Waste Volume Reduction using Surface Characterization and Decontamination by Laser Ablation

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Introduction

Research need addressed: *The characterization and decontamination of concrete surfaces*

DOE has 23 million cubic meters of concrete in contaminated buildings that requires characterization and possible decontamination. The radionuclide contamination in much of the concrete is present only on the surface or in the near-surface regions (typically less than one inch). Removal of the contaminated layer would greatly reduce the volume of waste requiring disposal. The goal of this work is to develop laser-based methods of characterization and decontamination. In addition, the work has broader implications in that the techniques under development may aid in other areas of concrete research, particularly in chemical analysis and ion transport in the cement matrix.

Laser ablation / effluent collection schematic

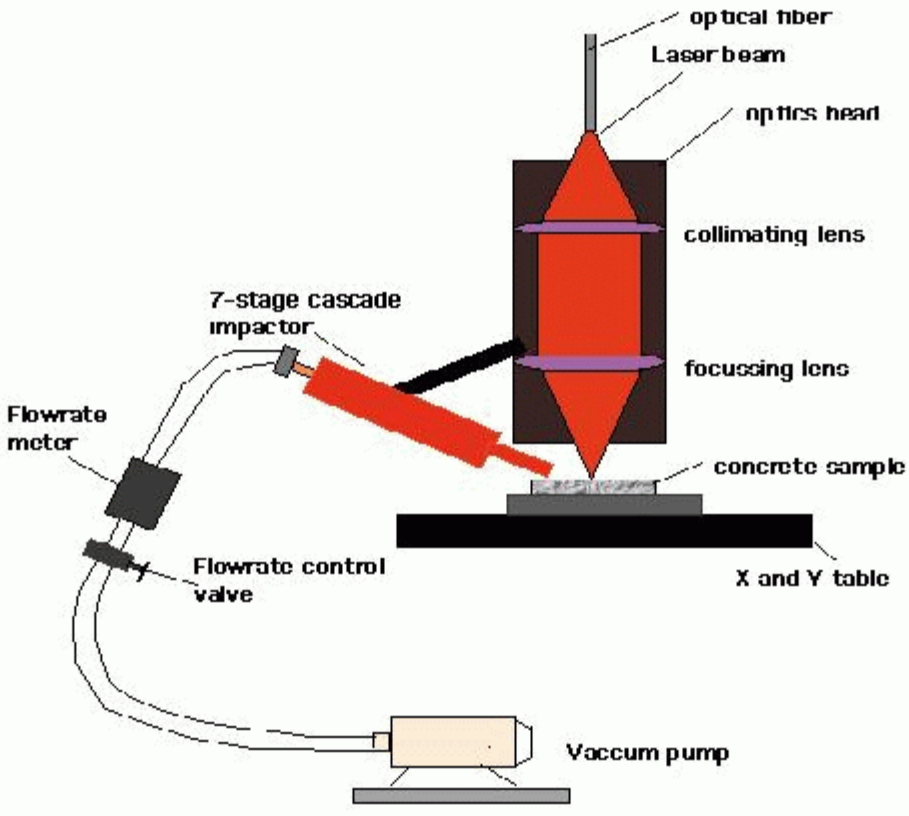
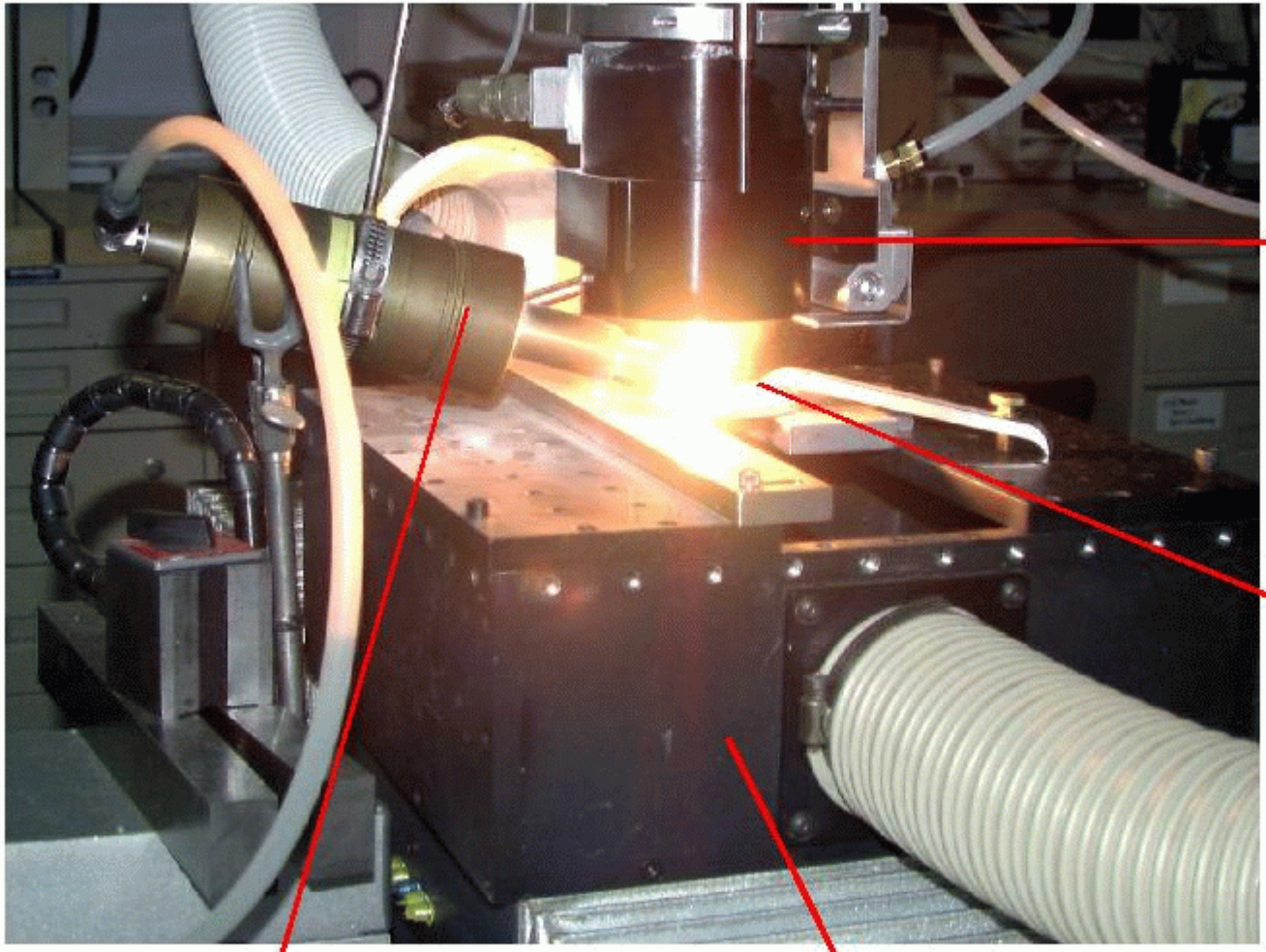


