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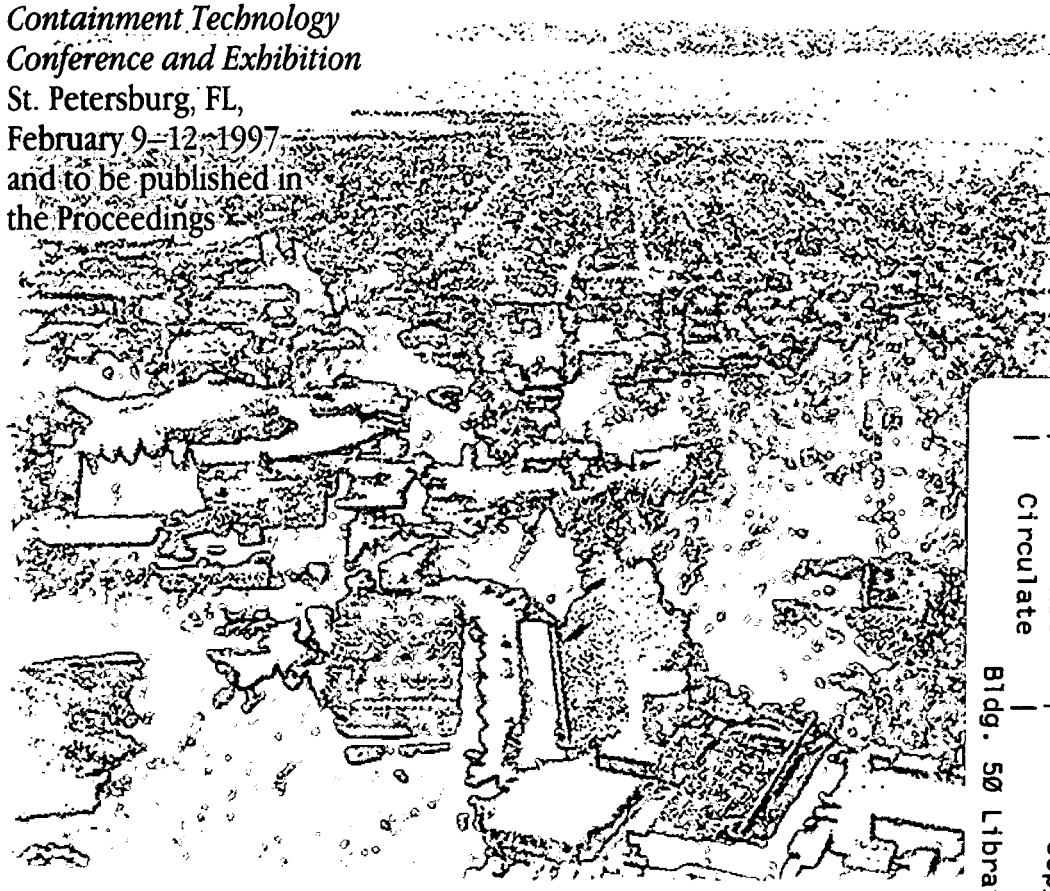

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ABSTRACT

The injection of solution grouts into the subsurface can be used to form underground barriers for the containment of contaminants. The technology requires identifying suitable grout materials, specifically fluids which exhibit a large increase in viscosity after injection and eventually solidify after a controllable period, thus sealing permeable zones. We have developed a new fluid property module for the reservoir simulator TOUGH2 to model grout injection, taking into account the increase of liquid viscosity as a function of time and gel concentration. We have also incorporated into the simulator a model which calculates soil hydraulic properties after solidification of the gel within the pore space. The new fluid property module has been used to design and analyze laboratory experiments and field pilot tests in saturated and unsaturated formations under a variety of subsurface conditions. These applications include modeling barrier emplacement in highly heterogeneous soils in the vadose zone, grout injection into the saturated zone in combination with extraction wells for flow control, the design of verification strategies, and the analysis of barrier performance. In this paper we discuss the modeling approach and present simulation results of multiple grout injections into a heterogeneous, unsaturated formation.

