Computer Modeling and Laboratory Experiments to Design Sorbents for Remediating Radiologically Contaminated Soil and Water: Nanoscale Structured Surface Layers

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Abstract—This research illustrates the application of a combination of computer modeling and laboratory experiments to designing nanoscale structural surface layers of compositions for remediating radiologically contaminated soil and water. Natural sorbents and sorption materials with nanoscale structured surface layers of various structures are compared in terms of sorption efficiencies. Monte Carlo simulation to generate numerical calculations and chemical and physical analyses to verify the modeling predictions of the structure of metallic surface zones synthesized by ionic beams was performed. Fe, Al, and Al₂O₃ compositions were irradiated with Ti ions to form nanoscale structured surface layers of various structures. ⁹⁰Sr and ¹³⁷Cs were used in the laboratory experiments to assess sorbent performance. The results demonstrate that using the sorbents with new structural forms of a surface produces increased efficiency in potential radionuclide sorption.

Keywords—Ionizing Fluxes; Radiation Factor; Materials Science; Modeling; Natural Sorbents; Nano Structured Surfaces; Synthesized Compositions; Soil Solutions

I. INTRODUCTION

Environmental remediation of aquatic and terrestrial sites that have been contaminated by varying levels of radioactivity increasingly is done using various kinds of natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents. Using sorbents to remediate contaminated sites is attractive because a number of radionuclides such as ¹³⁷Cs and ⁹⁰Sr have relatively long half-lives, exhibit water solubility, and can be readily incorporated into the natural sorbents.

II. METHODS

The computer modeling portion of this research used Monte Carlo simulation to create new nanoscale metallic structures and assess their sorption performance. The Monte Carlo method using the program SCATTER [5] for the modeling description of ion interaction with substance by binary impacts was used for this simulation. The interaction effects and as stopping of incident particles at elastic and inelastic collisions, scattering inside iron samples, defect generation, particle reflections from the target as well as its dispersion by both incident and reflected particles were estimated according to the software program SCATTER of the study [8].

The laboratory experimentation phase was used to validate the computer modeling predictions. For that portion of this research, plasma technology was used to create Fe, Al, and aluminum oxide (Al₂O₃) compositions with nanoscale structured surface layers of various structures. Ionic plasma synthesis on exposure to cryogenic plasma streams of zirconium (Zr) and titanium (Ti) on Fe and Al respectively was used for formation of the compositions.
selected for evaluation. Prior to creating the structures, the input materials were polished and purified to remove oxide
skin and other contaminants before subjecting the input materials to ionic irradiation. The ionic irradiation was
carried out in a vacuum chamber (5 x 10^-6 mm Hg) at ion energy values from 0.1 to 2.0 keV. The structure and
properties of the synthesized compositions were determined using multiple methods of physical and chemical analysis,
including X-ray phase, local X-ray spectrum, micro durometeric analysis, nuclear gamma resonance (NGR), and
electronic microscopy.

III. RESULTS

This section summarizes the results of the computer
modeling using Monte Carlo simulation to generate
numerical calculations and the empirical analysis of the
structure of metallic surface zones synthesized by ionic beams.

A. Monte Carlo Modeling

The first stage of this research employed Monte Carlo
modeling to test the theoretical assumptions about the
formation process of the nano compositions based on the
interaction of plasma streams with Al and Ti for varying

<table>
<thead>
<tr>
<th>Plasma Charge</th>
<th>Embedding Depth for Ion Energy of 1000 eV</th>
<th>Embedding Depth for Ion Energy of 2000 eV</th>
</tr>
</thead>
<tbody>
<tr>
<td>+1</td>
<td>0.35</td>
<td>0.93</td>
</tr>
<tr>
<td>+2</td>
<td>0.92</td>
<td>2.60</td>
</tr>
<tr>
<td>+3</td>
<td>1.73</td>
<td>4.70</td>
</tr>
<tr>
<td>+4</td>
<td>2.60</td>
<td>7.50</td>
</tr>
</tbody>
</table>

Fig. 1 presents a graphical depiction of the results of the
Monte Carlo simulation used to calculate the distribution of
Ti ions by embedding depth in Fe as a function of the initial
energy stream using a binary collision model for the
interaction of Ti ions with Fe. The initial energy stream
plasma charges and embedding depth for ion energy. In
order to illustrate the results, we focus this portion of the
paper on discussing the results for simulating the embedding
of Ti ions in Fe to form a nanoscale metallic composition.

Table 1 shows the calculations generated predicting
possible penetration depth into Fe for 100 eV and 2000 eV
for plasma charges of +1, +2, +3, and +4 for the direct
"embedding" of ions of multi-charge plasma of Ti. Table 1
reveals that holding the plasma charge constant and
doubling the ion energy are estimated to produce a
substantial enhancement in terms of embedding depth.
When the plasma charge = +1 the embedding depth
increases by a factor of 2.66, by a factor of 2.83 for plasma
charge = +2, by a factor of 2.72 for plasma charge = +3, and
by a factor of 2.88 for plasma charge = +4 shifting from an
ion energy level of 1000 eV to 2000 eV. Holding the ion
energy constant at either 1000 eV or 2000 eV and increasing
the plasma charge assumed as an input value for the
computer simulations similarly is predicted to enhance
embedding depth for these compositions. As a result,
increasing both the plasma charge and ion energy is
predicted to produce structures with the greatest depth of
embedding Fe in Ti, with a +4 plasma charge and 2000 eV
ion energy estimated to result in an embedding depth of 7.50.

evaluated in this study ranges from 300 eV to 1500 eV. At
the same time, the calculation of parameters of this process
was carried out by the Monte Carlo method using the binary
collision model as the interaction modeling of ions with
substance. The calculations predict the process of
interaction of ions with a target depends on both the value of
stream energy and the specific irradiated metal. The most
probable penetration depth of Ti ions in Fe increases
nonlinearly as the initial energy increases and peaks at
approximately 10 x 10^-8 m and then decreases rapidly. Thus,
the relative half-width of the distribution quickly increases
with the corresponding energy growth for Ti. For the energy
range used in this simulation, Ti ion penetration into Fe
surface layers average between 300 to 600 nanometers. The
computer modeling results support the conclusion that
binding structural surface layers can create nanoscale compositions.