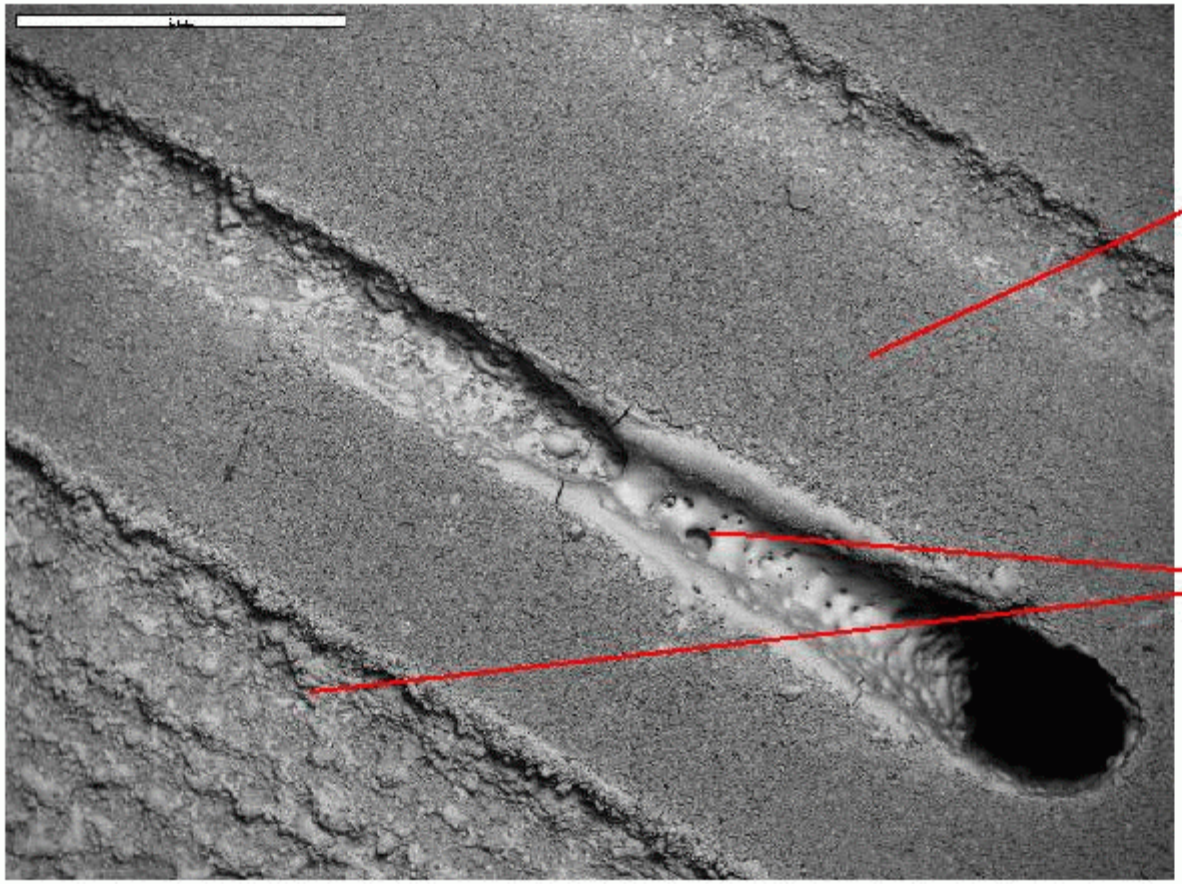
Figure 1. Schematical presentation of the filtration assembly

