Theoretical Study of Fluid Flow Accompanied by Solid Precipitation in Porous Media

CHIANG-HAI KUO and PHILIP J. CLOSMANN
Shell Development Company, Houston, Texas

This paper deals with compressible fluid flow accompanied by solid deposition in a porous reservoir. The isothermal fluid flow is considered to be radial and follows Darcy's law. The initially dissolved substances precipitate from the solution as a result of solubility reduction as the pressure declines. These deposits accumulate in the void spaces and hence inhibit the flow. An idealized model describing this transport mechanism has been proposed. On the basis of this model, a numerical technique has been developed to predict the amount of solid precipitation and the pressure distribution as functions of time and radial distance.

The method is applied to the production performance of a reservoir containing mainly hydrogen sulfide saturated with elemental sulfur. The solid sulfur builds up rapidly in the vicinity of the production well. A very negligible amount is formed close to the impermeable outer boundary. It is shown that plugging by solid sulfur in the porous medium can be reduced either by reducing the production rate or by choosing closer well spacing.

The flow of fluids in underground porous media may be complicated in at least two respects. First, fluids often occur naturally in a reservoir as a multicomponent system. Second, phase transformation often takes place during the course of flow. Because of the complex character of this multicomponent, multiphase flow behavior, a theoretical prediction of production performance cannot be easily formulated.

Among others, West, Garvin, and Sheldon (*10*) investigated the transient behavior of underground two-phase flow. They studied the case in which a gas phase is formed and separated from a liquid phase as a reservoir is depleted. A recent report by Kniazeff and Naville (*4*) on condensing gas flow covers another feature of the problem.

Still another phenomenon affecting underground fluid flow is precipitation of solid material from the fluid phase. The production of a reservoir fluid under such conditions could be troublesome if the deposits were to accumulate in significant amounts in the void spaces and thus inhibit the flow. A typical example, as will be discussed in detail later, is production of a dense fluid consisting mainly of hydrogen sulfide saturated with sulfur. An underground reservoir containing such a fluid has been found recently in Texas (*7*). Elemental sulfur precipitates from the solution as a result of solubility reduction as the pressure declines. The deposit will finally fill up such a portion of the pore space, especially in the vicinity of a production well, that serious plugging of the porous medium will occur.

Since no information on field experience is available in the literature, it is desirable to treat this problem theoretically. A model is proposed in this paper to describe the deposition of a solid phase and its effects on fluid flow behavior. The amount of solid precipitation and the pressure distribution in the reservoir formation are predicted by numerically solving a set of nonlinear partial differential equations. The optimum design of a scheme for producing this type of fluid is also suggested.

THEORETICAL MODEL

An idealized model of the homogeneous porous reservoir considered in this study is sketched in Figure 1. This reservoir, with a finite thickness h, is bounded by impermeable cap and base rocks and by an impermeable cylindrical outer boundary of radius r_e. The vertical velocity component and gravity effects are considered to be unimportant. In this case the velocity of isothermal horizontal fluid flow through this porous medium is governed by the one-dimensional form of Darcy's law (*6*):

$$v = -\frac{k}{\mu}\frac{\partial p}{\partial r} \tag{1}$$

The effective permeability to the flowing fluid k is a fraction of the total rock permeability K and is normally a function of the fluid saturation S. The fluid viscosity μ varies only slowly with the reservoir pressure p and will be assumed constant in this work.

As the reservoir is depleted the contained fluid flows radially toward the production well, which is located at the center of the reservoir. Some of the initially dissolved

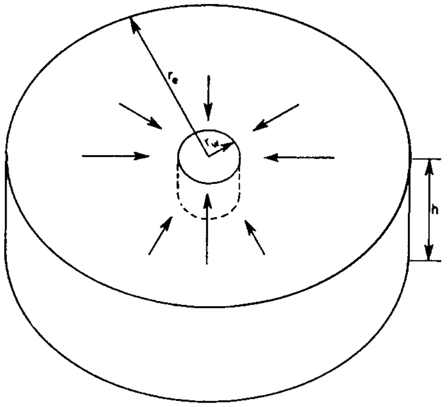

Fig. 1. Idealized model.

