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Mathematical Modeling of In-Situ Uranium Leaching

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Abstract

This paper presents the development and results of a computer model of in-situ uranium leaching. This model uses a streamline-concentration balance approach and is useful with a wide range of reservoirs. It can be used with any type of well system, in a reservoir with or without boundaries, and with any form of descriptive kinetics. The model also includes the effects of dispersion and consumption of oxidant by minerals other than uranium. The effects of well pattern, variable uranium concentrations, and the presence of oxidant consumers on uranium production are discussed.

Introduction

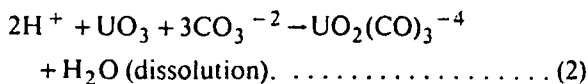
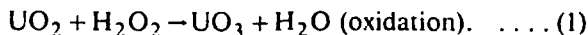
The sandstone uranium deposits of south Texas represent a possible major energy source. These deposits consist mainly of widely scattered roll fronts (pods) of unoxidized uranium minerals in loosely packed sands.¹ It is thought that these deposits were formed by the downdip migration of groundwater carrying oxidized uranium leached from the host rock, Catahoula Tuff. When the uranium-bearing waters reached a reducing zone, the uranium was precipitated, forming mainly the mineral uraninite, UO₂. Much of the uranium ore in the area is low grade (<0.05% U₃O₈) and is at depths of 100 to 1,500 ft.

Since 1960, various companies have been mining some of the higher-grade deposits to depths of up to 200 ft, using conventional strip-mining techniques. The concomitant surface disruption is extensive, and the costs of mining and transporting to a mill such large amounts of material prohibit the utilization of low-grade ore.

A mining technique that may overcome these difficulties to some extent and ultimately make more of the south Texas uranium deposits amenable to

recovery is in-situ solution mining. This technique consists of pumping through the ore body a chemical solution that will dissolve the uranium minerals so that they may be leached from the ore and recovered from the solution. For this process to be economically feasible, a low-cost solution must be available that will dissolve a large portion of the uranium present, the uranium must be easily recoverable from the leach solution, the physical attributes of the ore body must be such that the leach solution can be pumped through the ore without great loss to the surroundings, and environmental hazards must be avoided.

The leaching process and its chemistry are basically simple. Uranium is generally found to have one of two oxidation states - oxidized, U⁺⁶, or unoxidized, U⁺⁴. In the oxidized +6 state, uranium forms many soluble ions, among them the uranyl ion UO₂²⁺, the uranyl dicarbonate ion UO₂(CO₃)⁻², and the uranyl tricarbonate ion UO₂(CO₃)₃⁻⁴. Hostetler and Garrels³ have investigated the equilibria of uranium minerals with natural solutions and found that under oxidizing conditions, stable soluble ions exist over a wide range of pH. The results suggest that to dissolve uranium minerals, one must provide an oxidizing agent to oxidize reduced uranium to the +6 state and a complexing agent that will form stable complex ions with U⁺⁶. A typical set of reactions is as follows:



The oxidation step represented by Eq. 1 uses

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hydrogen peroxide as the oxidant. It is also possible to oxidize the uranium using air, pure oxygen, or sodium chlorate. Hydrogen peroxide is being used in most present operations, though evidence⁴ suggests that it dissociates rapidly to water and oxygen at reservoir conditions. If this is the case, pure oxygen may be the preferred oxidant because it is less expensive than hydrogen peroxide.

The physical operation of an in-situ uranium solution mine is similar in many respects to conventional oilfield operations. After a suitable ore body has been located and mapped by exploratory drilling, a pattern of wells must be placed to recover a large fraction of the value in a reasonable time. A second factor concerns confining the leach solution to the region of interest to avoid environmental problems. Often, this can be achieved by the efficient placement of production and injection wells; in other cases, guard wells may be required. This paper considers the problem of well placement, the use of guard wells, uranium production rates, the effect of oxidant concentration, and many other questions concerning the in-situ leaching of uranium. The approach used here is to develop a computer model that can be used to simulate a variety of production situations. The results can then be compared and reasonable operations parameters identified.

The Flow Model

In this model, the flow is assumed to be single phase and two-dimensional so that the fluid velocity at a point can be calculated using the well-known source and sink equations that describe the flow of an incompressible fluid through a porous medium as a function of the location and volume rates of production and injection wells (see Appendix A). Given the fluid velocities, streamlines can be traced. A typical example is shown in Fig. 1.

