Radiotracer Medical Imaging Technologies Applied To Environmental Remediation Systems

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Abstract— We demonstrate an alternate application of high-sensitivity radiotracer imaging technologies developed for nuclear medical imaging. There are several available radioisotopes, namely ^{51}Cr and $^{99\text{m}}\text{Tc}$, that are particularly appealing for studying environmental remediation techniques, notably for modeling toxic element runoff through the soil near former nuclear facilities such as the Hanford reactors in Washington state (USA). ^{52}Cr (stable) and ^{99}Tc (2.3×10^6 year half-life) are important reactor-generated contaminants that have entered the groundwater. There are numerous studies being conducted to model transport of these compounds in different types of soil such as sand, clay, dirt and gravel. The speed and character of contaminant transport through medium depends on chemical composition of the particles, chemical composition of water, physical properties of the medium (particle size, homogeneity, temperature) and on content of different types of microorganisms. Measurements are typically performed by flowing water containing ^{52}Cr or ^{99}Tc through sediment columns (tubes a few cm in diameter and 0.5-1 meter long) and monitoring the in- and out- flows of contaminants. We will support the effort to develop different methods of reducing the contaminant transport speed by using SPECT imaging to study transport of the single-photon emitting surrogates ^{51}Cr and $^{99\text{m}}\text{Tc}$. By quantitatively imaging the contaminant distribution in the sediment column as a function of time, a much richer set of data can be obtained. We will compare contaminant transport in various media under different conditions including standard diffusion and flow at different speeds and volumes. Experimental work will be supplemented with theoretical analysis and modeling of transport processes, including studies of diffusion and chemical exchange using a standard multi-compartment model. In this conference record, we present the results of two studies of technetium $^{99\text{m}}$ transport in hydrated sand, including both planar and tomographic SPECT images of the columns acquired at different conditions.

I. INTRODUCTION

In addition to providing diagnostic information for patients in clinical settings, different medical imaging technologies find their uses in a variety of fields, including earth and ocean studies. In the field of environmental remediation, there is a strong interest in understanding how contaminants, especially heavy metals and radioactive ions, are transported through soils and groundwater aquifers. Contaminants are frequently observed to migrate further than conventional theory predicts. Although alternative models have been developed to describe anomalously fast transport in the subsurface, experimental tests are needed. In this paper, we consider the application of single photon computing tomography (SPECT) methods to aid efforts of environmental remediation. Radiotracer imaging is a very sensitive technique for measuring the migration of contaminants in permeable materials without physically removing and

measuring numerous samples. Our efforts in this project are directed toward studying remediation activities such as ongoing work on the geobiochemical determinants of the dispersion of contaminants or precipitation of contaminants (e.g., use of biostimulation to alter soil properties).

An example of the importance of radioisotope methods is given for implementation or containment procedures for the waste effluent from the Hanford reactors in Washington State. Despite the almost \$60 billion estimated cost [1], the remediation project will not solve the problem of toxic elements that have permeated the soil. Thus, part of the massive effort currently underway includes research on how to immobilize these elements in the soil before they reach the Columbia River through groundwater transport. Two of the many elements that enter the groundwater and eventually the Columbia River are ^{99}Tc (2.3×10^6 year half-life) and chromium: both the stable but toxic element ^{52}Cr and, before the reactor closures, ^{51}Cr (28-day half-life). ^{99}Tc is a key radionuclide found in nuclear fuel radioactive waste from processing spent fuel, as well as radioactive wastes associated with operating nuclear reactors. Cr arises from the sodium dichromate that was mixed with cooling water of nuclear reactors during Hanford's production years to the 1980s. Also, chromium is a major water-polluting agent found in different types of industrial waste.

The requirements for the contaminant transport imaging and corresponding modeling are commensurate to those in clinical SPECT, so existing SPECT cameras and existing medical radiotracers will be used. In this paper, we present initial research that establishes the groundwork for SPECT application in environmental remediation studies. Section II explains the relevant concepts and facts concerning waste propagation and containment strategies. Section III describes how nuclear medicine will be applied to study the waste propagation, and Section IV lists the results of preliminary experiments.

II. THEORY

Waste containment problem and possible solutions. A major problem in toxic and radioactive waste disposal is proper containment of the waste. Figure 1 illustrates the expected distribution of Cr and Tc waste propagation from the Hanford storage facility outflow. For this case, it is expected that ^{99}Tc and stable chromium will continue to contaminate the effluent until immobilized before entering the Columbia River. Thus, there is an urgent need is to block the contaminant discharge from the soils around the Hanford Site. The chemical method is to reduce the oxidation states of both technetium and chromium and thus render these elements immobile. Tc^{VII} is soluble, but Tc^{IV} can form a solid when associated with sediment. The reduction of Tc^{VII} can be accomplished by certain forms of iron or by microorganisms. Similarly, Cr^{VI} is immobilized as Cr^{III} , which can be achieved using glycerol polyacetate, a safe compound that upon hydration releases lactate and phosphate [2]. Our goal in

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this study is to establish the optimal medium parameters and methods of medium modification, which would lead to creating environments where Tc and Cr transport slows down as much as possible.