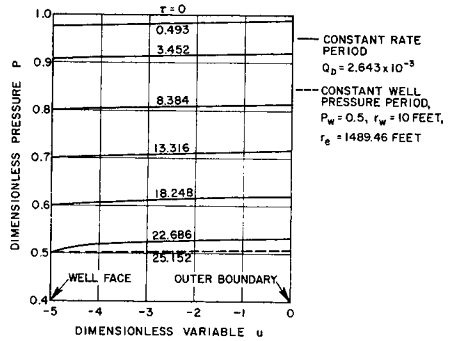

Fig. 2. Pressure distribution.

component precipitates from the solution as a result of solubility reduction as the pressure declines. The solid deposits formed are incapable of flow and accumulate in the void spaces. This assumption is reasonable, in view of the experimental results of Kennedy and Wieland (*3*). The equations of continuity for the fluid and the solid components can be written as

$$\frac{1}{r}\frac{\partial}{\partial r}(r\rho_f^f v) = -\phi\frac{\partial}{\partial t}(\rho_f^f S) \tag{2}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r\rho_s^f v) = -\phi\frac{\partial}{\partial t}[\rho_s^f S + \rho_s^s(1-S)] \tag{3}$$

where ρ_s^f denotes the mass of the solidifying component per unit volume of the solution and the porosity ϕ is assumed constant. The fraction of the volume occupied by liquid is $S\phi$. The fraction occupied by deposited solid is $(1 - S)\phi$. Defining the fluid mobility parameter M as

$$M = \frac{\dfrac{k}{K}}{\dfrac{\mu}{\mu_i}} \tag{4}$$

and the two dimensionless independent variables

$$u = \ln\frac{r}{r_e} \tag{5}$$

and

$$\tau = \frac{Kp_i t}{\phi r_e^2 \mu_i} \tag{6}$$

and incorporating Equation (1), we may obtain the following from Equations (2) and (3) (*10*):

$$e^{-2u}\frac{\partial}{\partial u}\left(\rho_f^f M\frac{\partial P}{\partial u}\right) = \frac{\partial}{\partial \tau}(\rho_f^f S) \tag{7}$$

$$e^{-2u}\frac{\partial}{\partial u}\left(\rho_s^f M\frac{\partial P}{\partial u}\right) = \frac{\partial}{\partial \tau}[\rho_s^f S + \rho_s^s(1-S)] \tag{8}$$

The dependent variables are the dimensionless pressure P and the fluid saturation S. By expanding and eliminating terms involving saturation derivatives in Equations (7) and (8), we obtain Equation (9):

$$e^{-2u}\left[\frac{\partial}{\partial u}\left(M\frac{\partial P}{\partial u}\right) + aM\left(\frac{\partial P}{\partial u}\right)^2\right] = [aS + b(1-S)]\frac{\partial P}{\partial \tau} \tag{9}$$

Elimination of $(\partial/\partial u)[M(\partial P/\partial u)]$ from Equations (7) and (9) yields Equation (10):

$$-e^{-2u}(a+c)M\left(\frac{\partial P}{\partial u}\right)^2 = \frac{\partial S}{\partial \tau} - [(a+c)S + b(1-S)]\frac{\partial P}{\partial \tau} \tag{10}$$

where

$$a = \frac{1}{\rho_s^s}\frac{\partial \rho_s^f}{\partial P} + \frac{\rho_s^s - \rho_s^f}{\rho_f^f \rho_s^s}\frac{\partial \rho_f^f}{\partial P} \tag{11}$$

$$b = \frac{1}{\rho_s^s}\frac{\partial \rho_s^s}{\partial P} \tag{12}$$

and

$$c = -\frac{1}{\rho_f^f}\frac{\partial \rho_f^f}{\partial P} \tag{13}$$

The fluid densities are functions of the pressure, but that of the solid phase is considered to be constant ($b = 0$). Equations (9) and (10) are solved numerically to obtain P and S as functions of u and τ for the following initial and boundary conditions.

Initially, the pressure is uniform across the formation, and the pore space contains only saturated fluid. At the impermeable outer radius there is no flow at any time. The well is produced at a constant mass rate before the pressure at the well face drops to a minimum value. Thereafter the rate is decreasing, while the minimum pressure is maintained at the well face. The well is shut in when the rate has declined to less than 5% of its initial value. These initial and boundary conditions can be mathematically stated as