Once streamlines have been defined, pressure drop along them can be calculated. In the approach used

here, the streamlines are divided into equal pressure increments or nodes that are used later in developing a finite-difference analog of the concentration balance. The fluid velocity at each node is recorded for future use in finite-difference equations.

The streamline model can be modified to approximately account for variations in reservoir thickness, provided they can be assumed to change linearly between wells.⁵ The calculation requires that the formation thickness be known at each point along a streamline. These values of thickness are entered using a grid system over the area of the pod, as shown in Fig. 2. This information is used to adjust the fluid velocities at each node. In the subsequent calculation of both uranium and oxidant concentrations, the modified velocities will be used. In this way, thickness variations are considered.

Most uranium pods are small in area compared with the aquifers in which they occur, and these are effectively unbounded systems as represented by Fig. 1. In many cases, it would be advantageous to minimize lixiviant escape from uranium-rich zones, because it is not then wasted on nonproductive areas, recovery rates are increased, and aquifer contamination is minimized. This last consideration is by far the most important one. To bound the system of production and injection wells, it is possible to drill additional wells (guard wells) outside the pod area. By properly adjusting the location and injection rates of groundwater into them, guard wells can be used to create a no-flow boundary around the periphery of the pod. The model uses a method of image wells developed by Lin⁶ to simulate the bounding that can be achieved by the proper injection rates through a given system of guard wells. Therefore, the program can be used to estimate how many guard wells are required to achieve effective bounding by varying their number and spacing until a satisfactory result is obtained. Of course, there are an infinite number of combinations that will provide adequate bounding.

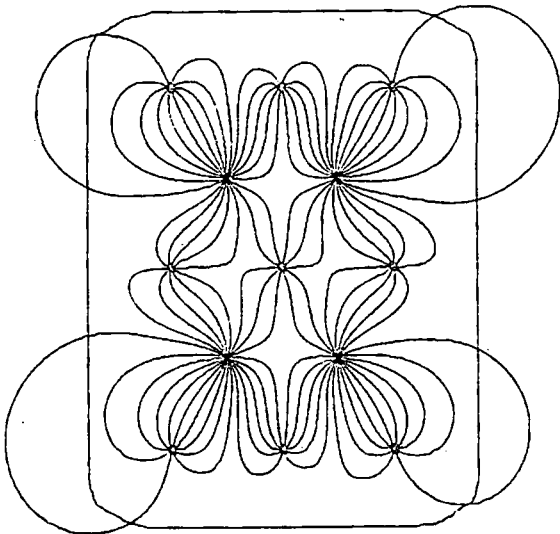


Fig. 1 - Streamline pattern for a regular, inverted five-spot in an unbounded aquifer. Rectangular region represents boundaries of the mineralized zone.

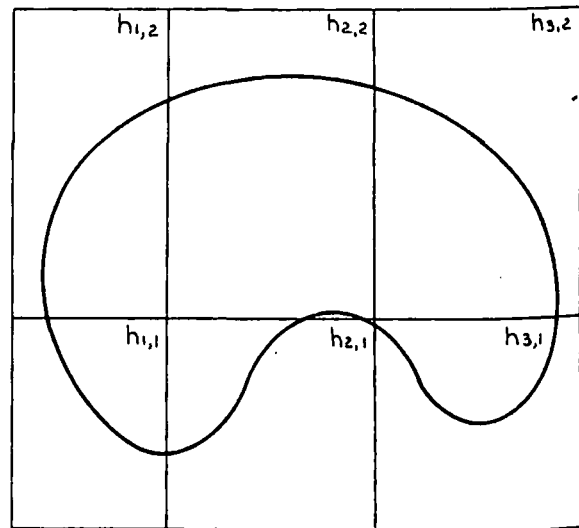


Fig. 2 - Typical thickness grid superimposed on the mineralized zone. It is used for calculating fluid velocities.

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To determine the "best set," economic considerations are required. Such considerations are beyond the scope of this work.

