp. Approximately 10^7 CFU g of dry weight^{-1} were found in the solid samples from submersible pumps and well casings; the leach solutions contained ca. 10^5 CFU ml^{-1} (Table 1).

The changes in hydraulic conductivity with time in both cores are shown in Fig. 3. Inoculation of microorganisms into core 1 caused a decrease in conductivity; an attempt to increase the conductivity by raising the pH of the leach solution to 10.8 was not successful. The addition of hydrogen peroxide on day 137, however, did cause an increase in the conductivity of core 1 to the preinoculation level; no changes in the conductivity of core 2 were detected.

Concurrent with the conductivity increase of core 1 after the addition of hydrogen peroxide to the leach solution, the number of viable bacteria

TABLE 1. Number and genera of microorganisms identified in samples collected from a south Texas uranium in situ mining operation

Sample location	Sample type	No. of microbes (CFU ml^{-1})[a]	Genera of microbes identified
Polyvinyl chloride pipe in production well (well casing)	Solid	1.4×10^7	Pseudomonads, xanthomonads, Biogroup VE
Submersible pump in production well	Solid	2.4×10^7	Pseudomonads, xanthomonads, *Micrococcus*
Inside submersible pump in production well	Solid	1.4×10^7	Pseudomonads, xanthomonads, Biogroup VE
Outflow from ion-exchange plant	Liquid	5.6×10^5	Pseudomonads, xanthomonads, Biogroup VE, *Micrococcus, Bacillus*
Monitor well in ore zone	Liquid	4.7×10^5	Pseudomonads, xanthomonads, *Bacillus*
Production well	Liquid	9.5×10^4	Pseudomonads, xanthomonads, Biogroup VE, *Micrococcus*
Monitor well in overlying aquifer	Liquid	1.5×10^5	Pseudomonads, xanthomonads, Biogroup VE

[a] For solid samples, numbers are reported as CFU g^{-1} (dry weight).

detected in the effluent decreased from 10^5 CFU ml^{-1}, the number observed on days 73, 110, and 137, to 1 CFU ml^{-1} over a period of 20 days (Fig. 4). Core 2 effluent was consistently free of microorganisms, and its hydraulic conductivity remained relatively stable over the course of the experiment.

The hydrogen peroxide contents of the effluents of the inoculated and control cores were, respectively, 65 and 70% of the influent content. Dissolved oxygen concentrations in the influent and effluent solutions of both cores were 7.9 and 8.0 $\mu g\ ml^{-1}$, respectively.

Upon completion of the experiments, core 1 was cut and a cross section was examined. No structural changes or precipitates were apparent.

DISCUSSION

Each core initially exhibited a decreasing hydraulic conductivity that eventually stabilized. The inoculation of core 1 was not performed until stabilization, which required 20 days, had occurred. Initial periods of unstable conductivity have been observed in other uranium-leaching experiments and have several causes, including the migration of loose, fine particles in the sample and the swelling of clays. The low calcium content of the ore and the absence of precipitates in the cross section of core 1 after leaching indicate that plugging by precipitates, such as calcium carbonate, was unlikely. The conductivity reductions observed here are well within the range of those obtained in experiments performed by other workers (R. S. Schechter and L. W. Lake, personal communication) evaluating the response of uranium ores to potassium carbonate leach solution.

Microorganisms isolated from the in situ uranium-leaching environment were identified as aerobic, heterotrophic organisms commonly found in the soil. It was not determined if these microorganisms were capable of growing in the subterranean in situ leaching environment. Inoculation of a mixed culture of these microbes into uranium cores in concentrations of 10^5 CFU ml of pore space of ore^{-1} effected a permeability decrease by a factor of 3 in 16 days; this lowered permeability was sustained for 93 days in the presence of a pH 10.8 potassium carbonate leach solution. The number of bacteria inducing the permeability loss in the laboratory core is similar to the number found in leach solution samples collected at a site experiencing plugging problems.

The addition of hydrogen peroxide (0.2 g $liter^{-1}$) to the leach solution increased the permeability of the core to preinoculation levels and eliminated the bacteria in the core. Similar concentrations of hydrogen peroxide have been