1. INTRODUCTION

The injection of solution grouts into the subsurface is a technique used to encapsulate pollutants, to emplace underground barriers for containment of contaminant, and to prevent the spread of existing plumes. Furthermore, on-site containment and control of ground-water flow patterns can prevent off-site migration during active cleanup operations, and may increase the efficiency of traditional remediation techniques. A number of barrier fluids have been identified as suitable for permeation grouting of soils in both the saturated and unsaturated zone. These fluids exhibit a low initial viscosity which increases at a controllable rate after injection. The grout eventually solidifies, sealing the permeable zones of an aquifer. Work on the identification of appropriate barrier fluids and the generation of basic knowledge on their rheological properties is reported in Moridis et al. (1994). We will focus here on an aqueous silicon-based chemical grout, the gelation of which is induced and controlled by increasing the ionic strength of the gelling agent.

Numerical models are essential tools for the design of subsurface barrier systems, the optimization of the emplacement strategy, and the assessment of the barrier's integrity. Computer programs designed to simulate in-situ gelation for the application of enhanced oil recovery have been previously presented (Scott et al., 1985; Hortes, 1986; Todd, 1990). The main emphasis in these works is on the kinetics of the gelation process. We are interested in the performance of shallow barriers emplaced in both the saturated and the vadose zone. Each application poses its specific problems. In the saturated zone, the dilution of the solution grout by native water has to be considered. Emplacement issues such as the absence of strong gravity effects and the question of how to achieve complete volumetric coverage between injection ports and near plume boundaries require special attention (Moridis et al., 1996). Due to space limitation, we focus here on vadose zone applications. When grout is injected into unsaturated soils, the grout plume slumps under gravity and spreads due to capillary forces, leaving the soil only partially saturated. The reduction of porosity and permeability is thus a function of final gel saturation, and multiple injections may be required to achieve complete filling of the pore space. Modeling these processes includes the simulation of multiphase flow effects, the concentration and time-dependent increase of viscosity of the grout-water mixture, and a model description of the hydraulic properties of the partially grouted soil. For the purpose of this work, we have extended the capabilities of the TOUGH2 code (Pruess, 1987; 1991), a numerical simulator for non-isothermal flows of mul-

ticomponent, multiphase fluids in porous and fractured media. In this paper we describe the physical processes considered and the modeling approach used for the development of the numerical simulator, and discuss its application for the design of a subsurface barrier system.

2. MODELING APPROACH

2.1 Process Description

Injection of a water-based grout into an unsaturated porous medium leads to a system which consists of three separate phases, namely the solid grains, a non-condensable gas, and an aqueous phase containing chemical grout in dissolved or colloidal form. After the initiation of the gelling process, the viscosity increases, turning the gel-water mixture into a non-Newtonian, visco-elastic fluid that eventually solidifies. The appearance of a new phase, i.e. the solidified gel, leads to changes in the physical and chemical properties. Contact angles and interfacial tensions vary with the chemical properties of the gel-water mixture, and adsorption and filtration of gel clusters may occur. By the time the grout is completely gelled, the resulting "new" porous medium has a lower porosity, a new pore structure, reduced permeability, and different wettability characteristics.

Controlled barrier emplacement becomes very difficult if the gelation kinetics are significantly affected by the soil mineralogy or pore water chemistry. Detailed knowledge about chemical and hydraulic heterogeneity over a range of scales would then be required to predict the behavior of the gel. A robust technology therefore relies on a solution grout material that is minimally affected by pH, salinity, and activity of the soil and pore water. In this case, no need for chemical interaction modeling arises. We use surface-modified colloidal silica grouts especially manufactured for this type of application. In our modeling approach, the following major assumptions are made:

(i) The chemical process of gelation is not explicitly modeled. Instead, we calculate the viscosity of the liquid phase as a function of grout concentration and time. The viscosity of pure grout as a function of time is measured in the laboratory and represented by a *gel time curve*. Mixture viscosity varies with the concentration of gel in the aqueous phase, and is described by a *mixing rule*.

(ii) The grout is treated as a miscible, aqueous solution, i.e. it does not form a separate phase. After completion of the gelling process, we assume that the gel, which is a fluid of very high viscosity, solidifies. By doing so, the porosity of the grouted soil is reduced. The new porous medium thus has a lower permeability and different characteristic curves in the region affected by the grout. The transition of the grout from a highly viscous fluid to a solid part of the matrix is described by the *solidification model*.

