

TRITIUM TRANSPORT THROUGH A LOW-PERMEABILITY NATURAL GAS RESERVOIR

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ABSTRACT

The U.S. Department of Energy and its predecessor agencies conducted a program in the 1960s and 1970s to evaluate technology for the nuclear stimulation of low-permeability gas reservoirs. Two of the sites were located in the Piceance Basin of Colorado, and the devices were detonated in low-permeability Cretaceous sandstones and shales. At each of the sites, transport of long-lived gas-phase radionuclides is of greatest concern -- these are primarily ³H, ¹⁴C, and ⁸⁵Kr. Using TOUGH2 and EOS7r, we conducted simulations to explore the controls and evaluate the length and time scales of ³H transport toward a nearby hypothetical producing gas well. The simulations were conducted such that diffusive transport occurred for a 35-year period starting from the nuclear detonation and prior to the start of gas production. The permeability in the vicinity of the hypothetical production well was then changed to account for hydraulic (production) fractures, and the simulator was restarted to evaluate 30 years of gas production from the well. Depending upon the location of the producing well and the magnitude of production, tritium transport could be controlled by either diffusion or advection associated with gas removal. After gas production ceased, the remnant pressure field controlled gas migration for a time equal to the amount of time that gas production occurred (i.e., an additional 30 yr). We found that in locations where diffusion was dominant, the mass fraction field of tritium was highly dependent upon the chosen tortuosity model.

INTRODUCTION

Several sites exist in the U.S. and former Soviet Union where nuclear devices were detonated in low-permeability gas reservoirs to increase reservoir permeability (Rubin et al., 1972; Nordyke, 1996). Three stimulation projects were undertaken in the U.S., although none resulted in successful gas production. Two gas reservoir stimulation projects were conducted in the former Soviet Union, at least one of which resulted in a 20-fold increase in produced gas over that expected without nuclear

stimulation. Nothing has been reported of the other experiment.

The three U.S. experiments were conducted in fine grained, low-permeability sandstones and shales located in the Piceance Basin of Colorado (Projects Rulison and Rio Blanco) and the San Juan Basin of New Mexico (Project Gasbuggy). Gaseous radionuclides (primarily ³H, ¹⁴C, and ⁸⁵Kr) were included with the natural gas flared to the atmosphere as a result of production testing after the detonations.

All three of the stimulation sites are located in actively producing gas fields. As a result, there is interest in evaluating subsurface radionuclide migration toward producing gas wells. This paper reports part of an investigation of subsurface transport at the Rio Blanco site, where three 33-kt nuclear explosives were simultaneously detonated at 1780, 1899, and 2039 m below the land surface. We discuss some of the processes (variable intrinsic permeability, initial gas saturation, choice of tortuosity model) controlling radionuclide migration toward a hypothetical well producing natural gas from the same horizon as the middle test shot. This paper is a condensation of a larger report (Cooper et al., 2005) addressing complexities such as parameter uncertainty more fully than is presented here.

GEOLOGIC SETTING

In theory, most of the dynamics of gas and liquid flow in petroleum reservoirs are understood. The development of an accurate model, however, can be hampered by a lack of data. In particular, local and regional pressure gradients are often not understood, as flow in petroleum reservoirs is generally controlled by gradients induced by production wells (Gerritsen and Durlowsky, 2005). In parts of the Piceance Basin of Colorado, gas production occurs from localized sandstone lenses in the low-permeability Mesaverde Formation. These lenses are not always associated with individual strata, due to uplift and tilting of rocks associated with the development of the Overthrust belt. Thrusting resulted in fragmented hydraulic gradients, and the

extremely low permeabilities helped preserve pre-Overthrust fluid pressures. The result is hydraulic gradients with no relation to the current geologic structure, and an absence of discrete gas/water contacts in the formation. Produceable gas is sometimes found structurally downdip from water-saturated formations, with no obvious trapping mechanism aside from the low permeability of the reservoir in which it exists (Johnson, 1989).