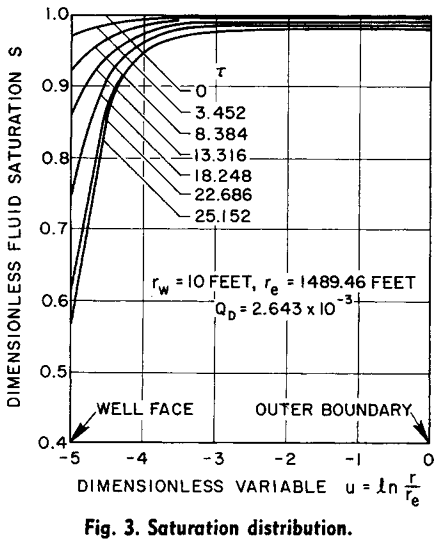

Fig. 3. Saturation distribution.

$$\left.\begin{array}{l} 0 > u > \ln\dfrac{r_w}{r_e}, \quad \tau = 0, \quad P = 1, \quad S = 1, \\ u = 0, \quad \tau > 0, \quad \dfrac{\partial P}{\partial u} = 0, \\ u = \ln\dfrac{r_w}{r_e}, \begin{cases} \tau_m > \tau > 0, & Q_D = \dfrac{\rho_f}{\rho_f^*} M \dfrac{\partial P}{\partial u} = \text{constant}, \\ \tau > \tau_m, & P = P_w, \end{cases} \end{array}\right\} \tag{14}$$

where Q_D is the dimensionless mass flow rate, related to the flow rate Q by the equation

$$Q = -Q_D\left(\frac{2\pi K h p_i \rho_f^*}{\mu_i}\right) \tag{15}$$

A numerical method has been developed to solve Equations (9) and (10) with these initial and boundary conditions. The details of this method are described in the Appendix.*

APPLICATION TO SULFUR-HYDROGEN SULFIDE SYSTEM

The developed method has been applied to the production performance of a reservoir containing mainly hydrogen sulfide saturated with elemental sulfur. This system has been found to exist in an underground rock formation (*7*). Furthermore, the fluid properties and the phase behavior of this mixture have been widely studied (*3*, *8*). For instance, the solubilities of sulfur in the methane-carbon dioxide-hydrogen sulfide system were measured by Kennedy and Wieland (*3*). They showed that the solubility of sulfur is greater for a higher content of hydrogen sulfide in the mixture at a given pressure and temperature. If a reservoir fluid is initially undersaturated with sulfur, the deposition of solid sulfur occurs at a later time during the production. In order to give a specific example, it is assumed that before production starts, the hydrogen sulfide-sulfur mixture exists in an underground reservoir at 5,000 lb./sq. in. and 200°F. as a dense fluid. The minimum well pressure is arbitrarily specified as 2,500 lb./sq. in. in order to exclude conditions where more than two phases occur. Within this pressure range the solubility data of Kennedy and Wieland (*3*) and the compressibilities measured by Reamer, Sage, and Lacey (*8*) are correlated to obtain the following approximate functions:

$$c = -0.3863 + 0.2438P \tag{16}$$

and

$$a = 0.7663 - 0.2438P \tag{17}$$

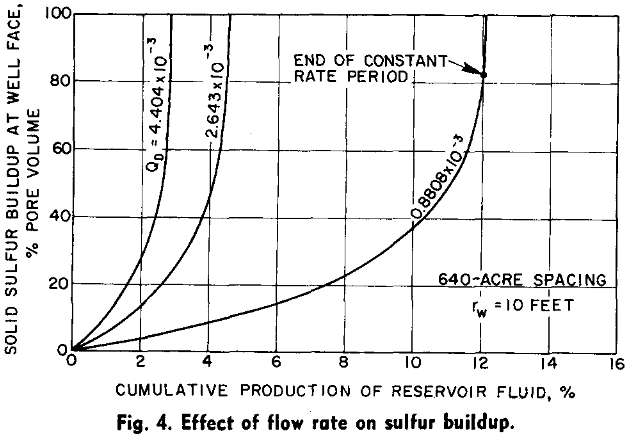

Fig. 4. Effect of flow rate on sulfur buildup.

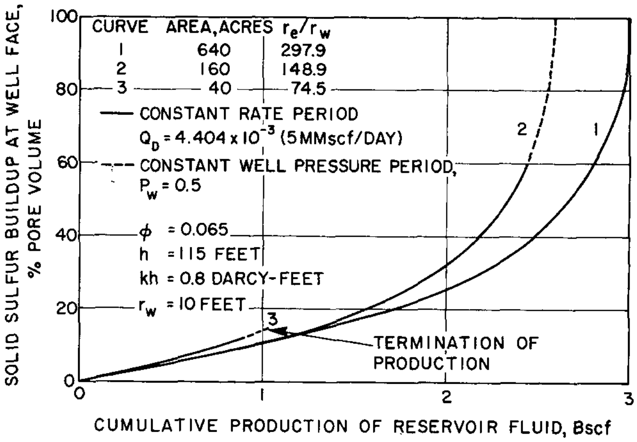

Fig. 5. Effect of well spacing on sulfur buildup.