Laser Ablation System

•1.6 kW pulsed Nd:YAG laser system with fiber optic beam delivery

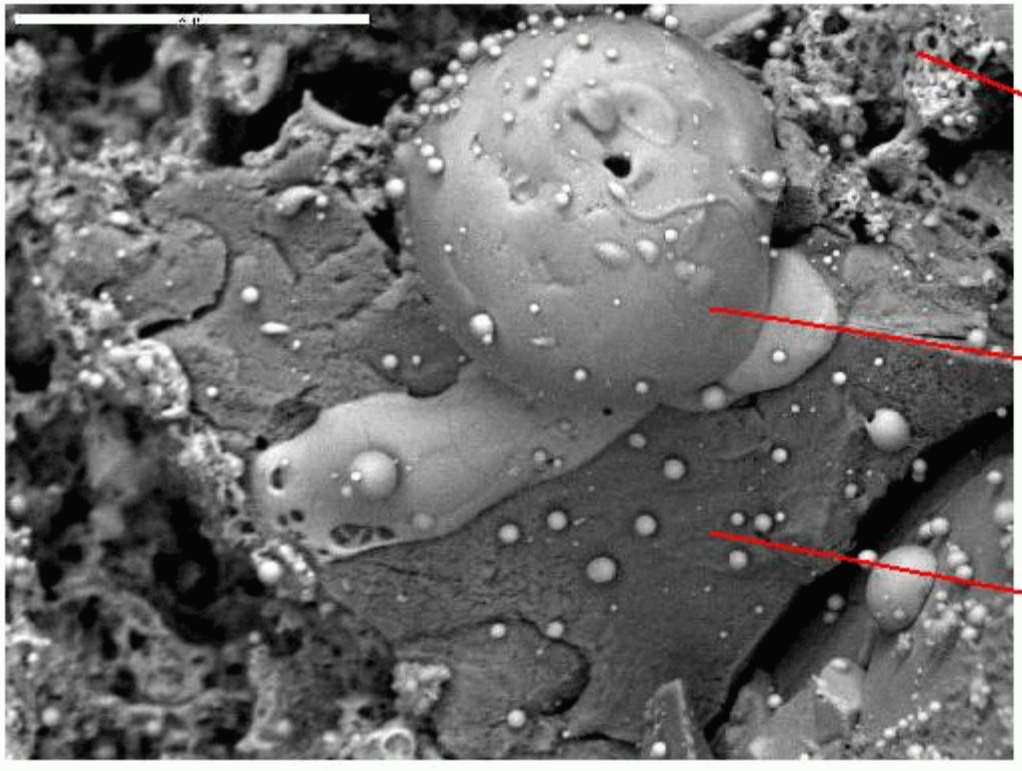


Cement surface after laser ablation



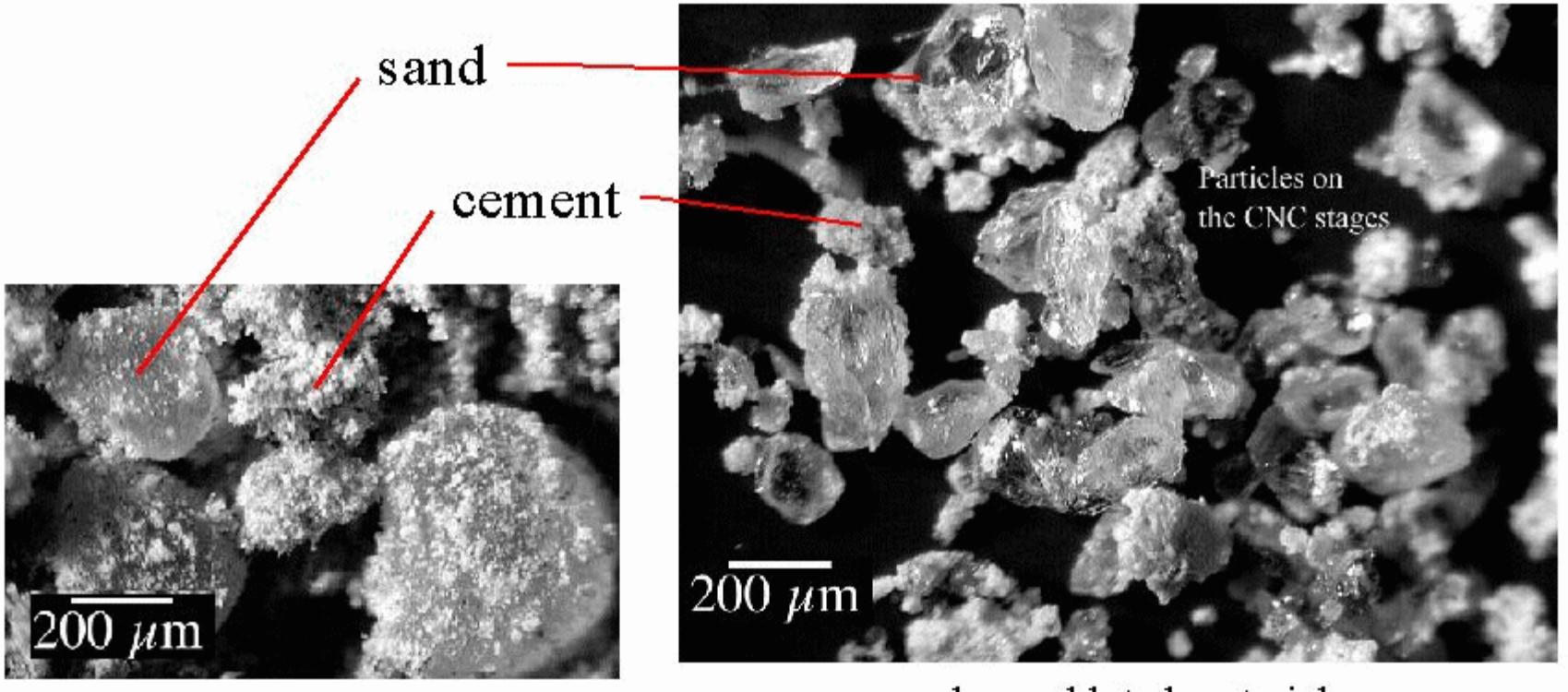
Scanning electron micrograph showing melting of the ablated cement surface. Ablation tracks are ~0.6 mm wide by 0.5 mm deep. Pulse repetition rate = 400 Hz.

Cement surface after laser ablation



High magnification scanning electron micrograph of ablated cement surface showing melting and foaming of cement matrix due to rapid melting and dehydration, and a fractured sand grain.

