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Project Summary

Physics of Immiscible Flow in Porous Media

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This project addresses the conceptual formulation, numerical implementation and experimental validation of a model for the movement of organic chemicals which are introduced into soils as nonaqueous phase liquids via surface spills or leakage from subsurface containment facilities. Numerical procedures were investigated and implemented for solution of the governing equations for flow in three phase systems under the assumption of constant gas phase pressure, coupled with the equations for three phase component transport with equilibrium phase partitioning. A two dimensional finite element code was developed which was used to evaluate a proposed constitutive model for properties governing multiphase flow. The code was also applied to investigate various hypothetical problems involving leakage from underground storage facilities.

Continuum-based mathematical models for fluid flow in porous media require knowledge of relationships between fluid pressures (P), saturations (S) and permeabilities (k) for the fluids and porous media of concern to enable prediction of convective velocities and saturations. A concise parametric description for S-P relations based on a scaling procedure formulated to separate porous medium-dependent and fluid-dependent effects has been developed. Relative permeability-saturation relations are predicted from the scaled S-P functional, which is assumed to reflect the pore size distribution of the medium. The general form of the model provides a unified theoretical framework for the description of three phase k-S-P relations subject to arbitrary saturation paths, including effects of hysteresis and nonwetting fluid entrapment. Model calibration is predicated on the availability of two phase saturation-pressure measurements alone. Data requirements may be further reduced by employing interfacial tension data to determine scaling coefficients.

Experimental studies were carried out to validate the constitutive model for cases involving monotonically draining water and total liquid saturation paths. Procedures were developed for the direct measurement of fluid pressures and saturations in three phase systems. Measurements of S-P relations for static two and three phase systems provided model validation and demonstrated the feasibility of using interfacial tension data to simplify model calibration. Model validation under transient flow conditions was investigated by comparison of numerically simulated and experimentally measured results. Two phase investigations were conducted involving measurement of water displacement during constant pressure NAPL infiltration and redistribution event. These validation studies indicated the k-S-P model provides a reasonable description of the system behavior under the imposed experimental conditions and may be satisfactorily calibrated using relatively simple procedures.

This Project Summary was developed by EPA's Robert S. Kerr Environmental Research Laboratory,

Ada, OK, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Contamination of groundwater by organic chemicals has become a serious threat to subsurface water resources. Among the most widespread and hazardous contaminants are organic liquids of low water solubility introduced into the subsurface environment via surface spills, leaks from underground storage facilities, or seepage from improperly designed or managed landfills or land disposal operations. Numerous contamination incidents have been documented resulting from petroleum sources. Such contamination problems generally involve complex mixtures of multiple organic constituents which may move in aqueous and nonaqueous liquid phases and in the gas phase and which may undergo chemically and biologically mediated degradation with time. Modeling of these systems requires consideration of multiphase fluid flow and multicomponent transport and reaction within each phase. The development of efficient numerical algorithms for such problems presents many difficulties. An even more fundamental impediment, however, has been the substantial lack of information concerning constitutive relationships governing multiphase flow and transport.

These studies focused attention on the characterization of properties governing multiphase flow with the following specific goals:

- develop a parametric model for hysteretic permeability-saturation-pressure relationships in three phase porous media systems,
- develop experimental methods and specialized apparatii to directly measure fluid pressures and saturations in three phase systems for purposes of model validation and refinement,
- develop experimental and computational procedures for routine model calibration from two phase system measurements,
- implement a two-dimensional finite element model for liquid flow and single component transport in a three phase system at constant gas pressure and experimentally validate flow model for selected saturation paths.