EXPLOSION PHENOMENOLOGY

Production tests in a well near the Rio Blanco nuclear emplacement hole indicated that the formation pressure prior to the detonations was 19.2 MPa. The formation pressure exceeded the lithostatic pressure within 10 seconds of the blast, causing fracturing in the Mesaverde Formation. The extreme temperatures from the blast vaporized much of the rock, water, and gas, resulting in three underground cavities. Each cavity was estimated to have a radius of 21 m and height of 84 m. Fractures from the explosions were estimated to extend 63 m horizontally from the emplacement hole. Within one minute of the detonation, the formation pressure fell to pre-detonation formation pressure. Molten rock formed a puddle of lava (puddle glass, or melt) several meters deep at the bottom of each cavity. In theory, when the gas pressure could no longer sustain the weight of the roof of the cavity, the roof should have collapsed into the cavity, forming a high-porosity rubble called a chimney. Within several days of the detonations, the three cavities cooled to below 600 K, and condensation of steam began. At this time, the rate of cooling of the cavity gas sharply decreased. Within one month, most of the steam condensed, causing the formation pressure to drop to 11.1 MPa. At this point in time, formation gas flowed into the cavity, resulting in a rise back to the pre-test formation pressure in the cavity/chimney.

As the cavity cools, radionuclides are distributed into four phases: the nuclear melt glass, as surface deposits on rubble in the chimney, dissolved in water, or in the gas phase (Borg et al., 1976; IAEA, 1998). Most of the fission products from the detonations are refractory and are incorporated into the glass. These will leach very slowly out of the glass as the glass itself slowly dissolves in groundwater. More volatile radionuclides, or those with a gaseous precursor (such as ^{137}Cs , which is produced by the decay of ^{137}Xe), occur both in the melt glass and as more easily dissolved deposits on rock surfaces. Though surface-deposited nuclides are more readily dissolved into groundwater than those in the melt glass, many are reactive and tend to sorb strongly onto mineral surfaces. Several radionuclides are mobile in groundwater, with the most significant being tritium. At Rio Blanco, tritium is also the most abundant of the gaseous radionuclides.

Because tritium is an isotope of hydrogen (half-life 12.26 yrs), it is able to form radioactive water and possibly methane (CH_4) molecules. Water exists in both the gas and liquid phases, while methane exists (under reservoir conditions) in only the gas phase. With respect to tritium, it appears that the pressure and temperature conditions are not sufficient for ongoing isotopic exchange reactions involving hydrogen and methane (Smith, 1975). Some tritiated methane, however, probably formed under the extremely high pressure and temperature conditions associated with the nuclear detonation. In addition, tritium can be present as hydrogen gas (either ^3HH or $^3\text{H}_2$), which can be as much as 13 percent of the gas phase (Toman and Tewes, 1972). Monitoring during flaring activities suggests that the majority of tritiated methane and hydrogen gas were removed during the production tests. All of the tritium, therefore, is assumed to be bound in the water molecule (i.e., little remaining tritium exists as either hydrogen gas or tritiated methane).

CONCEPTUAL FLOW AND TRANSPORT MODEL

The Mesaverde Formation forms a low-permeability, two-phase, fractured gas reservoir with volumetric gas saturation about 0.4. Oil, if present, was disregarded as an active phase. The formation intrinsic permeability was estimated as $3.4 \times 10^{-16} \text{ m}^2$, while the fracture permeability (associated with the nuclear detonation and production well hydrofractures) was estimated at $2.96 \times 10^{-14} \text{ m}^2$. The TRUST function (with the exponent provided by measured data on cores) was used for the capillary pressure relationship while the relative permeability was modeled with Corey's curves. Aqueous phase diffusion was neglected because diffusion coefficients are four orders of magnitude less than those in the gas phase. Low diffusion coefficients, coupled with low aqueous-phase velocities ($<10^{-11} \text{ m s}^{-1}$), are the reason that dispersion can be ignored in the aqueous phase. In natural porous media, mechanical dispersion in the gas phase can often be neglected as it is usually dominated by gas-phase molecular diffusion. This dominance occurs because gas phase diffusion coefficients are typically $\sim 10^{-5} \text{ m}^2 \text{ s}^{-1}$. For gas flow through porous media, a maximum velocity (u) might be 10^{-4} m s^{-1} (about 10 m day^{-1}), and a dispersivity (α) value (a characteristic pore diameter) for fine-grained sandstones could be 10^{-6} m . The mechanical dispersion coefficient, D_h , would be $D_h = \alpha u \sim 10^{-10} \text{ m}^2 \text{ s}^{-1}$, which is five orders of magnitude less than a molecular diffusion coefficient for a typical gas.