The streamlines for a bounded system are shown in Fig. 3; if these results are compared with those of Fig. 1, the effect of confining the streamlines is evident. The guard wells distort both the streamlines and the fluid velocities from those of the unbounded system. Both these changes are taken into account when calculating uranium production.

Uranium Balance

Once the flow system has been defined, bounding has been completed if desired, and each streamline has been divided into nodes at which velocities are stored, the next step is to calculate the uranium concentration at each of the nodes as a function of time. This is done using a component balance that allows for chemical reactions and dispersion. The balance equation is first written in a rectangular coordinate system that includes the effect of dispersion and then is converted into one in which the streamlines and potential lines form the coordinates (see Appendix B). Assuming that dispersion along a line of constant potential (perpendicular to flow) can be neglected, the component balance can be written along a streamline as

$$\alpha(\Phi, \psi) \frac{\partial c}{\partial \Phi} + \beta(\Phi, \psi) \frac{\partial^2 c}{\partial \Phi^2} - R = \phi \frac{\partial c}{\partial t}, \quad \dots \dots (3)$$

where α and β depend on the velocities, which are known but are independent of the concentration, c .

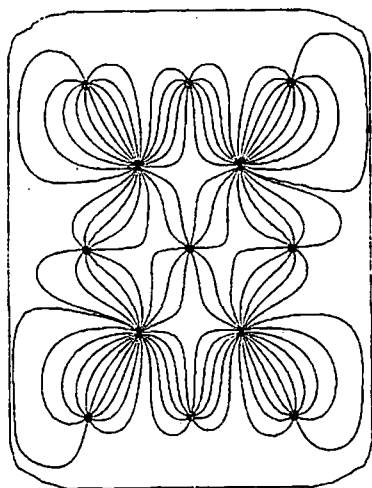


Fig. 3 - Streamline pattern for a regular, inverted five-spot bounded by guard wells around the mineralized zone.

R is the reaction term that describes the rate at which uranium is oxidized and leached from the solid surface, and Φ is the potential function that, for a horizontal system, is the pressure measured along a streamline. The form of the reaction rate used in this paper is

$$R_{Ur} = \epsilon (W_{Ur} - W_{RUr})(1 - \phi)\rho. \quad \dots \dots (4)$$

This rate expression was found to adequately describe the flow results reported by Galichon *et al.*⁴ The results presented here are not general in that the constants used in the rate expression are expected to vary from site to site. It is necessary then to develop the appropriate rate expression using ore samples from the deposit of interest and using the leach solution to be employed in the field.

It should be noted that ϵ depends on number of factors, including the oxidant concentration. The calculations reported here use an experimentally determined relationship between the rate constant (ϵ) and the peroxide concentration (the equivalent oxygen partial pressure would serve equally well). Grandstaff⁷ finds that the rate is proportional to the partial pressure when pure uraninite is dissolved in carbonate solutions at temperatures ranging from 2° to 23°C. Pearson and Wadsworth⁸ found the rate constant depends on the square root of the partial pressure at higher temperatures. The experimentally determined ϵ used here is shown in Fig. 4. The other factors given in Eq. 4 have been discussed previously.⁴ Eq. 3 is solved for each streamline at selected times. It is solved using a fully discrete, finite-difference approximation with an unconditionally stable Gaussian row reduction

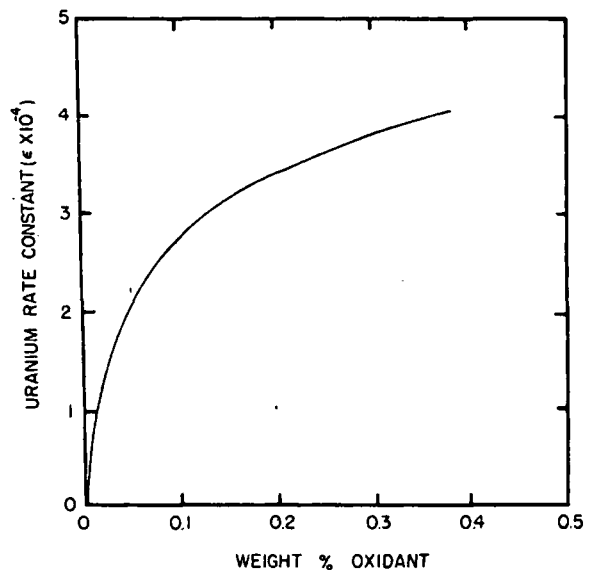


Fig. 4 - The uranium rate constant shown as a function of oxidant concentration (from Galichon's data.¹²)