Effluent from laser ablation of cement: fractured & dislodged material

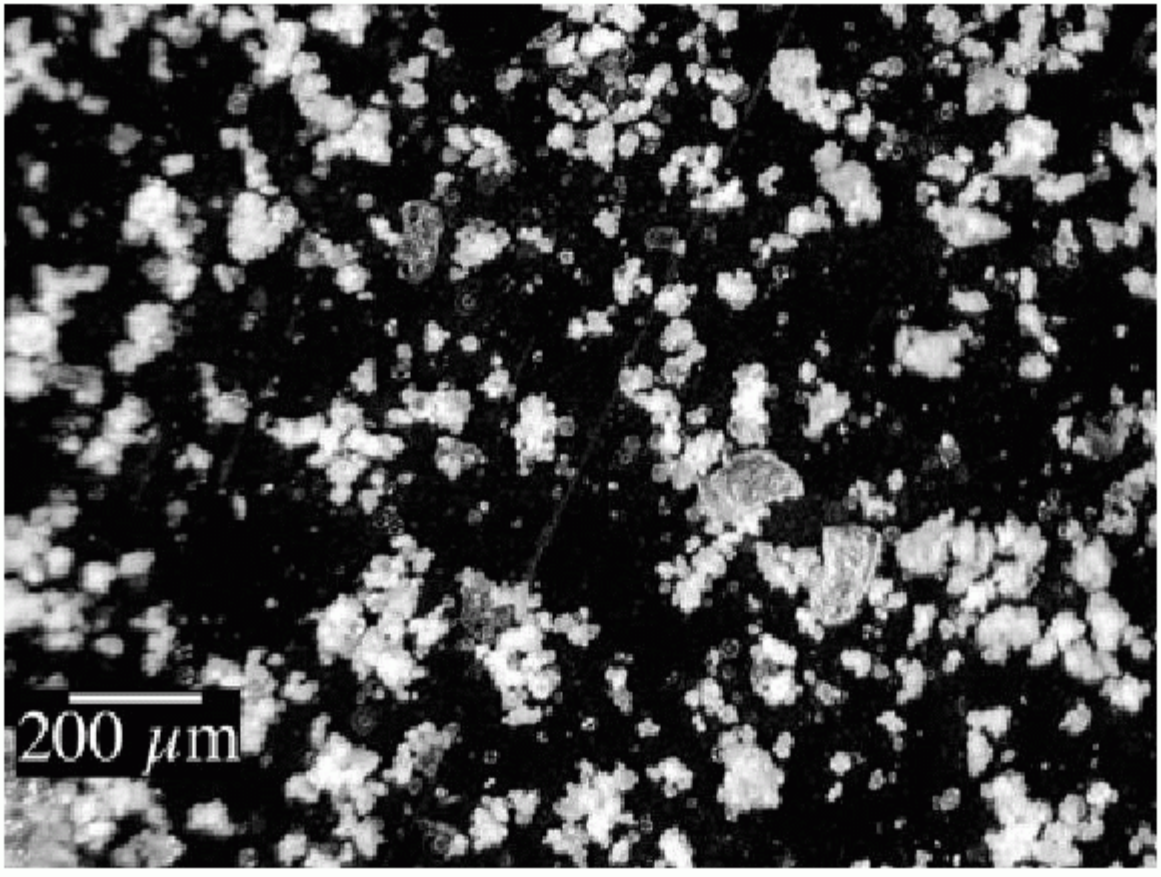


mechanically crushed material

laser ablated material

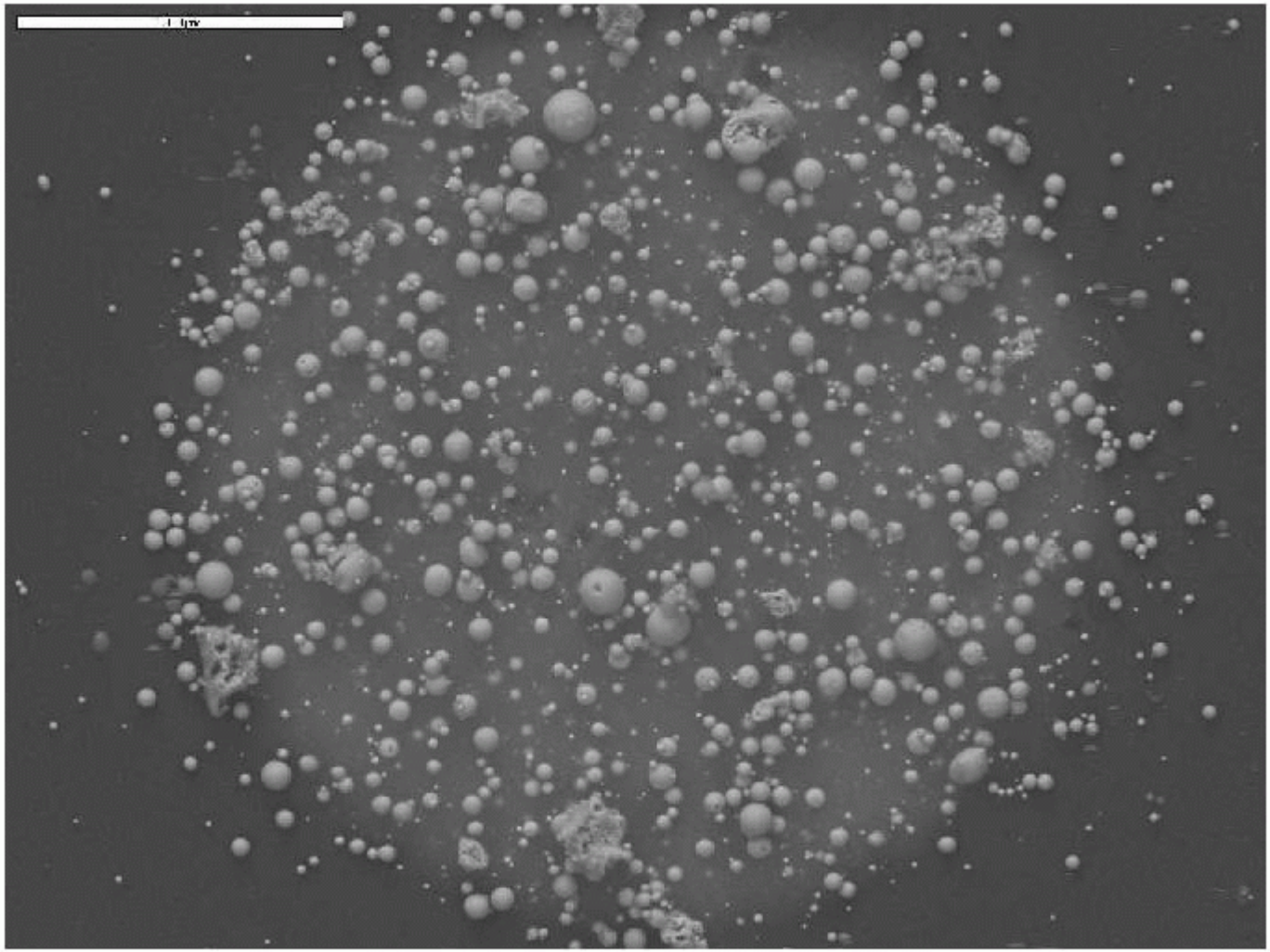
The ablation effluent from a single experiment spans sizes ranging from chunks hundreds of microns across to sub-micron aerosol particles. These optical micrographs show a mixture of fractured sand grains and cement matrix material, along side of a mechanically crushed sample for comparison. Rapid thermal expansion during laser heating produces shock waves that fracture and dislodge chunks of material.

Effluent from laser ablation of cement: disaggregated material



This optical micrograph shows intermediate-size effluent particles, mostly disaggregated cement matrix material.