Figure 1 is a diagram of the flow and transport domain. Following the nuclear detonation, tritium diffuses radially outward from the nuclear cavity. After 35 years, a production well is located 290 m

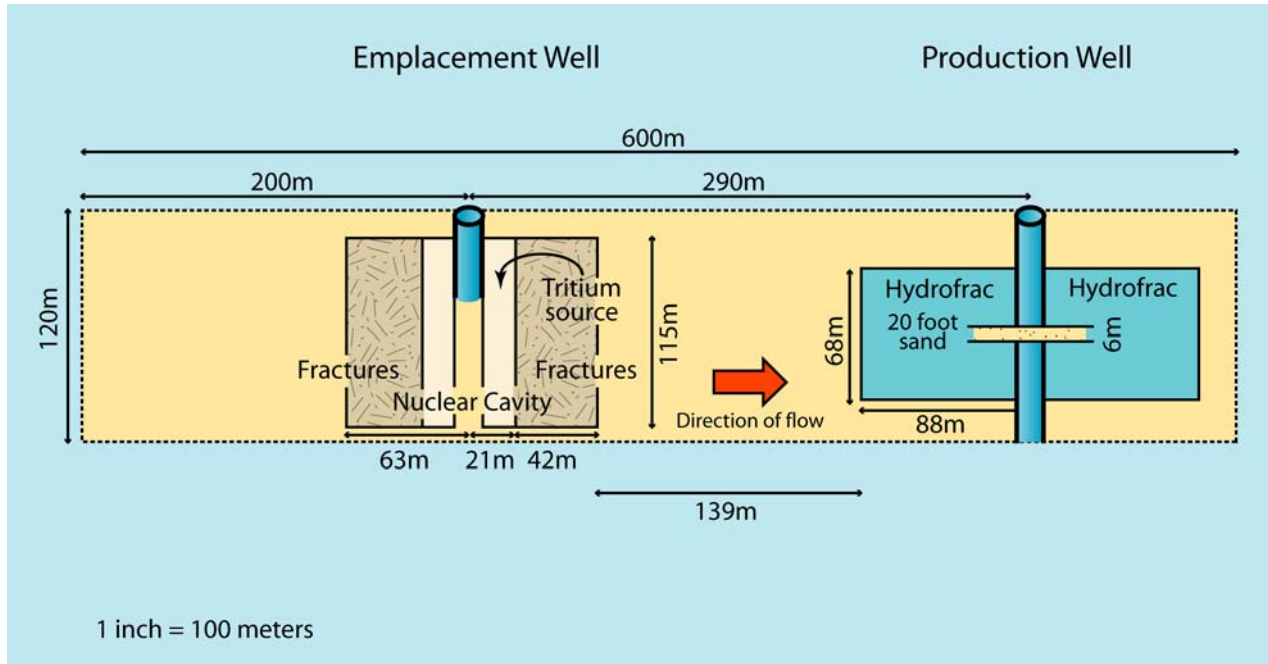


Figure 1. Vertical cross section for gas flow and tritium transport to a producing gas well. The boundary conditions are no flow and no solute flux on the upper and lower horizontal planes, prescribed pressure and prescribed (zero) mass fraction of solute on the left and right sides, and no flow and no solute flux on the planes parallel to flow in the transverse direction (i.e., into the page).

from the cavity, and production begins. Gas production occurs for 30 years, at which time it ceases. The effects of permeability, gas saturation, and production rate on tritium migration are examined in the results.