technique.⁹ The finite-difference analogy to Eq. 3 uses an implicit technique with a two-point backward difference for the derivative of concentration with respect to time. The derivatives of concentration with respect to pressure are expressed using five-point centered differences at nodes between wells and three-point centered differences at the nodes next to wells. The three-point forms must be used near wells to maintain the pentadiagonal form of the coefficient matrix that is solved by the Gaussian technique. This finite-difference analogy has been shown to generate good results. Details of the computer program and the testing of the numerical procedure are available elsewhere.¹⁰ The output from the balance is the uranium concentration at each node of each streamline at every time step. The uranium concentration at each wellbore can then be calculated.

Other Modeling Considerations

Most aquifers contain varying concentrations of minerals, such as pyrite, that will compete with uranium for oxidant. These minerals consume some of the injected oxygen. Therefore, the presence of these minerals has the effect of delaying uranium production. Again, a laboratory study must be conducted using samples from the ore body of interest to determine the reaction expressions required to describe the minerals' effects.

To demonstrate the impact of these oxidant-consuming minerals, a reaction-rate expression is used in an oxidant-balance equation (having the same form as Eq. 3) to calculate the oxidant concentration at each node as a function of time. This rate expression is

$$R_o = \zeta M C_o / \delta \dots \dots \dots (5)$$

This expression has not been tested yet, but clearly ζ and δ will vary from formation to formation. The initial values of the oxidant-consuming mineral concentrations are stored and used in the rate expression and as the reaction proceeds:

$$\frac{dM}{dt} = - \delta R_o \dots \dots \dots (6)$$

This equation is integrated and the mineral concentration is allowed to decrease with time. Thus, given the amount of oxidant left at a node after reaction with the oxidant-consuming minerals, the appropriate uranium reaction-rate constant, ϵ , can be located from Fig. 4 and used in Eq. 4. Even if there are no oxidant-consuming minerals (save uranium) present in the pod, the oxidant balance must be solved together with the uranium balance to determine the oxidant concentration present at a particular node at a particular time and hence obtain the proper uranium reaction-rate constant.

Because most reservoirs are not homogeneous chemical systems, the model has been built to consider these differences. The same grid system used to store differences in reservoir thickness for the velocity approximation can be used to provide concentration variances. The uranium and pyrite

concentrations over the entire reservoir are stored along with the thickness changes in the grid system. They now can be located by the position of each node on each streamline and used in the oxygen and uranium concentration calculations.

In unbounded or partially bounded systems, some of the streamlines escape beyond the uranium-bearing zone, as shown in Fig. 1. The portions of a streamline that lie outside the uranium-bearing area do not contribute any new uranium production to the system. They may, however, contain pyrite since the process that deposited pyrite probably was not the same one that deposited uranium. Thus, along these portions of a streamline outside the uranium-bearing area, the oxidant may continue to be consumed. To consider this phenomenon, each node along a streamline is tested to determine if it is inside or outside the uranium-bearing area. This test is performed by taking the cross-product of two vectors. The first vector passes through the node and the closest boundary point to that node. The boundary points are a finite set of discrete points used to define the uranium-bearing area. The second vector passes through the closest boundary point and the next point taken in a clockwise direction, as shown in Fig. 5. If the cross-product is positive, then the node is inside the pod. If it is negative then the node is outside the pod and no uranium production is calculated. The only instability in this method occurs when the node, closest boundary point, and second boundary point are all on line, thus forming an angle of 180°. When this occurs, the cross-product is zero and no distinction can be made. Therefore, in the event that this happens, the model uses the boundary point behind the closest boundary point to form the first vector and that point and the closest boundary point to form the second vector.