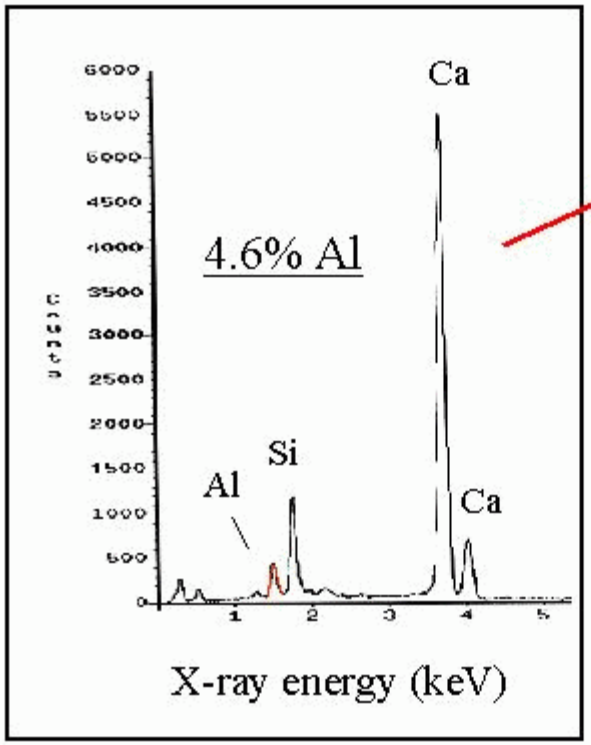
Effluent from laser ablation of cement: aerosol particles

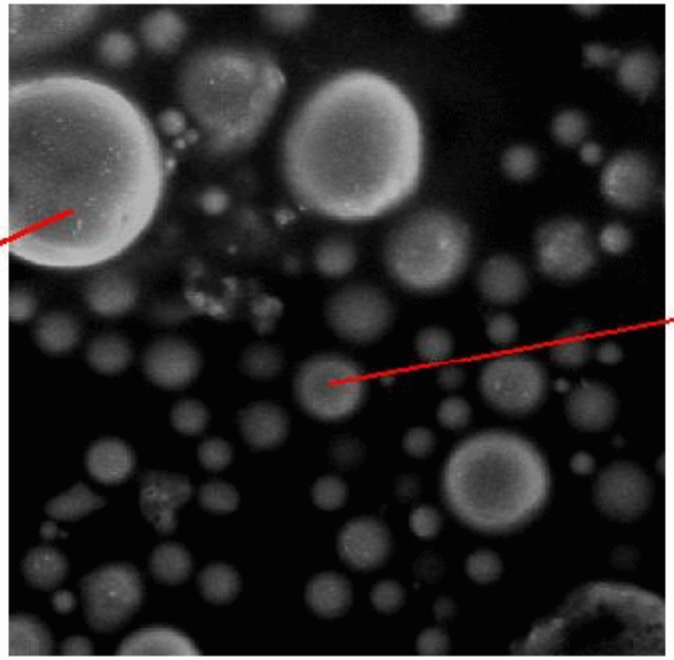


Electron micrograph of small aerosol particles collected on a high efficiency filter. Larger spheres with dimples/holes are formed from liquid spattered off the surface during ablation. Smaller solid spheres are formed by vaporization and condensation.

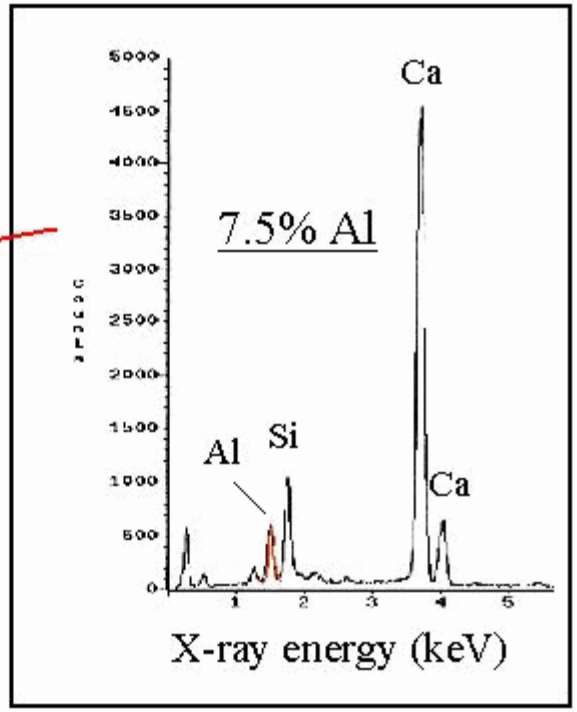
Energy dispersive x-ray analysis of aerosol particles