RESULTS

The first simulation (Figure 2) shows the transport of tritium away from a single cavity/chimney, toward a producing gas well located outside the current drilling exclusion boundary ($k = 3.4 \times 10^{-16} \text{ m}^2$ and initial liquid saturation, $S_l = 0.6$). Transport is from the cavity located near the left side of each slice toward the production well (the red vertical line) near the center of each figure. Each figure shows a vertical slice that bisects the flow field in the direction of flow (the x -direction). A second, parallel slice located 120 m away is also shown.

Tritiated gas fills the cavity within the first month of the nuclear detonation and diffuses radially outward from the cavity for the first 34 years. Just prior to the start of gas production (35 years after the detonation), the extent of the tritium is approximately 100 m from the center of the cavity (Figure 2a). Gas production occurs for 35 years (Figures 2b through 2c), after which time production ceases. The pressure gradient

induced by gas removal has only a limited effect on tritium transport, as radioactive gas is drawn only an additional 40 to 50 m during gas production. Figure 2d shows the mass fraction of tritium in the gas phase (X_g^{THO}) field 66 years after the nuclear detonations; at this time, tritium migration is balanced by its radioactive decay such that the plume dimensions have stabilized. Beyond this time (Figures 2e-h), the mass fraction field diminishes due to radioactive decay. Gas-phase tritium never reaches the production well under this scenario. Although not shown, tritium dissolved in the aqueous phase has a similar appearance to X_g^{THO} , with higher values of tritium mass fraction. Aqueous phase velocities are several orders less than those in the gas phase due to smaller liquid-phase diffusion coefficients, yet tritium in the aqueous phase has traveled the same distance as in the gas phase. The reason is due to phase change; that is, tritium moves away from the cavity in the gas phase and partitions into the aqueous phase. This partitioning acts to diminish X_g^{THO} at the expense of gaining mass in the aqueous phase. In combination with radioactive decay, this acts to constrain the dimensions of the plume, as both processes (phase partitioning and radioactive decay) work together to diminish mass fraction of tritium in both phases.