The following equation is used for the dimensionless mobility in a typical reservoir (*1*):

$$M = e^{-5.65(1-s)} \tag{18}$$

From these formulas the prediction of sulfur deposition can be made. The quantity $(1 - S)\phi$ is the fractional volume of sulfur deposited. With a well bore of a few inches radius, the pore space in the vicinity of a well face is filled up with precipitated sulfur before 1% of the reservoir fluid is produced. Thus, very frequent cleaning of the formation for sulfur removal is required.

Fortunately, a large effective well radius can be assumed for some cases. As an example, the use of acidization is well known in the field of petroleum engineering. For illustrative purposes the effective well radius r_w is taken as 10 ft. in these calculations. A number of well spacings are assumed, ranging from 10 acres ($r_e = 372.37$ ft.) to 640 acres ($r_e = 2978.92$ ft.), and three different production rates are employed.

Typical pressure and saturation profiles are shown in Figures 2 and 3 for one case. From Figure 2 it can be seen that the pressure gradient at the well face increases with time before the pressure declines to the specified minimum well pressure. The fluid can then no longer be produced at the specified constant mass rate. The prediction is terminated when the production rate becomes less than 5% of the initial value. In Figure 3 the saturation distribution is illustrated as a function of time. The deposited solid accumulates mostly in the vicinity of the well face, whereas a high fluid saturation is maintained for the remainder of the formation. This indicates that if cleaning of the formation for sulfur removal is desirable, it is necessary to clean only a short distance out from the well face. Figure 4 shows the effect of flow rate on sulfur buildup at the well face. A low producing rate has the advantage of achieving high cumulative production for a given sulfur saturation at the well face. This is because the lower rate of pressure decline results in a lower rate of buildup of solid sulfur. This in turn permits longer production before a given solid sulfur saturation is reached. Therefore, reducing the flow rate may be an effective way of alleviating the effect of sulfur plugging in certain cases.

The effect of well spacing on sulfur buildup at the well face is illustrated in Figure 5. As demonstrated by curve 1 of this figure, the porous space in the vicinity of a well face may be completely saturated with deposited sulfur before the formation is depleted to the specified minimum well pressure. Therefore, periodic treatment of the formation near the well for sulfur removal may be essential. On

---

*Deposited as document 8981 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm. microfilm.

the other hand, formation cleaning may not be necessary for the entire production life if a close well spacing can be chosen. In the range of well spacing shown by curve 2, the pressure may decline to the specified minimum value at the well face; yet at the time when the porous space at this point is filled up with precipitated sulfur, the pressure at the outer boundary is still at a higher value. Curve 3 of Figure 5 represents a case where formation cleaning may not be necessary for the entire production life. At smaller values of the ratio r_e/r_w the pressure gradient at the well face is not so steep as it is at greater values. Since smaller pressure gradients result in less sulfur deposition, less sulfur ultimately accumulates at the well face at low r_e/r_w ratios relative to that at higher ratios, and the sulfur deposition throughout the reservoir is more uniform.

From this discussion it is apparent that plugging in porous media due to solid precipitation can be reduced through proper design of the production scheme. This can be accomplished either by reducing the flow rate or by choosing closer well spacing. However, the former method increases the time required to achieve a certain cumulative production. The latter method requires the drilling of more wells for a given sectional area. Thus, how much of the reservoir fluid can be produced economically will be determined by the parameters for any given case.

In the present paper the precipitation and separation of a solid substance from a flowing fluid in a porous medium have been illustrated for the hydrogen sulfide–sulfur system. However, the method developed can be extended readily to other systems.

CONCLUSIONS

The following conclusions can be drawn from this study:

1. A mathematical model has been formulated to describe fluid flow accompanied by solid precipitation in a porous medium. A numerical method has been developed to predict the pressure and saturation distributions in a porous formation as functions of time and radial distance during depletion of the formation contents through a single well.
2. When applied to a reservoir containing mainly hydrogen sulfide saturated with elemental sulfur, the method predicts a rapid buildup of solid sulfur in the vicinity of the well. Only a negligible amount of solid is deposited close to the impermeable outer boundary of the reservoir.
3. The tendency toward plugging by deposited sulfur at the well face can be reduced either by reducing the production rate or by choosing closer well spacing.