16 μm sphere



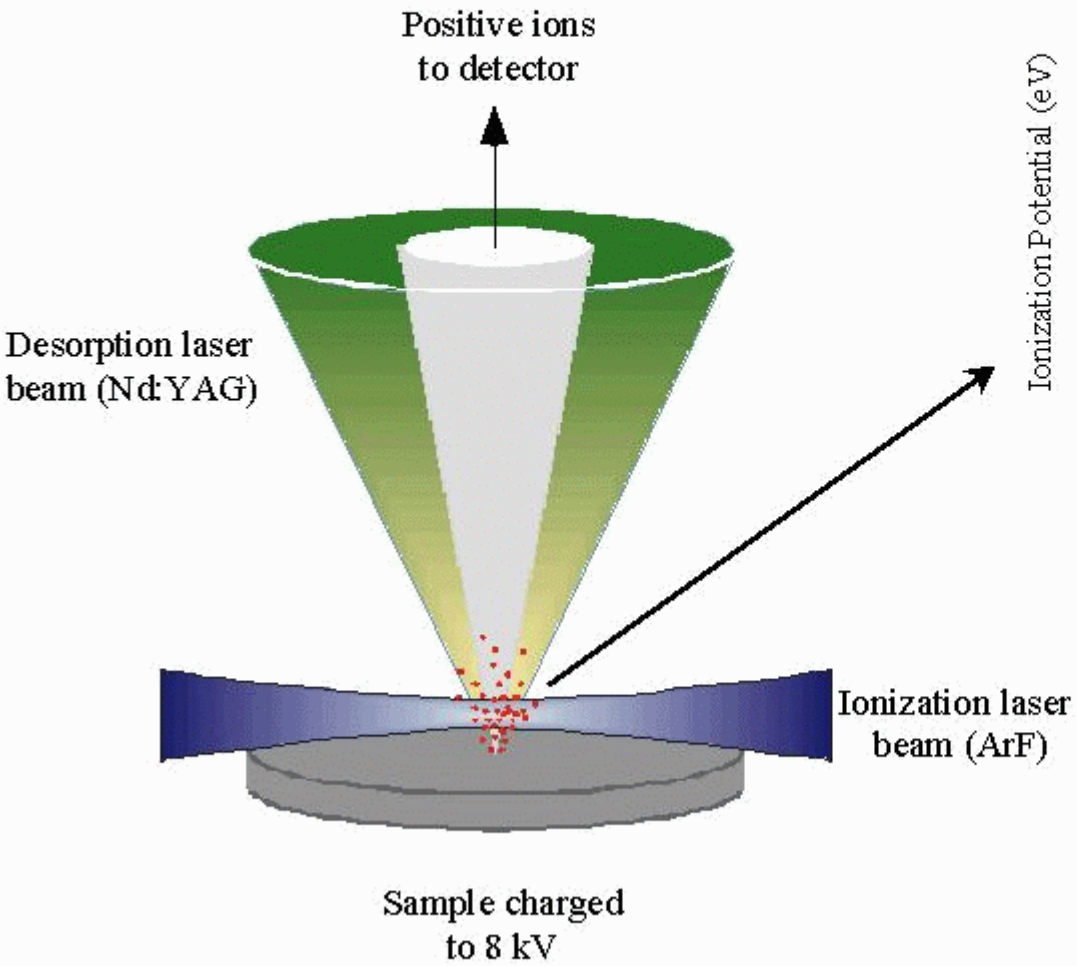


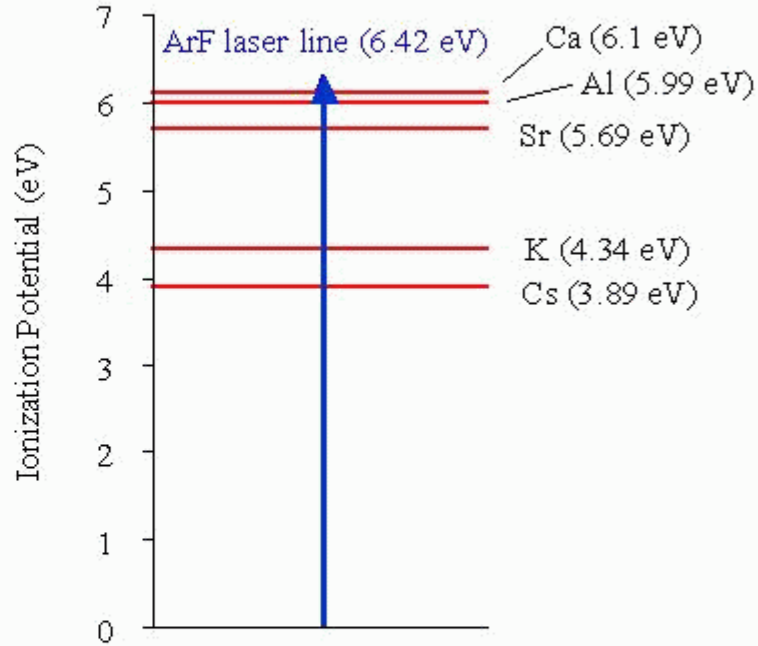
5 μm sphere



Aluminum content of small spheres increases as sphere diameter decreases, possibly indicating aerosol particle nucleation by an aluminum-rich phase. Hydrated Portland cement Al content is 2 - 2.5%.

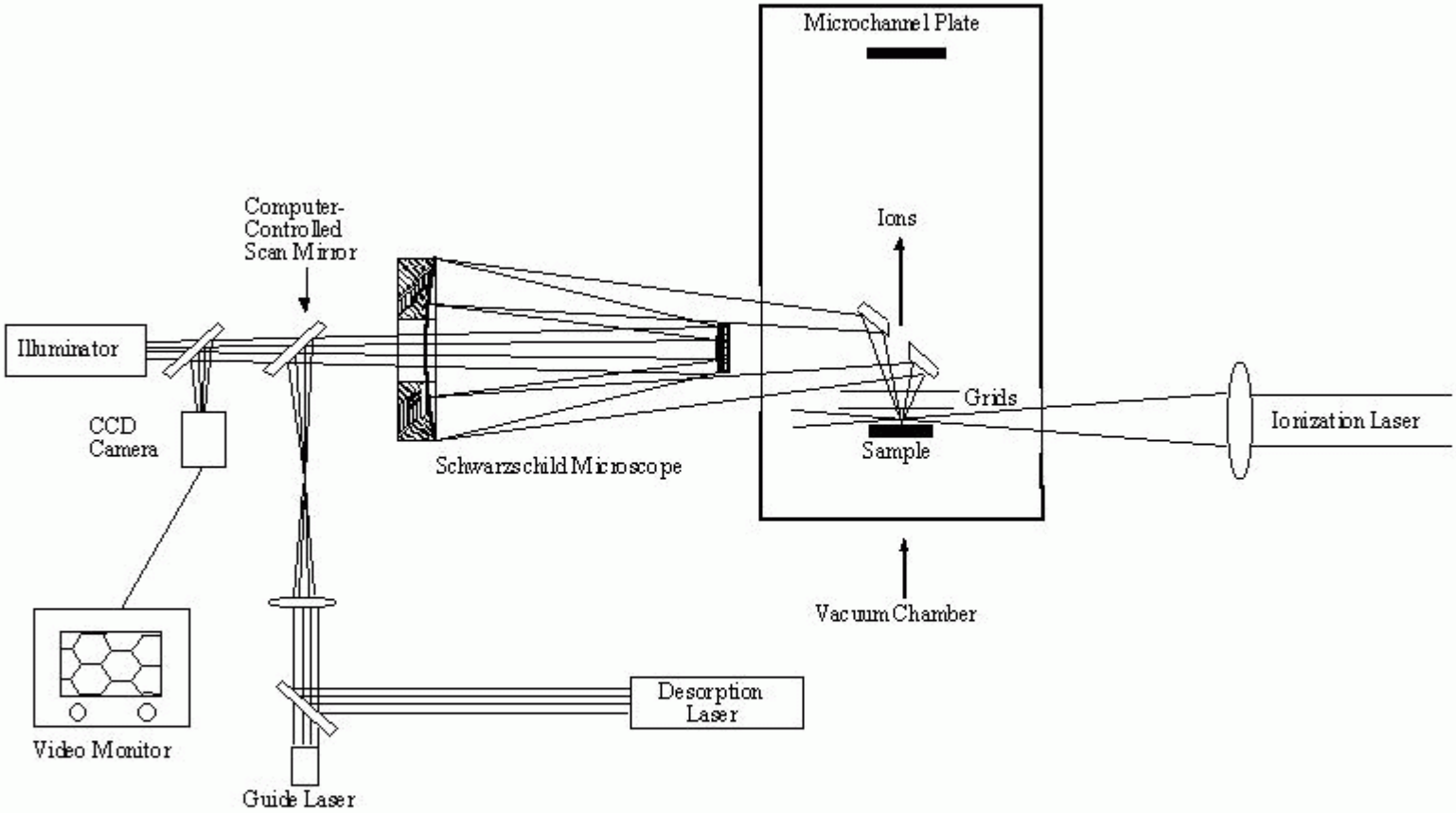
Laser Desorption / Laser Ionization Mass Spectrometry