Results

The economic viability of a particular leaching

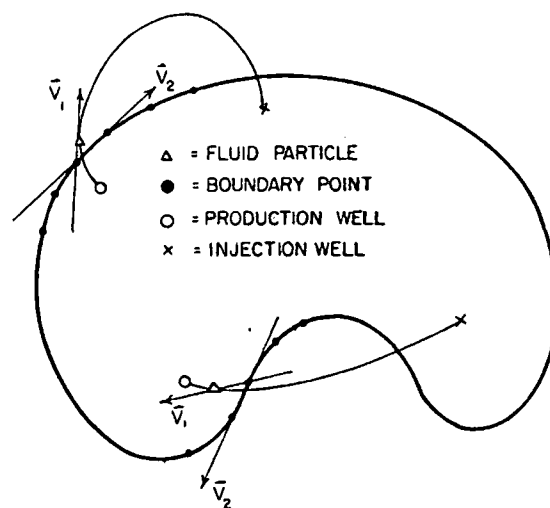


Fig. 5 - Sketch showing the vector relationships used to determine whether a point is within the mineralized zone.

project is related to both the concentration of the uranium in the produced leach solution and the time required to produce a significant portion of the mineral values. It is, therefore, critical to design a well pattern that will leach as much uranium-bearing rock as possible over a particular time period, while also minimizing the number of guard wells required to prevent the leaching solution from escaping into the aquifer. Theoretically, any well pattern will eventually sweep a pod given enough time. Thus, for a given time frame, the well pattern that recovers the most uranium would be judged the most efficient and probably the most desirable.

The importance of areal sweep has been demonstrated using the unbounded, inverted five-spot pattern of Fig. 1, the unbounded line-drive pattern of Fig. 6, and the unbounded, expanded, inverted five-spot pattern of Fig. 7. The production histories of these patterns after 6 months of production are presented in Fig. 8. It can be seen from this figure that even though the line drive has more wells than either of the other two patterns, it does not produce more uranium than the regular five-spot pattern. However, due to its greater areal sweep, the expanded inverted five-spot pattern produces in the given time roughly 1.4 times the uranium of the other two patterns. There is, however, a longer delay initially with the expanded, inverted five-spot because of the increased distance between wells.

As outlined earlier, if any oxidizable minerals such as pyrite are present in a pod, the uranium production should be delayed. To illustrate this, the system shown in Fig. 1 was modeled for 6 months, first with and then without pyrite. The production profiles are contrasted in Fig. 9. The presence of pyrite causes a marked delay in uranium production.

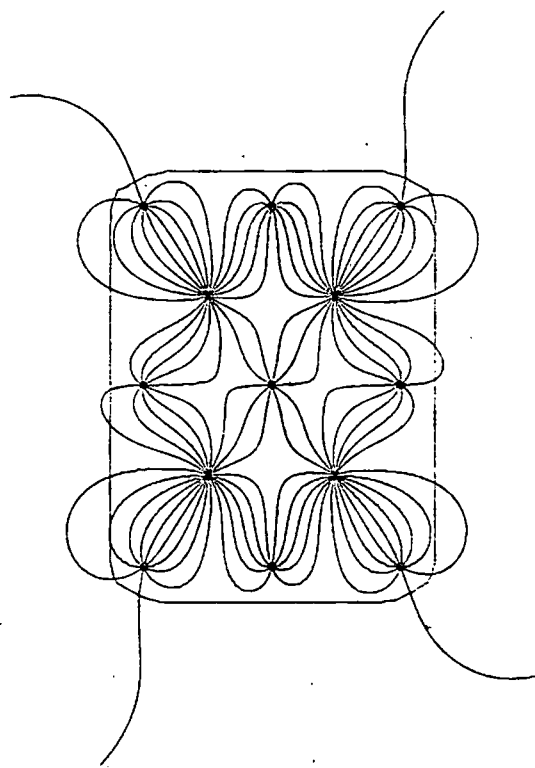


Fig. 7 - Streamline pattern for an expanded, inverted five-spot in an unbounded aquifer.