ACKNOWLEDGMENT

This research was carried out under the supervision of Dr. C. S. Matthews. Many of the mathematical results presented in this paper were originated by Dr. J. W. Killian of Shell Development Company. Discussions with Dr. H. A. Deans of Rice University and D. F. Hughes of Shell Canada, Ltd., are appreciated.

NOTATION

a = function defined in Equation (11)
b = function defined in Equation (12)
B = boundary of space domain D
c = function defined in Equation (13)
D = space domain of problem, shown in Figure 1 for a reservoir
F = function of u and τ as defined in Equation (A-1)
h = thickness of a formation, ft.
i = integer
I = integrals defined in Equations (A-10) and (A-11)
j = integer
k = effective permeability to flowing fluid fraction of K, darcies
K = total permeability, darcies
l = integer
m = integer
M = dimensionless fluid mobility, $(k/K)/(\mu/\mu_i)$
p = pressure, lb./sq. in.
p_i = initial pressure, lb./sq. in.
P = dimensionless pressure, p/p_i
P_w = dimensionless minimum pressure at well face
q = dimensionless pressure gradient at well face, $(\partial P/\partial u)_{1,l+1/2}$
Q = mass flow rate, lb./hr.
Q_D = dimensionless mass flow rate at well face
r = radial distance, ft.
r_e = outer drainage radius, ft.
r_w = effective well radius, ft.
S = fluid saturation, fraction of pore volume
t = real time, hr.
T = time part of weight function φ
u = dimensionless independent variable, ln (r/r_e)
v = fluid velocity, ft./hr.

Greek Letters

α = functions defined in Equations (A-12) to (A-14)
β = functions defined in Equations (A-12) to (A-14)
γ = functions defined in Equations (A-12) to (A-14)
ϵ = arbitrary constant
η = functions defined in Equations (A-12) to (A-14)
μ = fluid viscosity, centipoise
μ_i = initial fluid viscosity, centipoise
ξ = space part of weight function φ
ρ = density, lb./cu. ft.
ρ_f = density of fluid, lb./cu. ft.
ρ_f^f = mass of fluid component per unit volume of solution, lb./cu. ft.
ρ_f^s = mass of fluid component per unit volume of solid phase, lb./cu. ft.
ρ_f^* = fluid density at standard condition, lb./cu. ft.
ρ_s^f = mass of solid component per unit volume of solution, lb./cu. ft.
ρ_s^s = mass of solid component per unit volume of solid phase, lb./cu. ft.
τ = dimensionless time
τ_m = dimensionless time for constant rate production period
ϕ = porosity, fraction
φ = weight function
ω = functions defined in Equations (A-12) to (A-14)

LITERATURE CITED

1. Archie, G. E., *Bull. Am. Assoc. Petrol. Geologists*, **34**, 943 (1950).
2. Bruce, G. H., D. W. Peaceman, H. H. Rachford, and J. D. Rice, *Trans. Am. Inst. Mining Met. Engrs.*, **198**, 79 (1953).
3. Kennedy, H. T., and D. R. Wieland, *ibid.*, **219**, 166 (1960).
4. Kniazeff, V. J., and S. A. Naville, *Soc. Petrol. Eng. J.*, **37** (March, 1965).
5. Lapidus, Leon, "Digital Computation for Chemical Engineers," McGraw-Hill, New York (1962).
6. Muskat, M., "Physical Principles of Oil Production," McGraw-Hill, New York (1949).
7. *Oil Gas J.*, 115 (March 15, 1965).
8. Reamer, H. H., B. H. Sage, and W. N. Lacey, *Ind. Eng. Chem.*, **42**, 140 (1950).
9. Synge, J. L., "The Hypercircle in Mathematical Physics," Cambridge Univ. Press, New York (1957).
10. West, W. J., W. W. Garvin, and J. W. Sheldon, *Trans. Am. Inst. Mining Met. Engrs.*, **201**, 217 (1954).

Manuscript received December 9, 1965; revision received April 4, 1966; paper accepted April 5, 1966. Paper presented at A.I.Ch.E. Dallas meeting.