![Fig. 1 Cross-section of embedding of Ti ions in Fe as function of the initial stream energy](attachment:fig1.png)
B. Experimental Analysis

The second stage of this research employed a series of laboratory experiments conducted in order to evaluate the surface layers associated with the nanoscale structures synthesized by ionic beams. The experiments produce empirical results consistent with the Monte Carlo modeling predictions. Taken as a whole, the chemical and physical analyses confirmed displacement of the Fe atoms by the Ti ions results Fe dispersion, Ti concentration on a surface, and their mutual hashing that can be used at the composition formation to create new structural forms of sorption material surfaces.

Fig. 2 shows that the structure of surface zones synthesized by ionic beams depends on both the basic metal of the target and the metal of an ionic beam. Comparing the structure of Fe and Al or carbon (C) and Al$_2$O$_3$ surfaces after impact of Ti ionic beams reveals distinct differences in hard-grained structure and density. Moreover, the sedimentation pattern for C and Al$_2$O$_3$ impacted by Ti ions is less dense when compared to the sedimentation pattern obtained for either Al or Fe. Thus, the laboratory experiments confirm that formation of the distinct metallic structures observed under experimental conditions is a function of differentials in the energy transferred from ionic beams to atoms in a crystal lattice of metal during the irradiation process thereby influencing the nanoscale structure of surface zones.

Local X-ray spectrum analysis was used to investigate the distribution of Ti on Fe surface and in surface zones. Measurements indicated that the range for zone width of the modified structure in both Fe and Al was from 5 to 10 microns (μ). The range for zone width for Al$_2$O$_3$ was reduced up to 2-3 μ less.

NGR-spectroscopy revealed spectra of surface zones represented the well-solved Zeeman sextets for characterizing nanocrystalline structure formation. The additional fine structure of spectrum caused clusters of Ti atoms is shown on Fig. 3.

The macro clusters of Ti shown on Fig. 4 (white color) were marked on the samples by means of the local X-ray spectrum analysis.
X-ray phase analysis confirmed that the structure of surface zones of Fe and Al was composed of oversaturated solid solutions of Ti in Fe and Laves phases such as TiFe₂. Table 2 summarizes the chemical structure and properties of the base material and characteristics of the surface zones investigated. The accuracy of these results was confirmed by also investigating the structure of surface zones using electronic microscopy in addition to the x-ray phase analysis. The size of particles that formed intermetallics (e.g., a class of metallic materials that form ordered crystal structures) such as TiFe₂ was found on the average to be in the 100-700 nm range.

### Table 2: Characteristics of Surface Zones Synthesized in Fe and Al

<table>
<thead>
<tr>
<th>Chemical Structure of Base Material, Mass%</th>
<th>Properties of Base Material</th>
<th>Properties of Surface Zones</th>
<th>Width of α-Zone, μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe - 100%</td>
<td>Phase Structure</td>
<td>Lattice Period α-Fe, nm</td>
<td>α-Fe, α-Ti, Fe₂, Ti</td>
</tr>
<tr>
<td>Al - 100%</td>
<td></td>
<td></td>
<td>Al, α-Ti, Al₂, Ti</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td></td>
<td></td>
<td>Al₂O₃, α-Ti, Al₂, Ti</td>
</tr>
</tbody>
</table>

The next step in the experiments involved the use of micro durometeric analysis. Micro durometeric analysis was employed using samples of the Fe and Ti compositions to evaluate micro hardness of a surface. The structure of the synthesized surfaces exhibited increased mechanical properties. For example, the surface zones synthesized in Fe had higher values of micro-hardness on Vickers (H_V); both an initial condition (1200-1500 MPa) and an increased temperature of 700-1000°C (1000-1450 MPa) in comparison with initial Fe (600 MPa), Al and its oxides. Similar increases in surface hardness at the formation of compositions on the basis of Al and oxides also were demonstrated in the experiments.

The final portion of the laboratory experiments involved an evaluation of the sorption properties of the synthesized compositions. In this step, the decrease in ⁹⁰Sr and ¹³⁷Cs radioactivity in soil solutions using an Al₂O₃ sorbent was compared to the results using a composite sorbent. For this purpose, aliquot solutions in volume of 10-25 ml were selected. The measurements of specific activity were carried out before and after the introduction of Al₂O₃ sorbents in active solutions, as well as before and after the introduction of composite sorbents on the basis of Al₂O₃ with the new nano structured form of a surface. Fig. 5 shows the plot of sorption ability K. The graph indicates the change in activity for each radionuclide in relation to its alimentation in the liquid phase. The results demonstrate that the synthesized sorbents increased radiological decontamination efficiency on the average by 1.2 to 2.5 times in comparison with Al₂O₃.
nanocrystalline structure sorbents and precipitable radionuclides will accelerate the remediation of radiologically contaminated soil and water.

ACKNOWLEDGMENTS

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REFERENCES