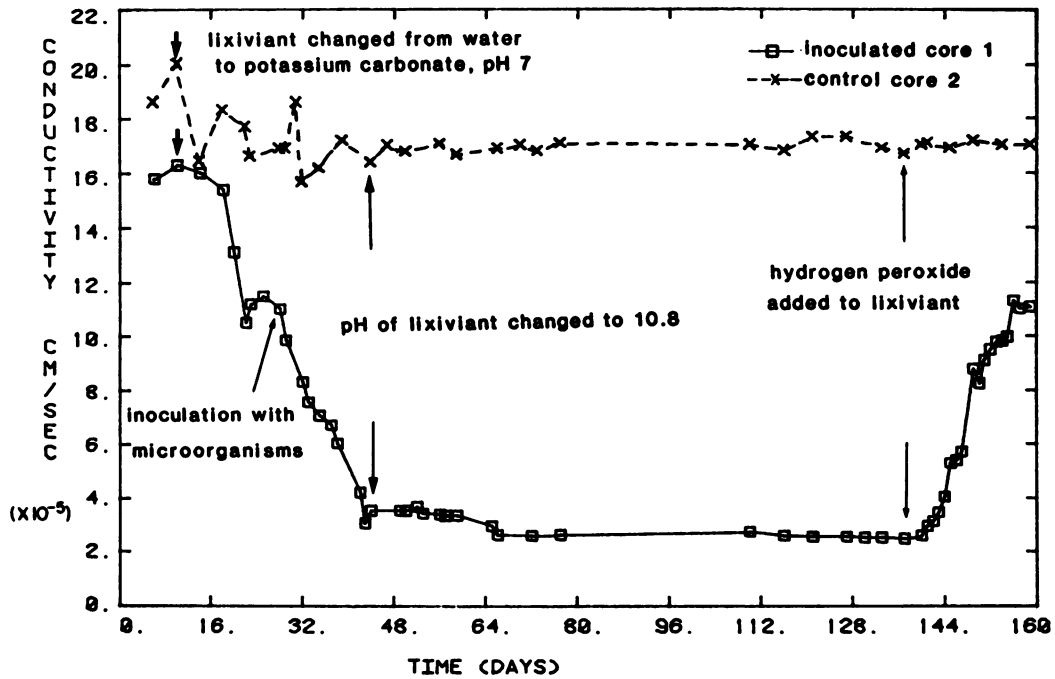

FIG. 3. Change in hydraulic conductivity in inoculated (core 1) and uninoculated control (core 2) cores of a uranium ore.

found to inhibit the growth of thiobacilli in laboratory experiments (2). Hydrogen peroxide degrades to produce oxygen that can at high concentrations be toxic to microorganisms. The concentration of dissolved oxygen in the effluents from the laboratory experiment was only 8

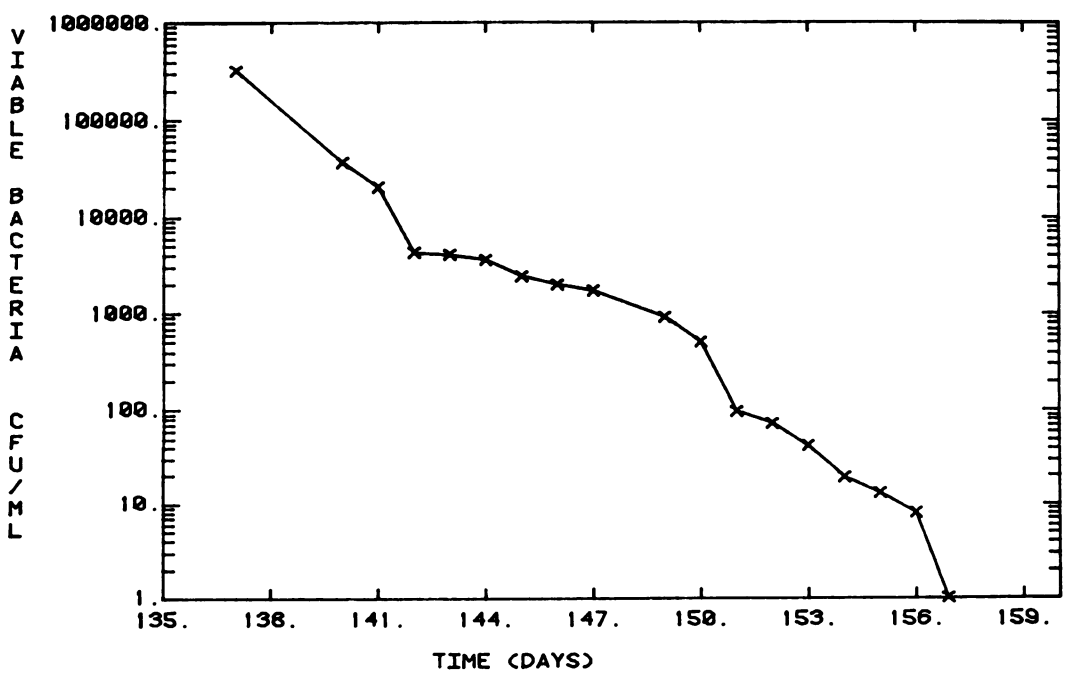

FIG. 4. Change in viable bacterial concentration with time in core 1 effluent after addition of hydrogen peroxide (0.2 g liter^{-1}) to leach solution.

$\mu g\ ml^{-1}$. However, it has been shown that *Bacillus* sp. and pseudomonads are capable of growing at oxygen concentrations of 35 $\mu g\ ml^{-1}$ (15). In the in situ leaching environment, however, hyperbaric oxygen concentrations may be high enough to be toxic to the microorganisms. It has been demonstrated that the growth of several microbes, including pseudomonads and *Bacillus* sp., was found to be inhibited by 35 to 70 μg of oxygen ml^{-1} (15).

The results of this study suggest that the injection of hydrogen peroxide through production wells into the ore body may diminish microbial growth at the production well and in the surrounding ore body. Hydrogen peroxide has the advantage of being environmentally safe, by degrading to water and oxygen. From the standpoint of site operators, it would be preferable to inject oxygen (as opposed to hydrogen peroxide) since it is normally already being used as the oxidant in the leach solution. Whether oxygen would be as effective as hydrogen peroxide is an open question. As the preceding paragraph pointed out, it is not known if the observed effects of the hydrogen peroxide were due only to the resulting elevated oxygen concentration. Other mechanisms must be considered, such as the possible toxicity of free radicals created as the hydrogen peroxide degrades.

Since microorganisms are continually being added to the ore body with the leach s utions, and microbes, adsorbed onto soil particles, can be desorbed by the increased flow associated with leaching, it will be necessary for hydrogen peroxide to be injected at regular intervals to prevent buildup of microorganisms near the production wells. Before hydrogen peroxide can be used efficiently to prevent plugging problems at in situ mining operations, the concentrations of hydrogen peroxide necessary to destroy the microbes, the distance through which the hydrogen peroxide can travel without losing its effectiveness, and the necessary duration and frequency of the hydrogen peroxide treatment must be determined. In addition, the potential problem of decreased permeability due to pore blockage by oxygen (created from hydrogen peroxide degradation), as noted by other investigators (8, 9, 12), must be further investigated.

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