Concrete Surface Removal and Characterization Using High Power Laser Ablation

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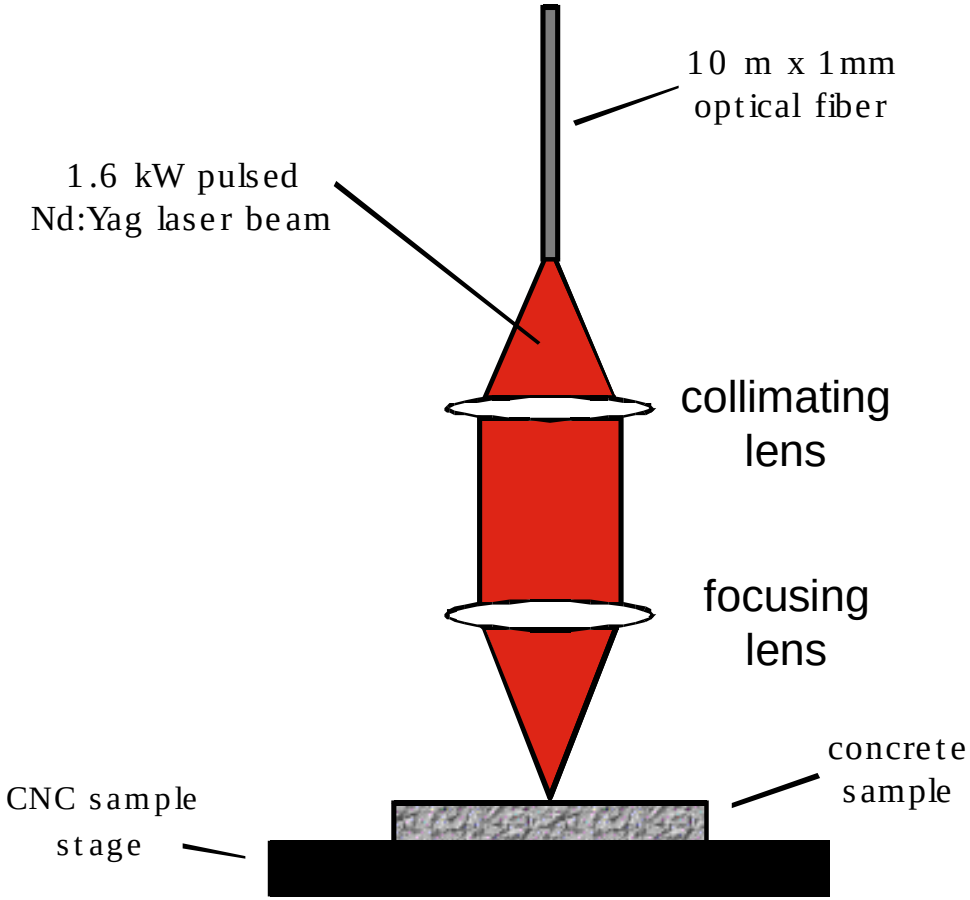
Introduction

- The U.S. Department of Energy owns 23 million cubic meters of concrete in contaminated buildings & facilities
- Much of the radionuclide contamination from water-borne sources resides on the surface or in the near-surface region ($\leq$ 3 mm)
- Many facilities need characterization for D&D planning and for validation

Goals

- To develop the scientific understanding and baseline data necessary for designing laser-based decontamination and on-line characterization methods
- To study the chemistry and distribution of contaminants in the surfaces and waste streams

Laser Ablation Schematic

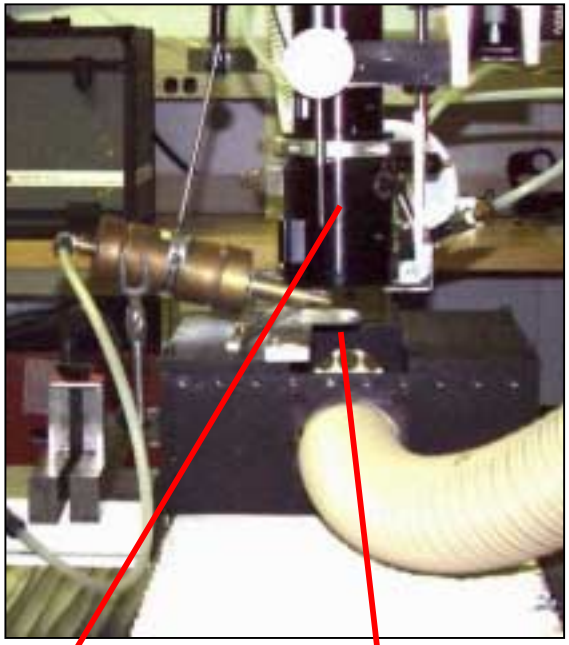


Potential advantages:

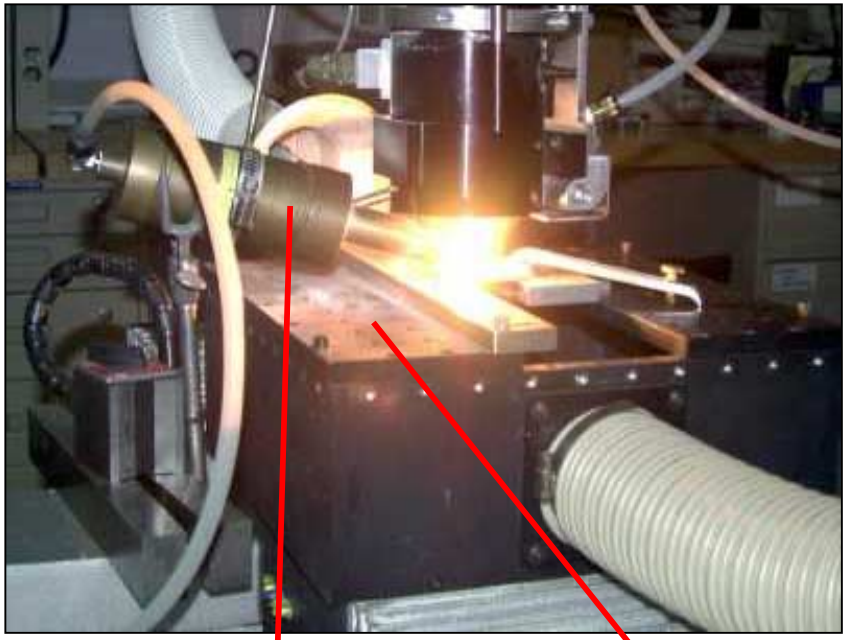
- No added waste
- Remote beam delivery
- On-line instrumentation
- Fine control over material removal
- Can work in corners, on cracked or irregular surfaces

Laser Ablation System

- 1.6 kW Electrox Nd:YAG laser
  - 0.5-10 ms pulse length
  - up to 32 kW peak power
  - 0-800 Hz repetition rate
- Fiber optic beam delivery (1 mm x 10 m cable)
- High efficiency vacuum shroud aerosol containment system (not shown)