To model the transport of organic contaminants introduced into the subsurface as nonaqueous phase liquids (NAPLs), it is first necessary to accurately describe the convective movement of coexisting fluid phases, i.e., water, air and NAPL. Continuum-based mathematical models for fluid flow in porous media require knowledge of relationships between fluid pressures (P), saturations (S) and permeabilities (k) for the fluids and porous media of concern to enable prediction of convective velocities and saturations. Unfortunately, such relationships are quite complex in three phase (air-NAPL-water) systems and very difficult to measure directly. Therefore, much effort was addressed to development of methods to describe k-S-P relations with maximum accuracy, while keeping procedures for model calibration to a minimum to facilitate practical application to real problems given reasonable resources. The approach to this problem was to develop a concise parametric description for S-P relations based on a scaling procedure formulated so as to separate porous medium-dependent and fluid-dependent effects. Relative permeability-saturation relations are predicted from the scaled function which is regarded as an index of the pore size distribution of the medium. The formulation permits calibration to be performed by measuring only two phase system S-P relations. Furthermore, it was found that scaling coefficients may be estimated with good accuracy from fluid interfacial tension data, thus enabling model calibration from only air-water S-P data and interfacial measurements. Validation of such calibration procedures is demonstrated for static two phase systems and for transient and three phase systems.

A critical assumption in the k-S-P model is that two phase S-P data can be used to predict three phase S-P relations. Although such assumptions have been commonly invoked in the field of petroleum engineering for many years, no direct validation has been reported due to a lack of experimental procedures for measuring three phase S-P relations. The investigators, therefore, developed a new experimental device expressly for the purpose of measuring three phase S-P relations and have utilized this apparatus to test the three phase S-P submodel for cases of monotonically draining water and total liquid saturation paths.

In lieu of evaluating the k-S submodel by direct measurement of three phase k-S-P relations, which is a v tedious and arduous task, t investigators validated the model comparing results of transient fl experiments with numerical simulatic based on the parametric mod Numerical procedures were investiga and implemented for the solution of governing equations for flow in two a three phase systems under t assumption of constant gas pha pressure in two dimensional spat domains using finite element methc which have been verified numerically a employed to investigate a number hypothetical problems. Experimen validation of the multiphase flow mo was achieved first for transient two pha experiments involving measurement water displacement by TCE and benzene-derivative hydrocarbon.

In order to validate the k-S-P mo for transient flow conditions in thr phase systems, procedures had to developed for the measurement of fl saturations and pressures in three pha porous media systems. To accompli this, a dual energy gamma attenuati device was designed, built and install in a laboratory dedicated to this functic The apparatus enables simultaneo determinations of air, water and NA saturations to be made with spat resolution of less than 1 mm on s columns or flumes. Soil columns we designed and constructed for use in t dual energy gamma system and we equipped with specially fabricat sensors capable of measuring NAPL a water phase pressures concurrent Instruments for measuring NAI pressures were adopted from the thr phase S-P apparatus design utilizing special technique for producing stak hydrophobic sensors by silanization ceramic tensiometers Experiments we performed involving simultaneo measurement of liquid pressures a saturations during transient flow in thr phase air-NAPL-water systems whi provided successful validation of tl three phase k-S-P model and h demonstrated the feasibility of employi model parameters calibrated using or two phase air-water S-P relations a interfacial tension data

The studies mentioned to this po have all been constrained to cases which hysteresis and nonwetting flu entrapment is ignored or eliminat experimentally by considering on situations involving monotonic water a total liquid drainage. In field situatior effects of hysteresis and especially flu entrapment may be expected to ha significant effects on flow and mass transport. The volume that a NAPL plume will eventually occupy before becoming effectively immobilized due to saturation path reversals will exert a major influence on the long term behavior of aqueous and gaseous phase transport from a contamination event, and this behavior cannot be modeled without explicitly considering entrapment effects on the k-S-P relations. Previous analyses of hysteresis in three phase systems have been incapable of dealing with arbitrary saturation paths and have generally considered hysteresis in k-S relations only.

To deal with the problem of hysteresis and fluid entrapment,the investigators developed a unified theoretical framework for the description of three phase k-S-P relations subject to arbitrary saturation paths. The theory extends their previous scaled parametric model, which it includes as a special case. Model calibration is still predicated on the availability of two phase data alone and requires only slightly more information than the nonhysteretic model. The investigators experimentally investigated calibration procedures for the hysteretic S-P submodel which also provided model validation for two phase main drainage and primary imbibition paths.

Numerical studies were initiated in the final stages of the project dealing with the analysis of coupled three phase flow and single component transport controlled by local phase equilibrium. The mathematical formulation for the problem was derived and implemented numerically for a two dimensional spatial domain.