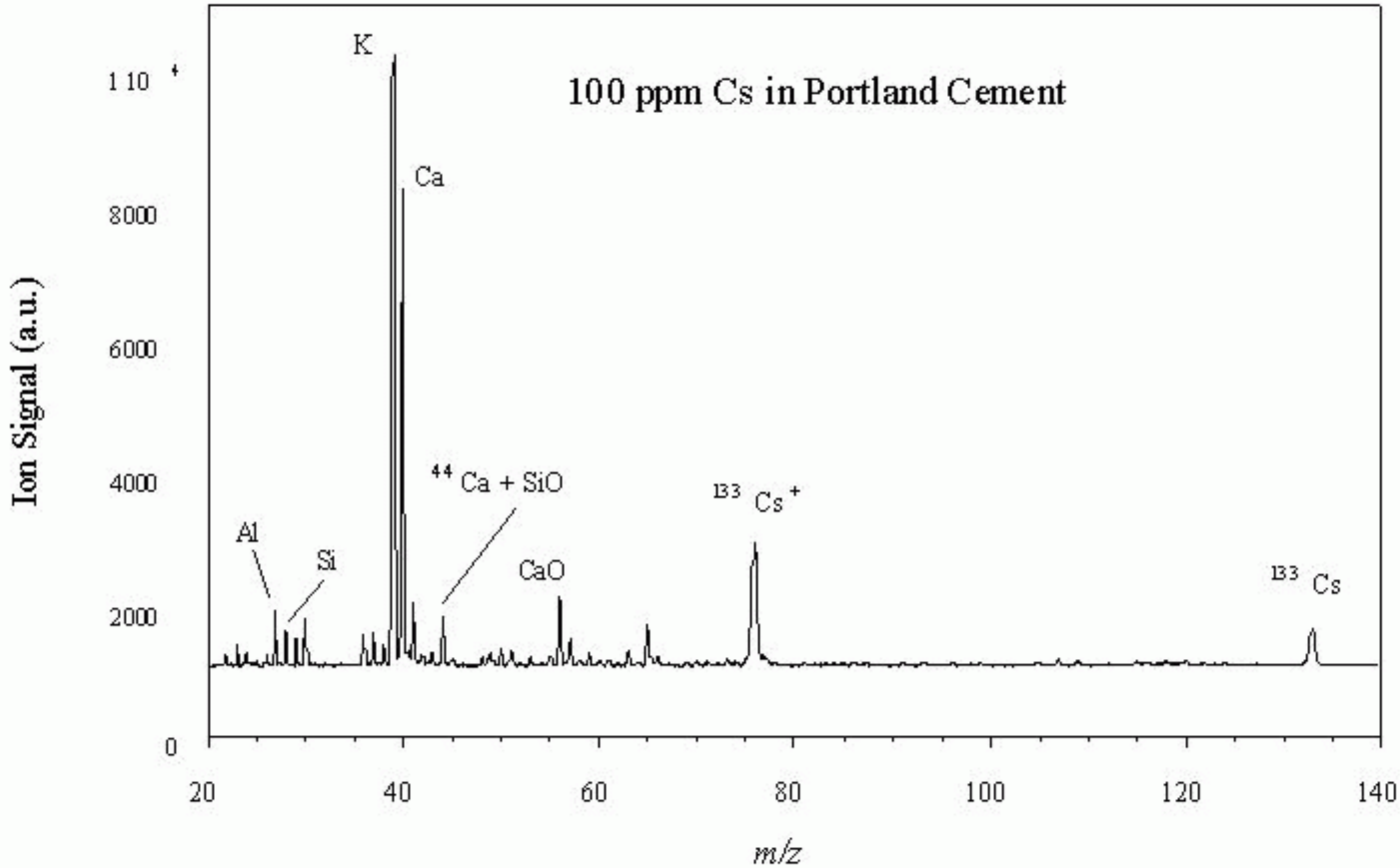
One-photon ionization of desorbed neutral atoms by ArF laser

Laser Desorption / Laser Photoionization Mass Spectrometer



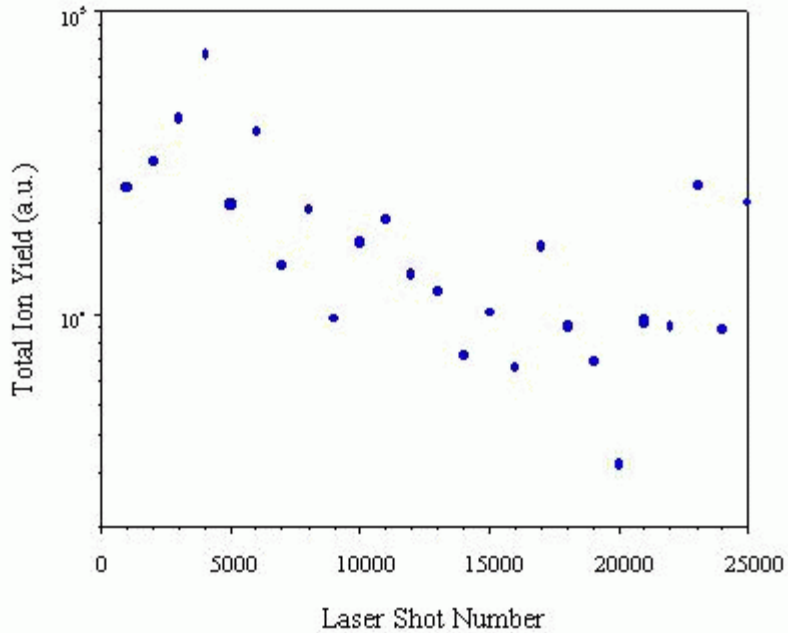
Sample is placed in a high vacuum chamber for time-of-flight mass analysis. Computer-controlled scan mirror can be used to generate chemical images of surfaces with micron spatial resolution.

