Environmental Management Science Program

Project ID Number 60283

Waste Volume Reduction Using Surface Characterization and Decontamination by Laser Ablation

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June 1, 1998

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Research Objective

Much of the contaminated concrete from nuclear facilities contains radionuclides only in the near surface region. Removal of the contaminated layer would greatly reduce the volume of waste requiring storage. The objectives of this research are to understand the depth-dependent concentration and chemistry of radionuclide-contaminated concrete surfaces, to determine the mechanism and efficacy of laser ablation in removing contaminated surface layers, and to chemically and physically characterize the captured ablation effluent which would become the stored waste.

Research Progress and Implications

This report summarizes work from the first eight months of a three-year project. Samples consisted of specimens of non-contaminated high density concrete from the Experimental Boiling Water Reactor at Argonne National Laboratory (sectioned into small coupons) and samples cast in the laboratory using Type I Portland cement and either silica, alumina, or sand. Some samples of concrete and cement were doped with Cs-133. Ablation experiments were done with a 1.6 kW pulsed Nd:YAG laser. The beam was delivered via a fiber optic cable, focused to a 0.5 mm spot, and rastered across the surface. The ablated material was collected with a vacuum shroud system and captured on a 0.2 micron filter. Virgin and ablated surfaces were chemically analyzed with laser desorption surface mass spectrometry.

The cement samples melted and vaporized under the influence of the ablation laser. There was no significant difference attributable to the different fillers (i.e. silica and alumina) used. Optical and electron microscopy showed smooth melted surfaces and aerosols composed almost exclusively of hollow spheres less than about 20 microns in diameter. In addition, the aerosols contained a minor fraction (less than about 5% by volume) of grainy agglomerates. Energy dispersive spectroscopy (EDS) showed that the chemical composition of the spheres was essentially identical to the original Portland cement, while the agglomerates were enriched in aluminum. This finding may have significance in light of literature reports showing that Cs ion binds preferentially to alumina and aluminum-rich silica phases [1, 2]. The Cs concentration in our samples was too low to be detected by EDS.

In contrast, ablation of interior sections of high density concrete resulted in a range of ablation mechanisms, from melting and vaporization to disaggregation, depending on whether the laser was focussed on aggregate particles or the cement matrix. The wide range of behavior is attributable to differences in optical absorption and melting points of the various materials present in the sample. The original exterior surface of the high density concrete was simpler than the interior sections, consisting of a 1-2 mm thick layer of Portland cement / sand matrix mostly free of large aggregates due to settling during casting. This material was ablated with the highest efficiency of any sample tested, with ablation rates up to ten times higher than the fresh Portland cement samples cast in our laboratory (0.4-0.8 cubic millimeters/pulse vs. 0.07 cubic millimeters/pulse). The material disaggregated under the influence of the laser, with particles up to 0.5 mm in diameter dislodged. The difference in ablation behavior may be due either to high sand content in the high density concrete or to the expected presence of calcium carbonate in the near-surface region of the sample after many years exposure to air and/or water [3-5].

Cs-doped Portland cement and high density concrete samples were analyzed in ultrahigh vacuum by surface mass spectrometry. Material was vaporized by a pulsed laser focussed to a small (~1 micron) spot on the surface. The desorbed atoms were then ionized by a second laser focussed to a spot above the surface, and analyzed by a time-of-flight mass spectrometer. Laser desorption mass spectrometry and sputtered neutral or secondary ion mass spectrometry are unique in their ability to analyze highly differentiated surfaces such as these with high sensitivity and lateral resolutions of 1 micron or less. The analysis showed Mg, Al, Si, and Ca, representative of the cement matrix, and Na and K, representative of the cement pore water and interstitial surfaces [6, 7]. Cesium was found to be associated with the pore water ions. Preliminary analyses of Portland cement samples doped with known amounts of Cs showed a Cs detection limit in the part per million range. The detection limit is expected to drop as the experimental technique is refined.

A sample of the exterior surface of the aged high density concrete was exposed to an aqueous solution of CsCl and ablated with the high power Nd:YAG laser. Comparison of the virgin and ablated surfaces showed a 30- to 300-fold reduction of Cs after removal of 0.6 to 0.8 mm of material in a single ablation pass.

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Planned Activities

Over the next six to twelve months laser ablation studies will be extended to a more diverse set of samples. Different kinds and amounts of fillers will be used to understand differences in ablation mechanisms. Quantitative surface chemical analyses based on laser desorption, sputtered neutral, or secondary ion mass spectrometry will be developed and used to analyze for cesium and strontium in both the ablated surfaces and captured effluent.

This work was supported by the U.S. Department of Energy, EMSP, under Contract W-31-109-ENG-38.