Discussions and Conclusions

Model for Nonhysteretic Multiphase Flow

The investigators have presented a parametric model for relative permeability saturation-pressure relationships in porous media which contain up to three coexisting fluid phases following monotonic saturation paths. The model provides simple closed form expressions for fluid saturation-pressure functions and their derivatives and for relative permeability-saturation functions.

By interpreting the scaled saturation-capillary head function as an index of the pore size distribution of the medium, expressions for the relative permeabilities of air, water, and NAPL in a three phase system are derived which also degenerate to appropriate functions for any subset two phase case. No additional parameters are introduced over those involved in describing the saturation-capillary head with the exception of a gas slippage correction factor for gas permeabilities. In fact, several parameters in the saturation-capillary pressure function do not arise in the permeability-saturation functions. Direct verification of the relative permeability model will require comparison of model-predicted permeabilities with experimentally observed values on two phase and three phase systems

The proposed parametric model for constitutive relationships governing multiphase convection is expected to provide an adequate representation of fluid-porous media systems subject to their meeting certain criteria implied in deriving the model. One difficulty to contend with is the assumption of a rigid porous medium with no significant fluid-solid phase interactions The data shown for a clayey soil, albeit of relatively low swelling potential, indicates that the proposed scaling procedure may not deteriorate seriously in fine-grained materials. However, even rather small changes in the pore size distribution, which in certain instances may not be clearly evident in the saturation-pressure relations (e g , development of small fissures due to clay shrinkage in the presence of low dielectic organic fluids), may markedly affect the permeability functions. In some instances such problems may be a substantial concern and research is warranted to investigate methods of dealing with their effects quantitatively

Another proviso on the model which should be emphasized is that the investigators have assumed saturation paths corresponding to monotonically decreasing wetting phase saturations in all instances, and for three phase flow that total liquid saturation also monotonically decreases. When these conditions are not met, hysteresis in the constitutive properties is expected to be of importance, particularly when imbibing and draining saturation-capillary head curves do not close at zero capillary head. For example, following an initial injection of oil into a water-saturated core, reflooding with water will commonly fail to achieve full water saturation due to entrapment of some oil.

Measurement and Estimation of Static Two Phase Saturation-Pressure Relations

The format proposed to scale saturation-capillary pressure relations of two phase air-water, air-organic and organic-water porous media systems was evaluated by applying the procedure to relations measured for four organic liquid systems in a sandy porous medium. Multiphase versions of the Brooks-Corey and van Genuchten retention functions were fit to the experimental data using nonlinear regression analyses

A procedure was outlined to obtain parameters of retention functions which are used to predict saturation-pressure relations in three phase systems that preserves a unique pore size distribution for rigid porous media. In addition, it was shown that fluid-dependent scaling factors used to scale saturation-capillary pressure relations of different two phase systems in the sandy porous medium investigated are well approximated from ratios of interfacial tensions provided the latter are measured using fluids subject to any impurities occurring in the actual porous medium system. Using scaling factors from interfacial tension data permits prediction of saturation-capillary pressure relations for any two phase fluid system in a porous medium from direct measurements of single two phase system and appropriate interfacial tension data Three phase system behavior may likewise be predicted

Measurement and Estimation of Three Phase Saturation-Pressure Relations

An experimental apparatus was developed to measure three phase air-oil-water S-h relations in unconsolidated porous media The apparatus is also capable of measuring S-h relations of two phase air-water and air-oil systems Measurements of three phase S-h relations were conducted and compared to two phase S-h measurements for three porous media to directly test assumptions involved in the prediction of three phase S-h relations from two phase measurement Good agreement between total liquid saturations in three phase air-oil-water systems and oil saturations of two phase air-oil systems as functions of air-oil water systems and oil saturations of two phase air-oil systems as functions of air-oil capillary head was observed Close agreement was also observed between water saturation versus oil-water capillary head relations measured in two and three phase systems.