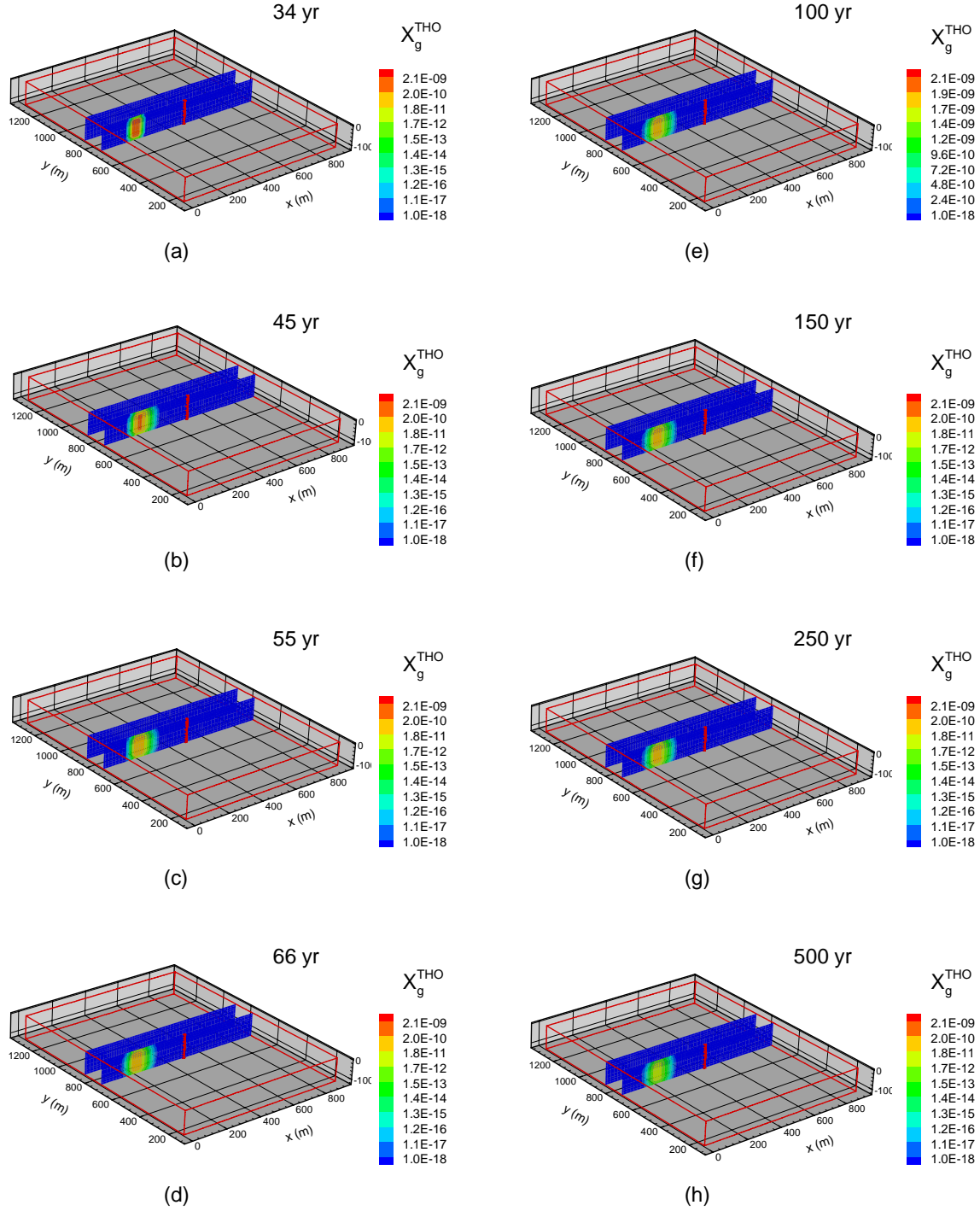


Figure 2. Mass fraction of tritium in the gas phase (a) 34 yr, (b) 45 yr, (c) 55 yr, (d) 66 yr, (e) 100 yr, (f) 150 yr, (g) 250 yr, and (h) 500 yr after nuclear detonation for the “reference” simulation, where $k=3.4 \times 10^{-16} \text{ m}^2$, $S_g=0.4$.

A simulation similar to the reference case, but with a one order of magnitude increase in intrinsic permeability, is shown in Figure 3a-d. The first 35 yr look similar to the previous simulation (and are not shown), as the difference is due solely to tortuosity associated with changes in saturation as a result of the different intrinsic permeabilities. Gas production, however (Figures 3a through 3c), has a slight effect of allowing tritium to migrate closer to the well, although only about 20 m closer when production ceases at 65 yr (Figure 3c) than in the reference simulation. The spread of tritium due to higher intrinsic permeability is evident in the second slice, located 120 m from main slice, where mass has migrated laterally in the y-direction.

A simulation was run in which the initial gas saturation was decreased from 0.4 to 0.2 as compared to the reference simulation (this required raising S_{lr} from 0.6 to 0.8 to prevent downward drainage of liquid water). The results of this simulation are presented in Figures 4a and b. There is no noticeable change in mixing when compared to the corresponding figures for the reference simulation ($S_g = 0.4$). Figures 4c and d show a simulation in which the initial gas saturation was increased from 0.4 to 0.6. Figure 4c shows that 30 years of production in conjunction with the high gas saturation has allowed tritium to migrate nearly 200 m from the center of the nuclear cavity. This is greater migration, by approximately 50 m, than any of the previous simulations. The reason for the further tritium transport is that the relative permeability has increased from approximately 0.45 to 0.70. Gas production acts to promote mixing and diffusion of tritium in the gas phase for high S_g ; however, after gas production ceases, tritium is more concentrated in the simulation with the low S_g . The “plume” tends to persist longer in the simulations with low S_g than in the simulations with higher S_g .

The choice of tortuosity model may significantly affect gas transport. Figures 5a and b show the X_g^{THO} field when a relative permeability based tortuosity model is implemented. There is little change in the mass fraction fields for tritium in either phase prior to the initiation of gas production from the well (not shown); however, the relative permeability model enhances transport in both phases once gas production has been initiated. This is seen at 45 yr (10 yr after the start of production) where tritium in both phases has diffused laterally (in the y-direction)

beyond the 120-m parallel slice located at $y = 860$ m. At 100 yr, the edge of the tritium field has come to within 50 m of the production well. In the liquid phase, the X_l^{THO} field is more uniform and never extends to within 100 m of the production well (Figures 5c and d).