Lens system holder Sample



Aerosol particle analyzer CNC sample stage

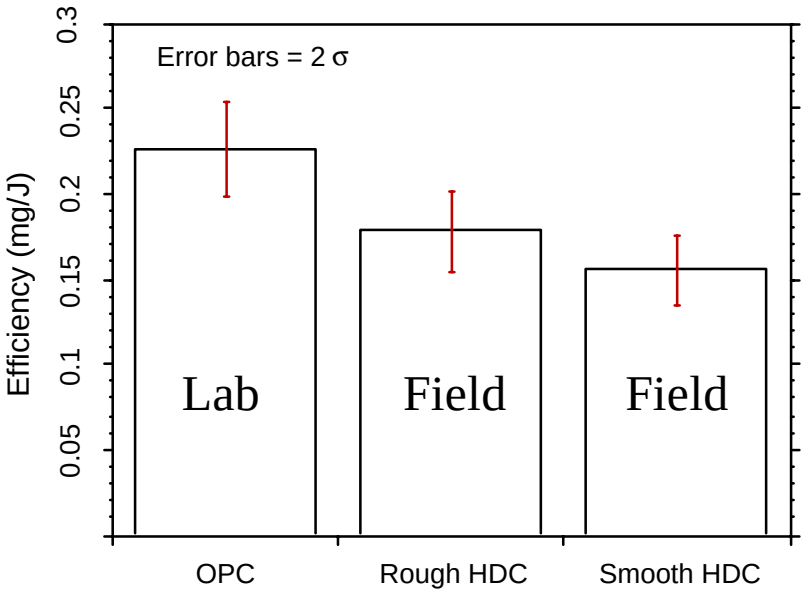
Ablated Cement and Concrete Samples

Lab sample:
40% Type I Portland cement
60% silica sand

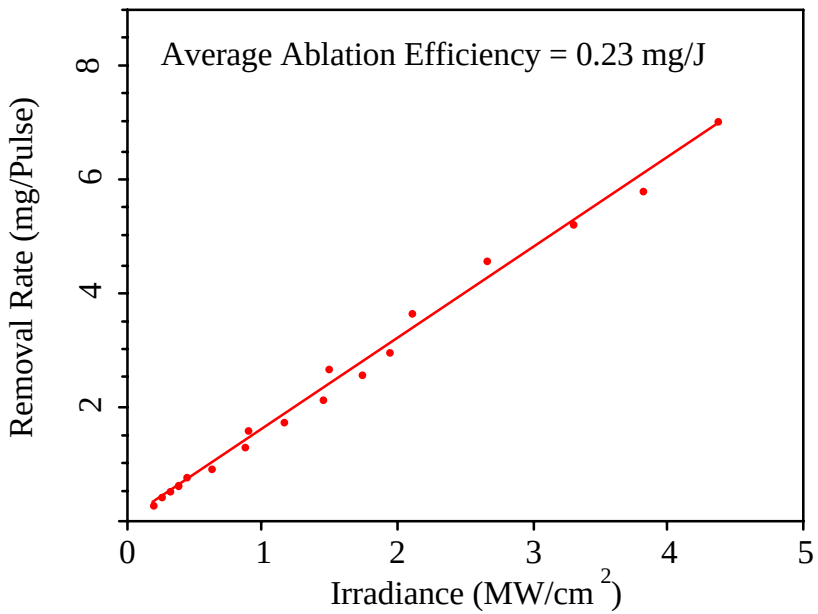
Field sample:
High density concrete
(10-20% cement, balance sand & aggregate)

Pulsed Laser Ablation Efficiency

Lab sample: 60/40 Sand / Ordinary Portland Cement (OPC)
Field sample: High Density Concrete (HDC) from Experimental Boiling Water Reactor at Argonne National Lab (10-20% OPC + sand & rocks)



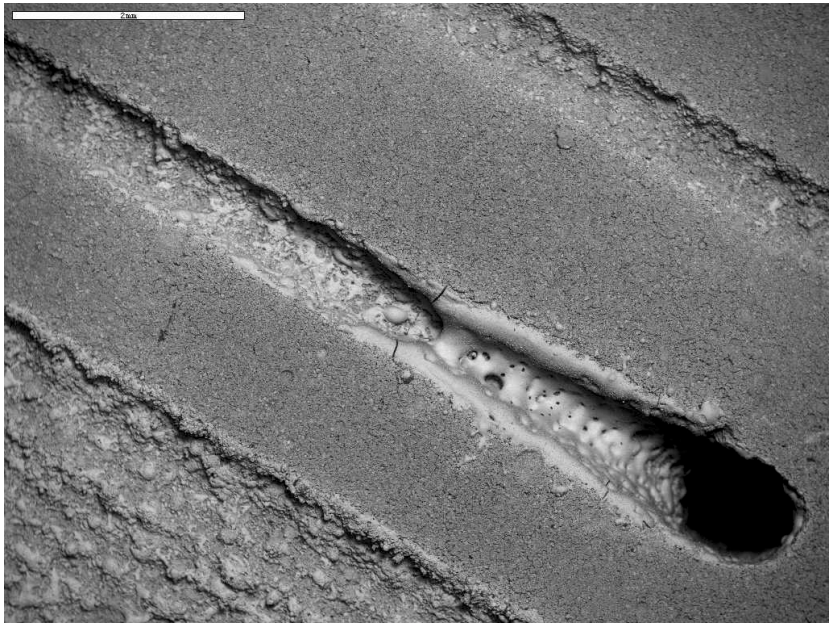
Lab samples ablate more easily than field samples.



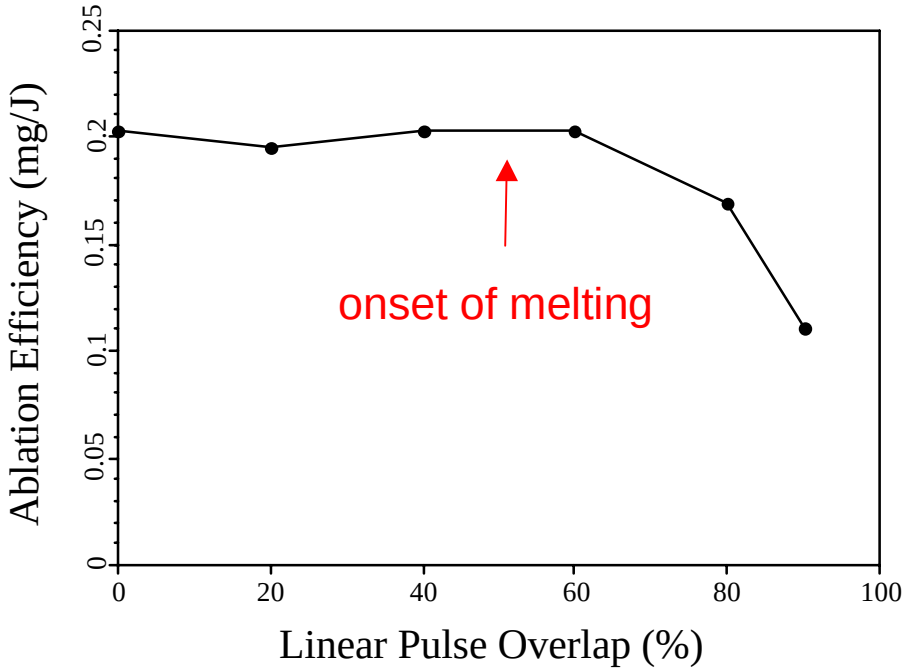
Efficiency is unaffected by irradiance (power density); implies that the ablation mechanism is also unaffected.