Agreement between two and three phase S-h relationships for monotonic saturation paths imply that researchers modeling flow of continuous oil and water phases in the vadose zone may employ more readily available two phase S-h measurements to predict flow phenomena in three phase air-oil-water systems if fluid saturations are assumed to be unique functions of capillary head (i.e., no hysteresis).

In addition to the proviso that results have only been presented for monotonically draining water and total liquid saturation paths, the procedures for predicting three phase S-h relations are expected to be valid only for strongly water-wet and rigid porous media. Soils and aquifer materials are generally expected to meet the criterion of being strongly water-wet, and at least in relatively coarse grained materials the rigid medium constraint may be sufficiently closely met as well. In finer textured porous media, the rigid medium assumption is most prone to cause deviations from theory and more extensive investigations for materials with a wide compositional range is warranted in the future.

The scaling procedure that we have advanced combined with a suitable parametric representation of the scaled retention function enables development of a multiphase retention model which can be used to describe two and three phase S-h relations. The results indicate that calibration of the model by fitting to two phase air-water, air-oil and oil-water S-h data yield an accurate description of two and three phase drainage S-h relations. If two phase air-oil and oil-water are not available, model parameters can be estimated from two phase air-water S-h data and relatively simple measurements of air-water, air-oil and oil-water interfacial tensions with only moderate deterioration in model precision.

Finite Element Model for Multiphase Flow

A finite element formulation based on the variational approach has been successfully employed to solve the coupled immiscible flow problem involving a three fluid-phase system of air, water, and NAPL in soil with air at constant pressure. For two hypothetical fluid storage tank problems, the leakage of organic fluids was analyzed and the NAPL plume movement predicted. The results show the potential of applying the finite element method developed here to the design of monitoring systems and remediation schemes for leaking underground storage tanks.

Model Validation and Calibration for Two Phase Transient Flow

Three methods of calibrating the k-S-P model developed in previous chapters were investigated. (1) direct measurement of S-P relations for air-water, air-oil and oil-water systems, (2) direct measurement of air-water S-P relations with scaling coefficients estimated from interfacial tension data, and (3) numerical inversion of transient oil-water displacement experiments using a nonlinear optimization procedure. The methods were evaluated by analysis of results for two NAPL-porous media systems.

Of the methods investigated for calibrating the proposed multiphase k-S-P model, all have yielded reasonably good descriptions of both static S-P relations and of transient two phase displacement experiments, although the equilibrium-based procedures (Methods 1 and 2) naturally describe S-P data with greatest accuracy, while the transient inversion procedure (Method 3) predicts transient flow behavior more closely. The substantial agreement between various calibration methods not only facilitates the selection of calibration procedures to ease experimental effort involved, but provides validation of the form of the proposed k-S-P model for the conditions involved in the present study.

Model Validation and Calibration for Three Phase Transient Flow

A three phase air-oil-water flow experiment was designed and conducted involving simultaneous measurements of liquid saturations and pressures during monotonic water and total liquid drainage. Oil and water saturations were measured with a dual gamma radiation apparatus after doping the liquid phases to enhance separation of gamma attenuation coefficients. Oil and water pressures were measured with hydrophobic (treated) and hydrophilic (untreated) ceramic tensiometers, respectively.

Measured water saturation vs c water capillary head data during the thr phase flow experiment agreed well w predictions using the proposed k-S model calibrated from static air-wa S-P relations and interfacial tension d (Method 1). Measured total liqu saturation vs air-oil capillary head d deviated more severely from the a water data predictions and exhibit marked scatter, which was inferred reflect, in part, errors in experimen measurements or perhaps due deviations from the assumption constant gas phase pressure. Moc parameters were also evaluated directly fitting to the observed thr phase S-P data (Method 2).

Good agreement was found betwe measured liquid saturation distributions time and space during the transient fl experiment and those predicted by t finite element multiphase flow code usi the proposed k-S-P model calibrat using either Method 1 or Method Measured liquid pressures agreed mc closely with simulations employi Method 2 parameters. Neverthele considering the practical advantages using Method 1 for model calibration, t accuracy of the simulations based these estimates was relatively good.