SUMMARY AND CONCLUSIONS

The presence of the Rio Blanco underground nuclear test within a natural-gas producing field prompted investigation of subsurface migration of tritium in the reservoir. The Rio Blanco test was conducted in the Piceance Basin of Colorado, in the Mesaverde Formation, where reservoirs occur in very low permeability sandstone lenses. TOUGH2 was used to explore some of the processes controlling tritium migration toward a hypothetical gas production well. Gas diffusion was found to be an important part of overall transport, with only limited additional migration driven by advection from assumed gas production from a well. Varying intrinsic permeability and gas production rates within reasonable limits only impact the simulations by less than a factor of two. Conversely, increasing the initial gas saturation of the formation allows more transport as a result of the association of saturation with the gas phase relative permeability. The tortuosity model also has a significant impact on the transport pattern, with greater transport calculated for a relative permeability model, as compared to a saturation-based model. Of importance in all simulations are exchange and decay processes. Though migration occurs essentially only in the gas phase, exchange into liquid water serves to effectively trap tritium mass along the gas pathway. The tritium half-life of 12.26 years allows for significant mass decay over the time scales considered here.

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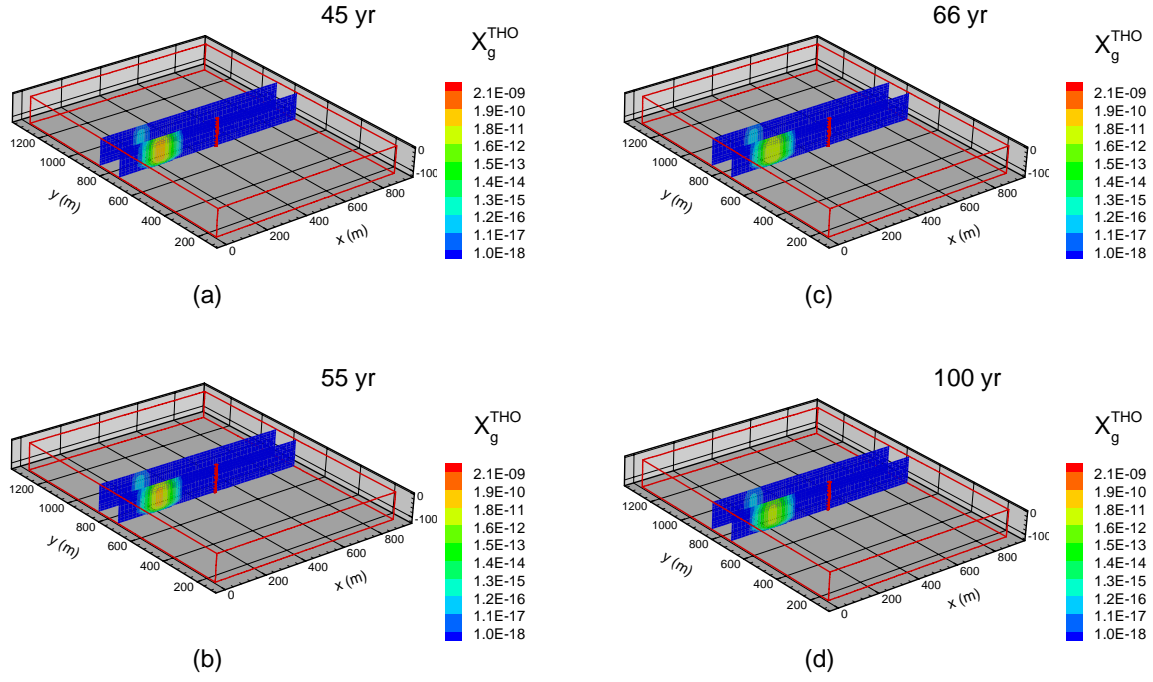


Figure 3. Mass fraction of tritium in the gas phase with intrinsic permeability one order of magnitude greater than for the “reference” simulation. The times are (a) 45 yr, (b) 55 yr, (c) 66 yr, and (d) 100 yr after the nuclear detonation.