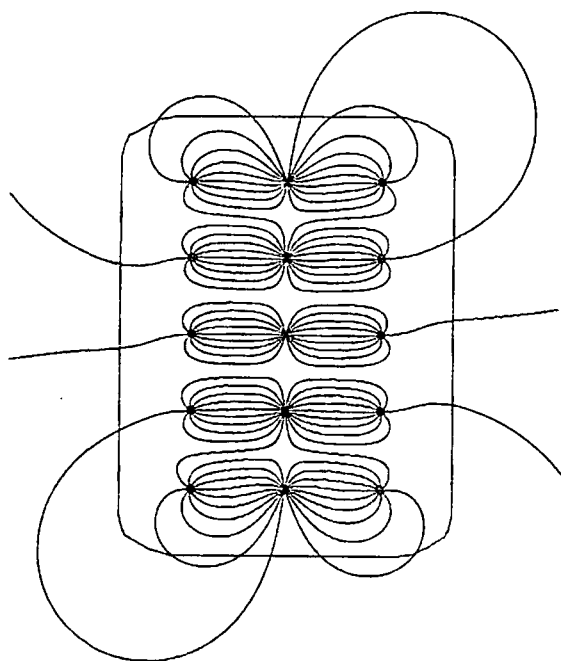


Fig. 6 - Streamline pattern for a line drive in an unbounded aquifer.

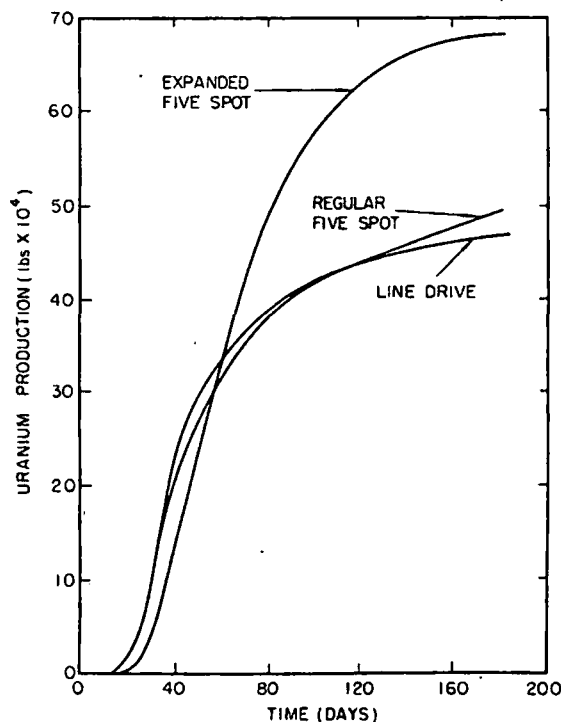


Fig. 8 - Cumulative uranium production with time as it depends on areal sweep, with all other variables held constant.

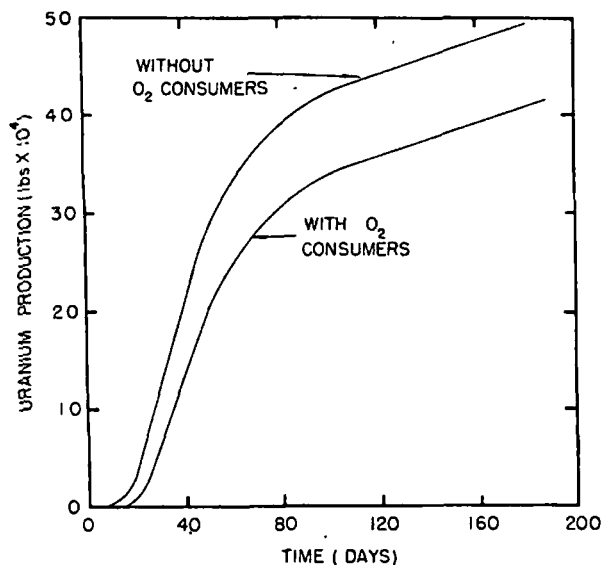


Fig. 9 - The effect of oxygen-consuming minerals in delaying uranium production.

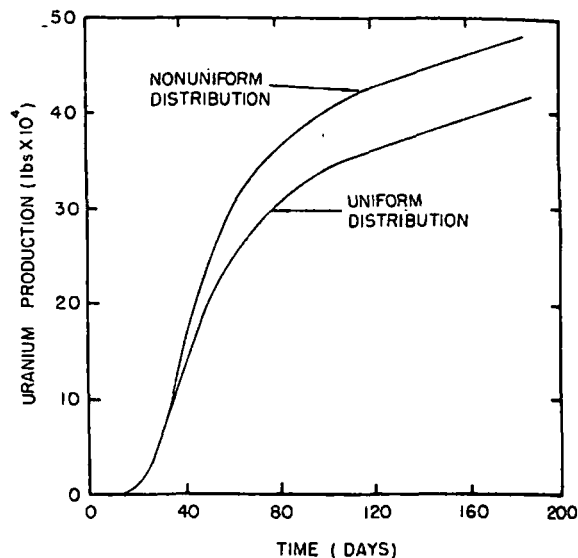


Fig. 10 - The effect of varying uranium concentrations within the mineralized zone.