Figure 1. Columbia River water plume mapped by salinity distribution in 1964, matches measured technetium-99 concentrations measured years later. [3]

Three main approaches to containing Tc and Cr waste as it moves through the soil will be addressed later in this project: toxin reduction to a non-soluble valence state by changes in pH, changes in iron content and by introducing species of anaerobic bacteria that reduce the oxidation state of the toxic ions. All three of these approaches can be modeled in columns and observed in real-time using radioactive isotopes of Cr and Tc and medical SPECT cameras.

III. METHODS

Imaging isotopes. Both Cr and Tc have isotopes with half-lives suitable for nuclear tomographic imaging. Technetium isotope ^{99m}Tc with 140 KeV γ -ray emission is a standard medical imaging agent used in cardiac imaging, renal imaging, and cancer diagnostics. The relatively short (6 hours) half-life of ^{99m}Tc limits studies to at most 4 half-lives (24 hours). Chromium radioactive isotope, ^{51}Cr emits 320 KeV γ -rays with half-life of 27.7 days. While longer half-life makes ^{51}Cr an ideal radiotracer for studying slow ground transport processes, the high energy of its radiation may require the use of medium energy collimators and additional shielding introduced to the SPECT scanner. In this work, we are presenting technetium-99m results. ^{99m}Tc was used in sodium pertechnetate (NaTcO_4) form.

In addition to the radionuclide imaging, some preliminary experiments have been performed with coloring agent added to the white quartz sand as shown in Figure 2(a). The experiments were analyzed both by means of visual inspection and by image analysis of color photographs of the dispersion in light-colored medium.

Medium preparation. Different types of media occurring in nature will be used in this project. These include soils, clay, sands, dirt, gravel, soaked in water of varying salinity and acidity. At the technology development stage, specially prepared materials will be used as propagation medium, too. Currently, we use both lab-grade quartz sand and standard construction-grade

sand with particle size range controlled by sieves. In addition to varying physical properties of the medium, different chemical compositions will be considered. Technologies of medium preparation and supporting models have been developed in the literature [4-5]. Specifically, we are studying differences in ^{99m}Tc transport through iron-enhanced media since iron compounds are principal candidates for reducing the valence state of technetium and chromium ions.

Imaging platform and acquisition setup. The imaging platform for both isotopes is our GE Millennium VG Hawkeye SPECT/CT system (Figure 1(a)).

Figure 2. (a) Concept experiment: using red dye diffusion and flow in different types of sand. (b) Experimental setup for planar SPECT of Tc diffusion or flow in sand using standard 1"-diameter tubes.

Experimental setup. The medium of interest is mixed with water and placed in vertical columns and positioned between parallel plates of SPECT camera as shown in Figure 2(b). Two types of imaging protocols are considered: planar and parallel beam tomographic imaging, both at 4.4 mm spatial resolution. In the future, sub-mm resolution pinhole SPECT will be added to provide high-spatial resolution 3D images.

Planar images will be collected to evaluate the vertical component of transport for the case of fast flow through the column. Time resolution of 1-2 seconds and spatial resolution of 4.4 mm are easily achievable for the planar imaging mode.

In order to extract maximum information about the flow properties in columns, three-dimensional images of isotope distribution are needed. These images are obtained by reconstructing tomographic data acquired using the SPECT camera. Two different acquisition methods have been used. In one scheme, the column is placed horizontally on the patient table of the scanner. This approach has been used in an experiment where the NaTcO_4 solution was propagating through the column from the top to bottom forced by gravity. In order to prepare the column for horizontal imaging, both ends are sealed. This precludes effective real-time flow measurement. The second approach uses the setup shown in Figure 3: NaTcO_4 solution is pumped upward. The same approach (upward pumping) is typically used in geophysics and geochemistry experiments. In order to generate a complete tomographic dataset, the setup has to be modified to enable column rotation in the field of view of the scanner, so the SPECT camera operates in the planar

imaging mode, and the planar data acquisition is synchronized with the column rotation driven by an external motor.

In addition to the SPECT acquisition, a CT-scan of the column is acquired and used for attenuation correction in emission image reconstruction.