(iii) Injection and redistribution of grout in the unsaturated zone is modeled using TOUGH2 (Pruess 1987; 1991). We consider multiphase flow of the three components water, air, and chemical grout. Flow processes account for relative permeability and capillary pressure effects, and allowance is made for appearance and disappearance of phases and components. The empirical gel time curve and mixing rule are used to describe liquid phase viscosity. The solidification model describes the transformation of viscous gel into a "new" porous medium.

The conceptual approach as well as the specific quantitative correlations described in the following sections are in need of detailed testing and verification through laboratory and field experimentation.

2.2 Gel Time Curve and Mixing Rule

Within the unsaturated zone, the pore space is occupied by two fluids: the gaseous phase, consisting of air and water vapor, and the liquid phase which is composed of water, grout, and dissolved air. The viscosity of the liquid phase depends on grout concentration and time. The increase of pure grout viscosity as a function of time is described by the gel time curve, a parameterized function which can be fitted to laboratory data. Based on the measurements of Moridis et al. (1994) we suggest the use of an exponential function of the form

Gel Time Curve: $$\mu_{gel} = a_1 + a_2 \cdot \exp(a_3 \cdot t) \quad (1)$$

where t is time, and a_1, a_2, and a_3 are fitting parameters. After injection, the grout suspension will be diluted due to mixing with pore water. The mixing rule calculates the viscosity of the liquid phase, μ_l, as a function of gel concentration, X_l^{gel}, and time. One of the following mixing rules can be used:

Linear Mixing Rule: $$\mu_l = X_l^{gel} \cdot \mu_{gel} + \left(1 - X_l^{gel}\right) \cdot \mu_w \quad (2)$$

Power-Law Mixing Rule: $$\mu_l = \left(\frac{X_l^{gel}}{\mu_{gel}^{\ b}} + \frac{1 - X_l^{gel}}{\mu_w^{\ b}} \right)^{-1/b} \quad (3)$$

Figure 1 shows the viscosity of the liquid phase as a function of time and gel concentration. The upper part of Figure 1 shows the viscosity measurements for a colloidal silica gel (symbols). The solid line is the fitted gel time curve (Eq. 1) which provides the viscosity as a function of time for pure gel (X_l^{gel}=1). A linear mixing rule (Eq. 2) has been applied and is visualized in the lower part of the figure where viscosity (in centipoise) of the liquid phase is contoured as a function of gel concentration and time. Strictly speaking, the concepts of a gel time curve and mixing rule are only meaningful as long as the grout is completely miscible with water, and as long as the grout-water mixture is Newtonian. For practical purposes, however, this model is reasonable since grout plume movements are very slow once a high viscosity has been reached.

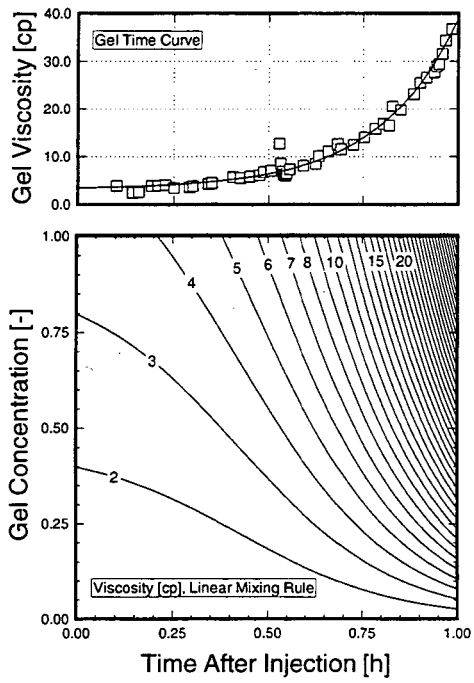

Figure 1. Viscosity in centipoise of pure gel as a function of time (gel time curve) and gel concentration (mixing rule).