With enough time, the uranium production will tend to that of the pod without any pyrite. Here again, the extent of the delay depends on the amount of oxidizable minerals present and their rate of reaction with oxygen.

Variable uranium content can have a dramatic effect on production. The pod represented in Fig. 1 was modeled first with a uniform initial uranium concentration of 0.003 g uranium per gram of rock and a second time with one-half the pod having negligible uranium concentrations, one-quarter having 0.002 g uranium per gram of rock, and one-quarter having 0.0093 g uranium per gram of rock. The average concentration for the second case was 0.003 g uranium per gram of rock. The production profiles shown in Fig. 10 show that the pod with the one high concentration quarter produced more uranium in the given time than the pod with the uniform concentration. This result is dependent on the rate expression used for the uranium reaction, and in actual situations will vary from site to site. The rate expression used for these examples, as seen in Eq. 4, contains the difference between the uranium present and unleachable or residual uranium, $W_{Ur} - W_{RUr}$. For the uniform pod this term has a value of 0.002 g uranium per gram of rock. In the nonuniform pod it has a value of 0.0083 g uranium per gram of rock, which is 4.15 times greater than in the uniform pod. So, if the effects of the other terms in the rate expression are neglected, the nonuniform pod should produce 4.15/4 or 1.04 times faster than the uniform pod. This figure is a bit lower than what actually occurred, but it points out that the rate expression controls how much uranium is produced from a pod in a given time, and the effects of high concentrations of uranium in a pod.

Summary

An in-situ uranium leaching model to predict the movement of reservoir fluid and to predict the

production and accumulation of fluid volumes and leachate concentrations has been described. It can be used to optimize well patterns and guard-well systems. The model can be used with any size and shape reservoir with variable thickness, with or without boundaries, with any configuration of wells, with or without guard wells, with areal differences in reservoir makeup, and with any form of descriptive kinetics. The model also considers dispersion and the consumption of injected oxygen.

The results indicate the importance of proper well-pattern selection and the possible benefits of guard wells. Economics determine their use.

Also, it has been found that significant variations in uranium concentrations in the produced fluids may be observed in most situations. These can result from any one or more of a number of factors, including variations in reservoir thickness or permeability, nonhomogeneous distribution of uranium values, or variations in concentration of oxidant-consuming minerals.

Nomenclature

- c = concentration
- D_o = molecular diffusion coefficient
- F = formation resistance factor
- h = formation thickness
- k = permeability
- K = dispersion coefficient
- M = pyrite concentration
- P = average formation particle diameter
- q = production or injection rate
- R = reaction rate
- RI = 2% of the average distance between wells
- t = time
- u = flux
- v = fluid velocity
- W = uranium concentration

- x, y = rectangular Cartesian coordinates
- α = a complex function of velocity appearing in the concentration balance.
- β = a complex function of velocity appearing in the concentration balance
- δ = stoichiometric coefficient
- Δ = increment
- ϵ = uranium reaction-rate constant, time⁻¹
- ζ = oxidant reaction-rate constant
- μ = fluid viscosity
- ρ = density
- σ = formation factor
- ϕ = porosity
- Φ = potential

Subscripts

- L = longitudinal
- m = mean
- n = number of wells
- o = oxygen or oxidant
- R = residual
- t = time
- Ur = uranium
- x, y = rectangular coordinate direction

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**APPENDIX A
Streamline Generation**

Starting with the Laplace line-source and sink equation

$$p(x,y) = p_m - \frac{\mu}{4\pi kh} \sum_{i=1}^n q_i \ln \{ (x-x_i)^2 + (y-y_i)^2 \} \dots \dots \dots (A-1)$$