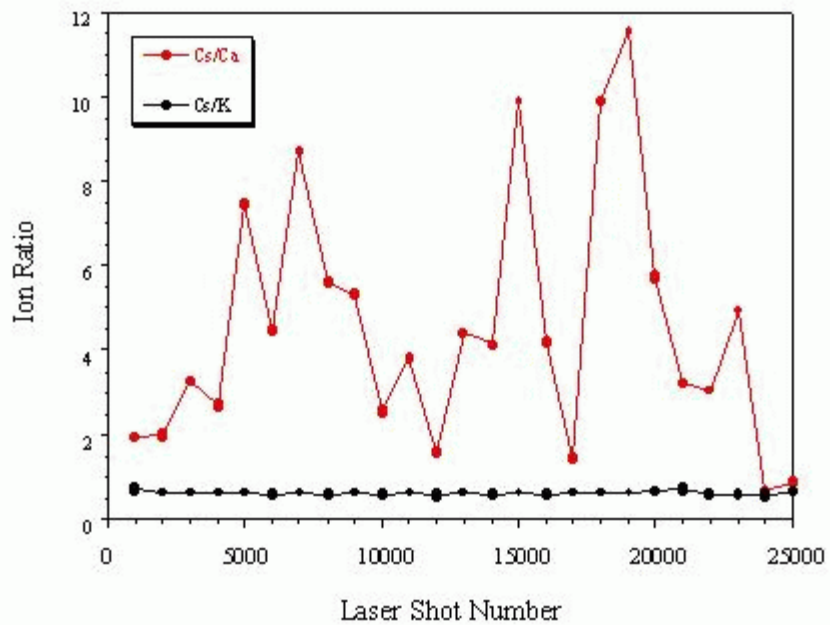
Laser Desorption / Laser Photoionization Mass Spectrum of Cs-doped Type 1 Portland Cement



The peak heights are not a measure of the relative amounts of each element. For example, cement contains many orders of magnitude more Ca than K, but since K desorbs and ionizes much more easily than Ca, it actually gives a larger signal in the mass spectrum. The Cs detection limit for this technique is 1-10 ppm.

Cesium Speciation in Type 1 Portland Cement

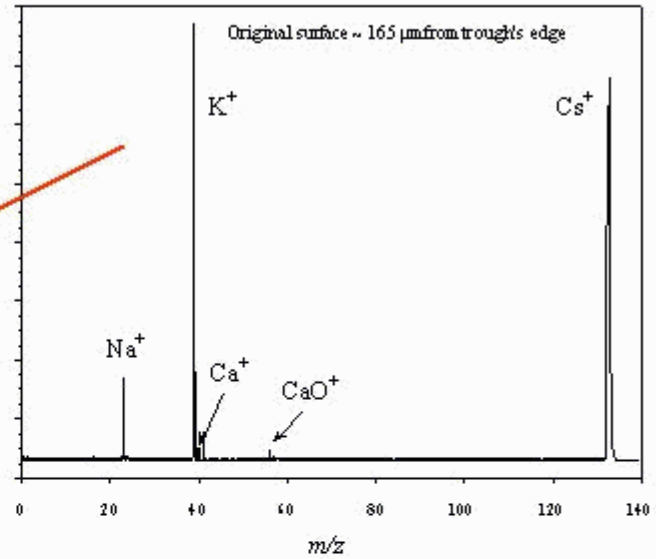


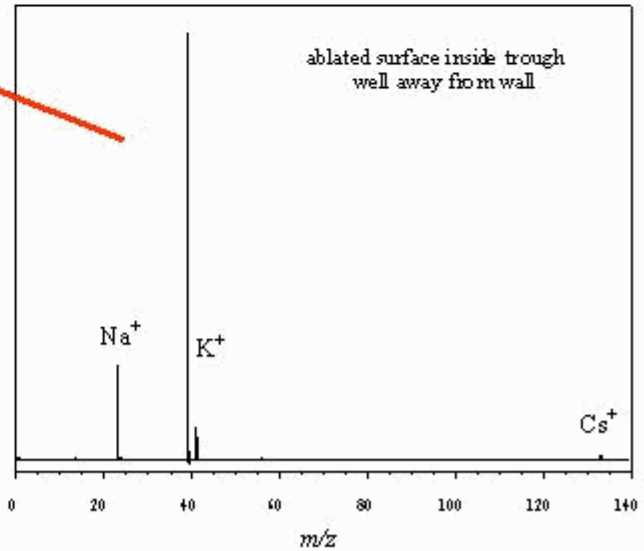


Potassium is present in cement primarily as ions in the cement pore water, while Ca is a cement matrix atom. In the laser desorption experiment, ion yields vary from shot to shot, and fall off as a function of total laser exposure (left figure), however the Cs ion signal always correlates with the K ion signal, but never with the Ca, regardless of sample history (right figure), showing that the Cs in Portland cement also exists primarily as an ionic pore water species.

Ablation & analysis of Cs-treated high density concrete

Sample of a high density concrete wall from the Experimental Boiling Water Reactor at Argonne National Laboratory treated with 0.4 M CsCl





Microbeam laser desorption mass spectra show substantial diminution of cesium in ablated areas. The ablation tracks are 0.5 to 1 mm deep and show a substantial Cs reduction.

Conclusions

- High power laser ablation of concrete produces several different types of effluent including fractured particles, disaggregated material, and aerosol particles
- Aerosol particles are nucleated by an aluminum-rich phase
- Laser mass spectrometric analysis shows cesium associated with cement porewater
- Cesium mass spectrometric detection limit is 1-10 ppm
- Laser ablation of only 1 mm of surface from a sample of Cs-treated concrete results in a substantial reduction of Cs