2.3 Solidification Model

As discussed above, the gel is modeled as a liquid, the viscosity of which increases with time as gelation proceeds, until solidification occurs. After complete solidification, the soil exhibits new properties that must be derived based on assumptions about the pore structure of the partially

grouted soil. The parameters to be recalculated are porosity, permeability, relative permeability and capillary pressure functions, and initial liquid saturation. All these properties are a function of the final grout content prior to solidification.

The Solidification Model is based on the assumption that all of the liquid in the pore space eventually solidifies if the grout in the liquid phase exceeds a certain minimum concentration, $X_{\min}^{gel}$. We introduce a parameter A as follows:

$$A = 1 \qquad \text{for} \quad X_l^{gel} \geq X_{\min}^{gel} \tag{4a}$$

$$A = \frac{X_l^{gel}}{X_{\min}^{gel}} \qquad \text{for} \quad X_l^{gel} < X_{\min}^{gel} \tag{4b}$$

Setting $X_{\min}^{gel}$ to 0.2, for example, means that all the liquid with a gel concentration greater than 0.2 eventually solidifies. The fluid with lower gel concentrations solidifies incompletely.

The liquid saturation at the time solidification occurs is denoted by S_l^{sol}. All soil characteristics and initial conditions referring to the new porous medium are denoted by a star (*). The porosity of the grouted medium is reduced by the amount of gel that solidifies:

$$\phi^* = \phi(1 - A \cdot S_l^{sol}) \tag{5}$$

The porosity reduction leads to a decrease of absolute permeability. The partial clogging of the pore space by grout is conceptually similar to the permeability reduction due to phase interferences in a multiphase flow system. The Permeability Reduction Model (PRM) describes the permeability reduction as a function of the solidified grout saturation:

$$k^* = k \cdot \text{PRM} \tag{6}$$

Because the grout-water mixture is the wetting fluid, the permeability reduction factor (PRM) is described by the relative permeability function of the non-wetting phase, $k_{r,nw}(S_{nw})$, evaluated at $S_{nw} = (1 - A \cdot S_l^{sol})$. The permeability reduction might in fact be stronger because not only are the small pores sealed by the wetting grout, but continuous gel adsorption at the pore walls reduces the diameter of the remaining larger pores. Other permeability reduction models can be found in Verma and Pruess (1988), Todd (1990), and Oldenburg and Spera (1992).

Due to the reduced pore sizes, the capillary pressure $p_c{}^*$ of the grouted soil is expected to be more negative for a given water content. We apply Leverett's scaling rule to calculate the capillary pressure of the new medium based on the capillary pressure function of the original soil:

$$p_c{}^*(S_l{}^*) = p_c(S_l^{ori})\sqrt{\frac{k}{k^*} \cdot \frac{\phi^*}{\phi}} \tag{7}$$

Note that the original p_c -function is evaluated at a saturation related to the original porosity value:

$$S_l^{ori} = A \cdot S_l^{sol} + S_l{}^* \cdot \left(\frac{\phi^*}{\phi}\right) \tag{8}$$

Given a certain liquid saturation $S_l{}^*$ of the grouted soil, Eq. (8) provides the original liquid saturation S_l^{ori} corresponding to the same water content. The capillary pressure p_c is then obtained and rescaled by applying Leverett's model (Eq. 7). Finally, the liquid saturation after solidification is calculated as the volume of the ungelled pore fluid divided by the porosity of the new medium.

Note that the solidification model has to be applied to each grid block of the discretized flow region to provide initial conditions and soil properties of the grouted medium for subsequent simulations.

3. APPLICATION

Numerical simulations were performed to design a horizontal grout barrier beneath a potentially leaking underground storage tank. A two-dimensional vertical model was developed, and a heterogeneous, anisotropic permeability field was generated with a geometric mean permeability of 1.55×10^{-12} m^2 and a standard deviation of one order of magnitude. We modeled multiple grout injections from two layers of horizontally drilled boreholes; the spacing between the wells is 1.0 m. The layout of the wells is shown in Figure 2. The first grout injection is made through the lower array of boreholes at a rate of 12 kg per minute and per meter borehole for an injection period of 1 hour. A gel time curve has been selected such that gel viscosity is doubled after 2 hours, and that it solidifies after 6 hours.