Eq. A-1 can be differentiated with respect to x and y to obtain expressions for the vectorial components of velocity

$$v_x(x,y) = \frac{1}{2\pi\phi h} \sum_{i=1}^n q_i \frac{(x-x_i)}{(x-x_i)^2 + (y-y_i)^2} \dots \dots \dots (A-2)$$

$$v_y(x,y) = \frac{1}{2\pi\phi h} \sum_{i=1}^n q_i \frac{(y-y_i)}{(x-x_i)^2 + (y-y_i)^2} \dots \dots \dots (A-3)$$

Define RI as 2% of the average distance between wells and assume that the velocity remains constant over this small distance. Sum the vectorial components of velocity to obtain the total velocity:

$$v(x,y) = (v_x^2 + v_y^2)^{1/2} \dots \dots \dots (A-4)$$

The time required for a fluid particle to move the distance RI is

$$\Delta t = RI/v(x,y) \dots \dots \dots (A-5)$$

The new location of the fluid particle along a streamline can be calculated by

$$x_{i+1} = x_i + v_x(x,y) \Delta t, \dots \dots \dots (A-6)$$

$$y_{i+1} = y_i + v_y(x,y) \Delta t. \dots \dots \dots (A-7)$$

This procedure is repeated for each streamline of each production well. Thus, the streamlines are traced between production and injection wells.

**APPENDIX B
Concentration Balance**

A concentration balance, which includes dispersion that varies in the axial as compared with the radial direction, can be written as

$$\begin{aligned} & -u_x \frac{\partial c}{\partial x} - u_y \frac{\partial c}{\partial y} + \frac{\partial}{\partial x} \left(K_{xx} \frac{\partial c}{\partial x} \right) \\ & + \frac{\partial}{\partial x} \left(K_{xy} \frac{\partial c}{\partial y} \right) + \frac{\partial}{\partial y} \left(K_{yx} \frac{\partial c}{\partial x} \right) + \frac{\partial}{\partial y} \left(K_{yy} \frac{\partial c}{\partial y} \right) \\ & - R = \phi \frac{\partial c}{\partial t} \dots \dots \dots (B-1) \end{aligned}$$

At this point, it is necessary to convert Eq. B-1 from rectangular coordinates to stream-function and potential coordinates. Eq. B-1 can be written as follows, provided it is assumed that dispersion perpendicular to flow can be neglected.

$$\frac{\mu}{k} \left\{ u^2 - K_L \left(1 + \frac{u_x^2}{u^2} \right) \frac{\partial u_x}{\partial x} - K_L \left(\frac{2u_x u_y}{u^2} \right) \frac{\partial u_x}{\partial y} - K_L \left(1 + \frac{u_y^2}{u^2} \right) \frac{\partial u_y}{\partial y} \right\} \frac{\partial c}{\partial \Phi} + \left(\frac{\mu}{k} \right)^2 \left\{ \left(\frac{u_x^2 + u_y^2}{u} \right)^2 \cdot K_L \right\} \frac{\partial^2 c}{\partial \Phi^2} - R = \phi \frac{\partial c}{\partial t} \quad \text{..... (B-2)}$$

Define

$$\alpha = \frac{\mu}{k} \left\{ u^2 - K_L \left(1 + \frac{u_x^2}{u^2} \right) \frac{\partial u_x}{\partial x} - K_L \left(\frac{2u_x u_y}{u^2} \right) \frac{\partial u_x}{\partial y} - K_L \left(1 + \frac{u_y^2}{u^2} \right) \frac{\partial u_y}{\partial y} \right\} \quad \text{..... (B-3)}$$

$$\beta = \left(\frac{\mu}{k} \right)^2 \left\{ \left(\frac{u_x^2 + u_y^2}{u} \right)^2 K_L \right\} \quad \text{..... (B-4)}$$

where

$$K_L = D_o / F\phi + 0.5\nu\sigma P,$$

which is the expression for longitudinal dispersion described by Perkins and Johnston.¹¹

All the terms in α and β are either known from the streamline generator or can be calculated. So, the concentration balance, Eq. B-2, can be reduced to

$$\alpha(\Phi, \psi) \frac{\partial c}{\partial \Phi} + \beta(\Phi, \psi) \frac{\partial^2 c}{\partial \Phi^2} - R = \phi \frac{\partial c}{\partial t} \quad \text{..... (B-5)}$$

The R term is the rate expression for either uranium or mineral oxidation. **SPEJ**

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