Figure 3. Basic setup of an experiment repeating a standard column study design used in geochemistry: water or radiotracer solution is pumped upward through a vertical column. Column rotates between the detector heads of a SPECT scanner. The scanner acquires planar data, so that acquisition time frames are synchronized with the column rotation. The acquired data are adjusted and re-formatted to create standard tomographic datasets.

Data analysis. Experimental studies can be used with theoretical and numerical models. A series of three-dimensional snapshots of bolus propagation through the system will be used to extract information about the local flow properties using simple compartment model methods similar to those described in [6].

IV. RESULTS

Diffusion model and color agent propagation. Theoretical model of Tc diffusion in water (diffusion coefficient $\alpha=10^{-5}$ cm²/sec) has been developed and studied numerically. During a few half-lives of ^{99m}Tc, the diffusion distance is only 2-3 cm, as shown in Figure 4(a). This is the lower limit of propagation speed, which will increase with the addition of flow. In porous media, α is expected to be smaller, so the radiotracer distribution is mainly determined by flow and surface tension effects.

Diffusion and flow experiments were conducted using the proof-of-concept model of standard food dye in different media, see Figure 1(a). Three types of media were used: standard white sand, street (gray) sand and hydrated pharmacy-grade the Sephadex G-25. First, only diffusion-driven transport was observed for about 24-hour period. Then, the bottoms of the test tubes were punctured and flow effects were added. Flow was maintained by adding additional clear water on top of the sand or gel with dye. Concentration of the color dye was deduced from optical images of the test tube. Although changes were visible in all media, only Sephadex environment provided sufficient clarity and adequate white background that allowed us to extract numerical value of the distribution, as shown in Figure 5(b).

Figure 4. (a) Standard diffusion model: numerical solution of Fick's diffusion equation with a constant boundary. (b) Vertical distribution of food dye concentration in Sephadex extracted from colored images (insert on the right).

Experiment 1. Sodium pertechnetate solution has been propagated through a vertical sand-filled column. A 63-mm wide and 50 cm-long quartz tube was filled with construction-grade sand. Sand particle sizes in 160-600 μ m range were selected by sieving the sand through a series of sieves with prescribed dimensions. One hundred μ Ci of activity in a form of radioactive slurry (30 ml) was added to the top of the column, and gravity driven by a steady flow of clear water, at approximately 1 ml/min. A series of planar images was acquired to trace the flow propagation, then the column was sealed, placed horizontally on the SPECT scanner patient bed and imaged. Sixty 10-second 128x128 pixel views were acquired at 3° per view and reconstructed using a standard ML-EM algorithm with attenuation correction. Both planar and tomographic results are provided in Figure 5.

Figure 5. (a) Planar images showing downward radioisotope propagation through a vertical column. (b) Sagittal slice through the reconstructed 3D volume: non-uniformity of bolus propagation is apparent. (c) Axial slices through the reconstructed volume with 1 cm slice-slice separation.

Experiment 2. An experimental setup shown in Figure 3 was established to track propagation of ^{99m}Tc sodium pertechnetate over the period of 7 hours. A 50 x 6.6 cm column was filled with pure lab-grade quartz sand, hydrated and agitated for 3 days to achieve optimal packing. Twenty mCi of activity was diluted in 250 ml of water and pumped up the column at approximately 4 cm / hour. The activity was introduced as a 3 ml bolus, followed by about an hour of pumping clear water, followed by pumping the radioactive solution. Six-minute tomographic datasets were acquired in 10-minute intervals. In Figure 6, we show the reconstruction results for the data acquired during the last interval, 7 hours after the start of the experiment. When compared to the experiment described in Figure 5, one can see that the spatial inhomogeneity of the activity front propagation is less pronounced. This fact is explained by a combination of factors, including upward pumping and better uniformity of the propagation medium.

Figure 6. Reconstruction results for the Tc solution pumped up through a quartz column. (a) Sagittal slice through the center of the column. (b) Axial slices.

The 4D dataset acquired in the present experiment will be analyzed further in order to calculate quantitative value of flow speed, which clearly varies within each axial plane and in order to evaluate possible inhomogeneities in the packing fracture.

V. CONCLUSIONS

Our preliminary results and theoretical analysis clearly demonstrate that nuclear medical imaging can be an effective tool in environmental remediation research. It can provide volumetric spatial and temporal information on the distribution of Tc and Cr compounds in different types of soils and other media subjected to different levels of water flow. Tomographic data on radioisotope concentration are quantitative and can be obtained as time-series. Availability of the 3D images of the contaminant distribution is crucial because of the non-uniformity of the radioisotope concentration even in homogeneous environments. Time-dependent data is essential for measuring the dynamic properties of contaminant propagation.

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