Figure 2 shows contours of grout content , i.e., the product of liquid saturation, grout concentration, and porosity, at the end of the first injection. Highest grout contents are in the immediate vicinity of the boreholes, where the initial soil gas and pore water are completely displaced due to the injection overpressure. After redistribution of the plume due to gravity and capillary forces, however, maximum grout contents are encountered beneath the injection ports due to gravitational slumping of the plume. The relatively fast gelation prevents the plume from slumping down even further. Note that the spreading of the plume leads to an incomplete occupation of the pore space by grout, i.e., partial plugging and insufficient permeability reduction will occur if only one injection is performed.

After solidification of the gel, the porosity of the grouted region is reduced, leading to the permeability field shown in Figure 3. Subsequently, a secondary injection is performed from the upper set of boreholes for 30 minutes. The grout from the secondary injection ponds on the low permeability layer produced by the primary injection (Figure 3), assuring high final grout saturations and thus enhancing the continuity of the subsurface barrier. The permeability field after solidification of the second plume is shown in Figure 4, along with the concentration contours of a potential spill from the underground storage tank. The plume is effectively contained by the barrier.

4. CONCLUDING REMARKS

Emplacing a subsurface barrier for temporary or permanent containment of contaminants requires identification of suitable barrier liquids, the development of an emplacement technology, as well as a numerical model for performing design calculations. In this paper we have presented a numerical simulator for modeling multiple injections of chemical grouts into the saturated and unsaturated zones of a heterogeneous aquifer. Assuming that the gelation process is not affected by soil and pore water chemistry, the code calculates the increase of viscosity as a function of time and gel concentration. A solidification model has been developed which assigns new properties to the zone partially occupied by grout. Application of the solidification model enables the repeated simulation of multiple grout injections, and performance studies of the barrier system can be conducted. Modeling capabilities are also essential for the verification studies including hydraulic, pneumatic, and tracer testing to assess permeability reduction and the continuity of the barrier. Furthermore, the design of a monitoring system can be supported by modeling.

Model calculations have been performed to design a horizontal barrier system in the unsaturated zone of a highly heterogeneous formation. Two layers of grout have been injected from a series of horizontally drilled wells. The simulations demonstrate that the combined action of gravity and capillarity leads to a substantial spreading of the gel plume, resulting in incomplete filling of the pore space. The second injection, however, assures high final grout contents due to ponding of the gel plume on the first layer of solidified grout.

We believe that uncertainties in formation characteristics and gel behavior should be addressed by appropriate grout specification and a robust design of the containment system. Nevertheless, accurate modeling of multiphase flow effects is crucial for the simulation of barrier emplacement in the unsaturated zone. The extended TOUGH2 code is considered to be a useful tool for the study of the behavior of grout plumes and for the design of subsurface barrier systems.

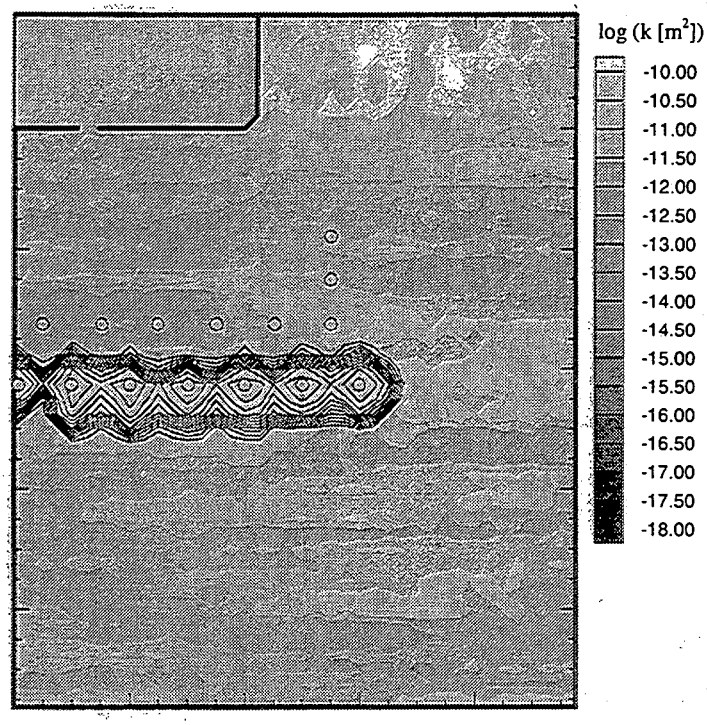

Figure 2. Initial permeability field (shaded) and gel content (contour interval is 0.04) after 30 minutes of grout injection.