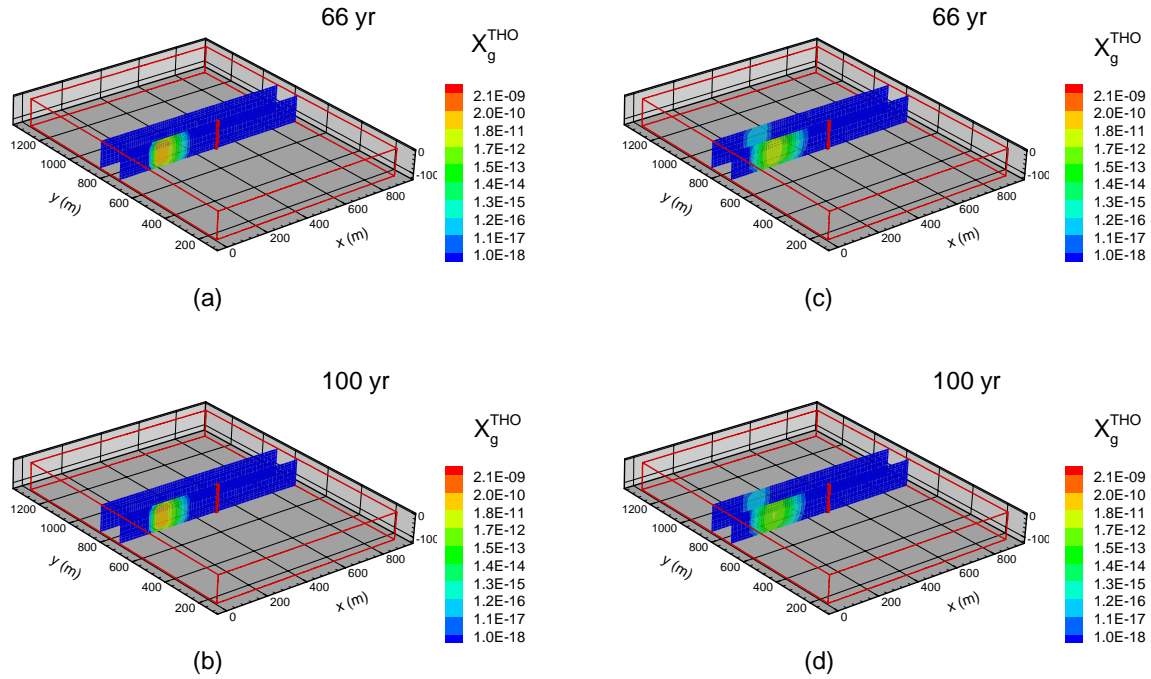


Figure 4. Effect of initial gas saturation on tritium mass fraction in the gas phase 66 and 100 yr after initial release of tritium: (a) and (b) show results for $S_g=0.2$, (c) and (d) for $S_g=0.6$.