Although the experimental regir considered in the present paper w limited to monotonically decreasing wa and total liquid saturation paths, t methodology is applicable to the study arbitrary saturation histories. Hystere and nonwetting fluid entrapme associated with complex saturation pat may substantially affect three phase S-P relations, and studies of the effec of such phenomenal will be necessary obtain confidence in our ability evaluate field scale multiphase flo behavior.

Model for Hysteretic Permeability-Saturation-Pressure Relations

A model is presented f permeability-saturation-pressu relationships in porous media syster containing up to three immiscible flu phases which accounts for hysteres and fluid entrapment effects. The moc was formulated by proposing a sing hysteretic scaled saturation-capilla head functional which describes wat saturation vs air-water capillary head two phase systems and water saturati vs. oil-water capillary head or total liqu saturation vs air-oil water capillary he in three phase systems and can employed to predict fluid permeabilities via a parametric model. The scaling of heads is obtained by a linear transformation and saturations are scaled by introducing the concept of apparent fluid saturations which include entrapped nonwetting fluid contents with those of the fluid under consideration.

By employing an adaptation of van Genuchten's retention function to characterize the pore size distribution of the system in conjunction with Mualem's relative permeability model, closed form expressions for relative permeabilities are obtained. Calculations for a hypothetical problem indicate that the proposed model predicts behavior that is in accordance with general behavior that has been reported in the literature for such systems. It may be noted that other parametric models could have been utilized to obtain similar ends, however,the evaluation of possible advantages in various various formations must be left to future investigations. Detailed measurements of three phase permeability-saturation-pressure relations will be necessary to rigorously validate the model.

Calibration of the proposed hysteretic k-S-h model was investigated for a sandy porous medium with p-cymene as the NAPL Two methods were studied· Method 1 in which model parameters were fit to a combined set of two phase air-water, air-oil and oil-water drainage and imbibition saturation-capillary head data, and Method 2 utilizing air-water drainage path data, single imbibition S-h points for air-water air-oil and oil-water systems and interfacial tension data. Predicted and observed drainage and imbibition data for all three of the two phase systems were described very closely by Method 1 parameters and only slightly less well by those using Method 2. This is a very encouraging result since the use of Method 2 greatly simplifies model calibration. The most tedious aspect of calibration in Method 2 and probably the most crucial in practice, is the determination of maximum residual nonwetting phase saturations. The development of simpler experimental procedures or empirical estimation methods for the latter would be of substantial value

Model parameters estimated by the two methods were employed to predict three phase k-S-h relations during a hypothetical NAPL contamination scenario Differences in paths predicted by the parameters obtained for the two calibration methods were generally small again providing encouragement for use of the simpler procedure. Fluid entrapment effects were provided to lead to greater hysteresis in three phase S-h relations than were evident in the two phase data. Effects of fluid entrapment on water relative permeability was not great for the saturation paths simulated, while effects on air relative permeability were large at high total liquid saturations.

The results indicate that hysteresis and nonwetting fluid entrapment effects in k-S-h relations of three phase systems may be quite substantial. However, since the proposed model is based on a number of hypotheses regarding the extension of two phase behavior to three phase systems, validation will require direct comparisons between model predictions and three phase system behavior.

Numerical Model for Multiphase Component Transport

A model is described for multiphase contaminant transport and its numerical implementation for a simple problem involving a single contaminant moving simultaneously in water, gas and nonaqueous liquid phases is demonstrated. A computational scheme involving decoupling of the solutions for flow and mass transport equations was implemented and found to provide satisfactory results. For future extensions to multicomponent transport, this procedure is expected to provide very substantial savings in computer execution costs. Much work remains to be done pertaining to the development of numerical solutions for multiphase transport to consider transport of complex NAPL mixtures, constituent interactions (e.g., cosolvent effects) and biochemical transformations, nonequilibrium phase partitioning, gas phase convection, hysteresis and other effects, as well as to improve and develop algorithms and computational procedures which lead to improved computational efficiency. Experimental studies should be pursued concurrently to further develop and refine constitutive models for multiphase flow and transport and to validate numerical codes which implement these.

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The complete report, entitled "Physics of Immiscible Flow in Porous Media," (Order No. PB 88-131 008/AS; Cost: $19.95) will be available only from:
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