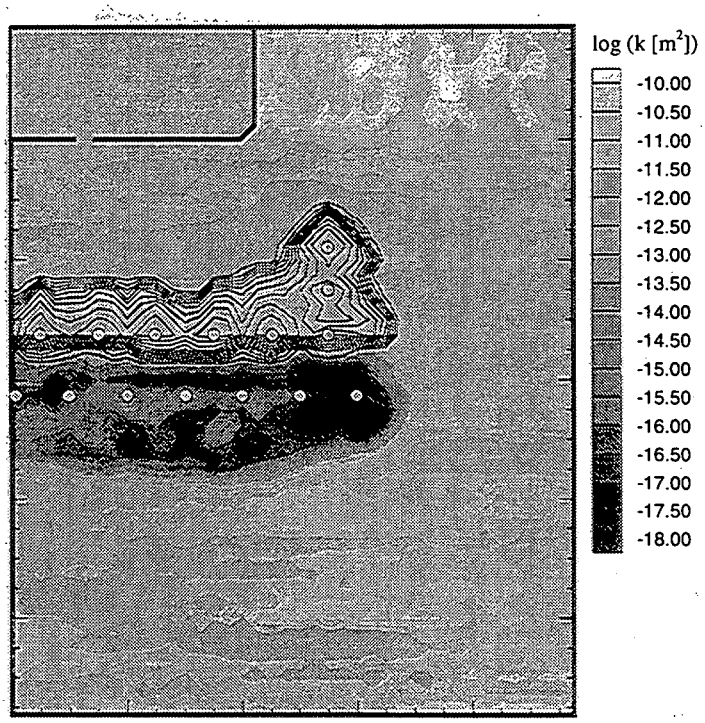

Figure 3. Permeability field (shaded) and gel content (contour interval is 0.04) prior to solidification of secondary grout plume.

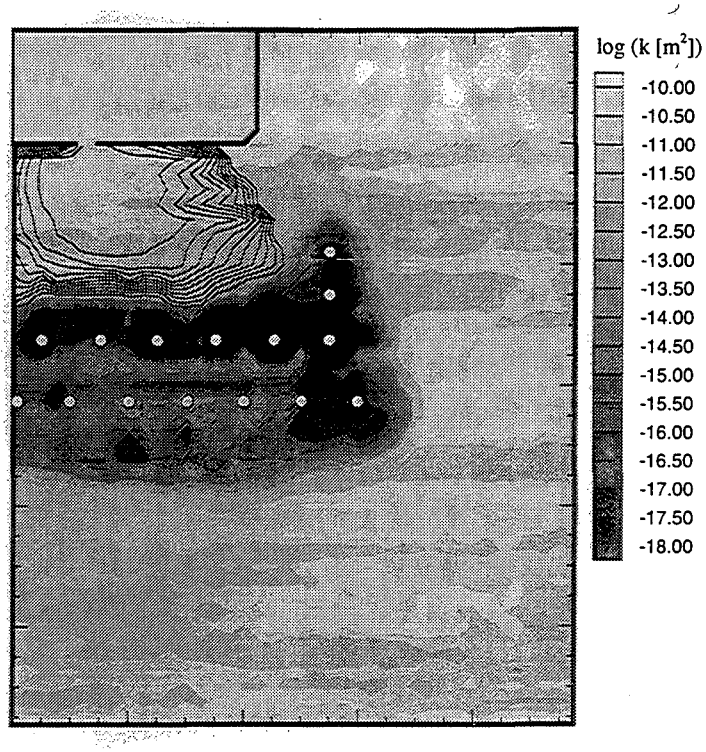

Figure 4. Final permeability field and containment of contaminant by subsurface barrier.

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