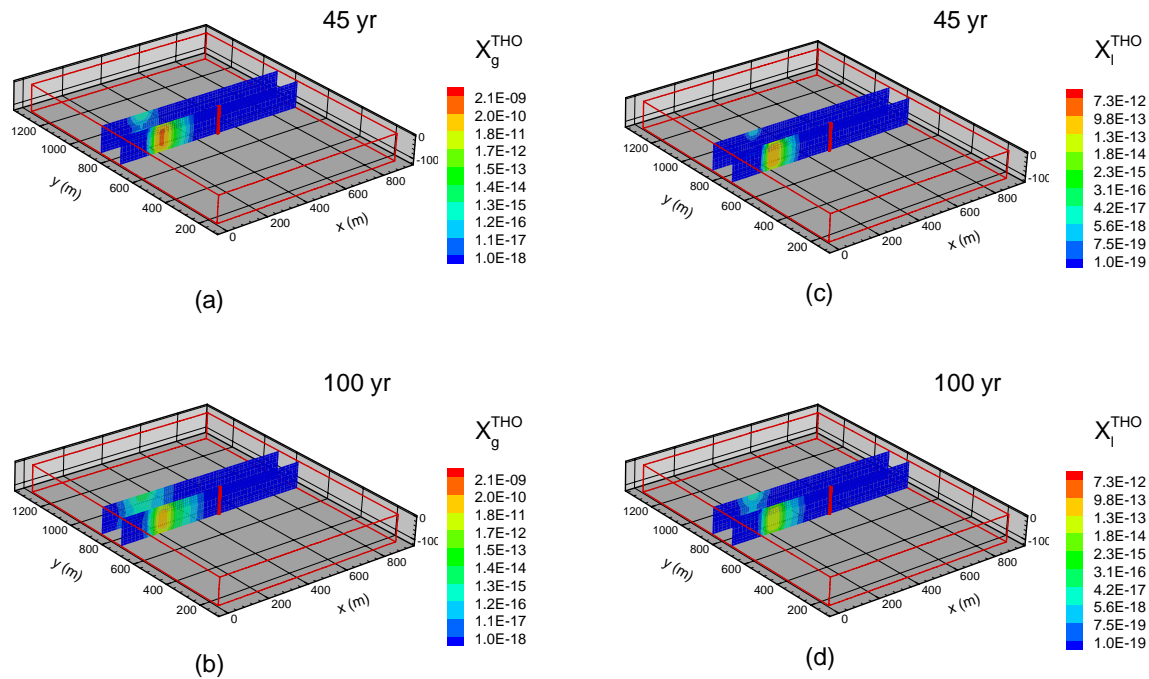


Figure 5. X_g^{THO} and X_l^{THO} for simulation with relative permeability tortuosity model. (a) and (b) show X_g^{THO} at 45 and 100 yr; (c) and (d) show X_l^{THO} for 45 and 100 yr, respectively.

REFERENCES

Borg, I.Y., R. Stone, H.B. Levy and L.D. Ramspott, *Information Pertinent to the Migration of Radionuclides In Groundwater at the Nevada Test Site, Part 1: Review and Analysis of Existing Information*, Report UCRL-52078 Pt. 1, Lawrence Livermore National Laboratory, 1976.

Cooper, C.A., M. Ye, J. Chapman, and C. Shirley, *Radionuclide Migration at the Rio Blanco Site, A Nuclear-Stimulated Low-Permeability Natural Gas Reservoir*, Report 45215, DOE/NV/13609-45, Desert Research Institute, Reno, Nevada, 2005.

Gerritsen, M.G., and L.J. Durlofsky, Modeling fluid flow in oil reservoirs, *Annu. Rev. Fluid Mech.*, v. 37, pp. 211-238, 2005.

International Atomic Energy Agency (IAEA), *The Radiological Situation at the Atolls of Mururoa and Fangataufa*, technical report in six volumes, published in Austria, 1998.

Johnson, R.C., Geologic History and Hydrocarbon Potential of Late Cretaceous-Age, Low-Permeability Reservoirs, Piceance Basin, Western Colorado,

Chapter E of *Evolution of Sedimentary Basins – Uinta and Piceance Basins*, U.S. Geological Survey Bulletin, 1787, E1-E51, 1989.

Nordyke, M.D., *The Soviet Program for Peaceful Uses of Nuclear Explosions*, Report UCRL-ID-124410, Lawrence Livermore National Laboratory, Livermore, Calif., 1996.

Rubin, B., L. Schwartz, and D. Montan, *An Analysis of Gas Stimulation Using Nuclear Explosives*, Rpt. UCRL-51226, Lawrence Livermore Laboratory, Livermore, Calif., 1972.

Smith, C.F., *Rio Blanco Gas Composition LLL Data Summary Calibration and Production Testing of RB-AR-02*, Report UCID-16762, Lawrence Livermore National Laboratory, 1975.

Toman, J., and H.A. Tewes, *Project Rio Blanco: Phase I Technical Studies*, Report UCID-15968, Lawrence Livermore Laboratory, 1972.