Ablation Efficiency vs. Pulse Overlap

|← 2 mm →|

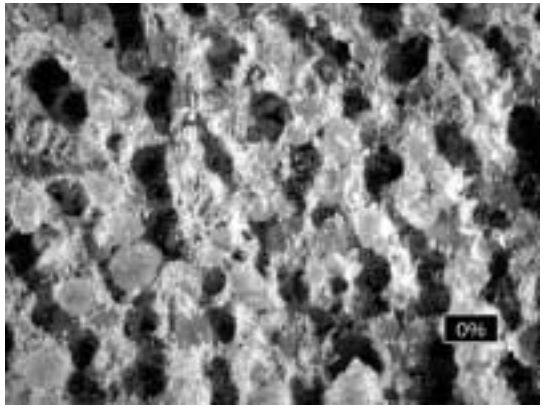


SEM micrograph showing melting of the ablated cement surface as pulse overlap increases and the surface absorbs consecutive laser pulses.

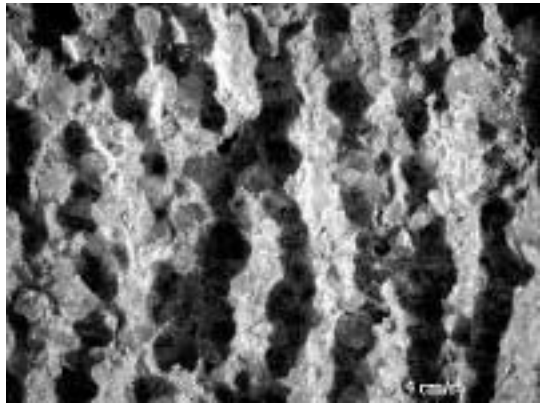


- Most efficient ablation occurs via a thermal shock mechanism.
- Melting harms efficiency by pumping laser energy into the solid/liquid phase transition and allowing stress relaxation.

Concrete Surface After Laser Ablation

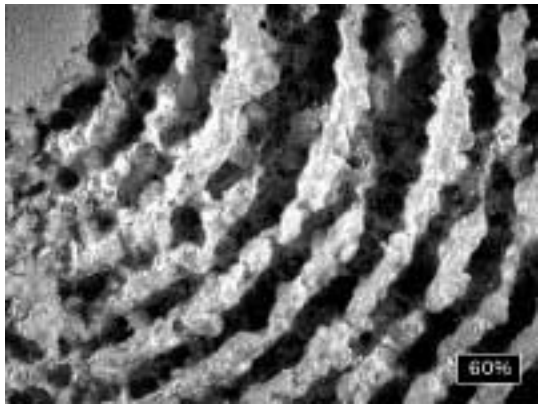


O% pulse overlap

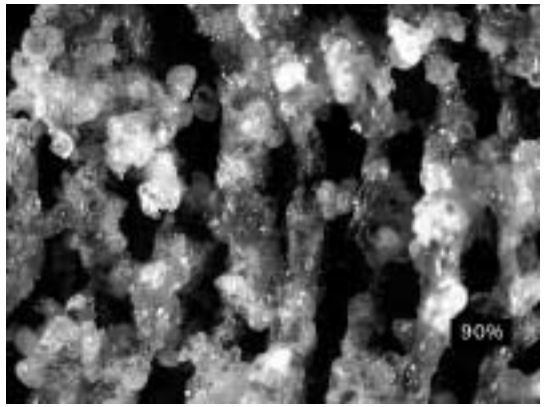


4O% pulse overlap

Up to 60% pulse overlap ablation grooves deepen and little or no melting occurs.



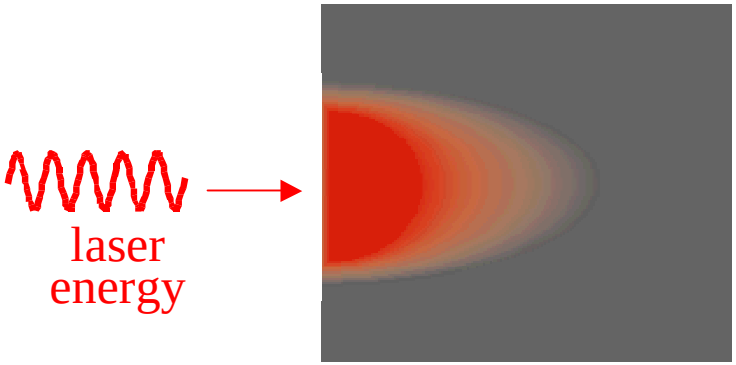
6O% pulse overlap

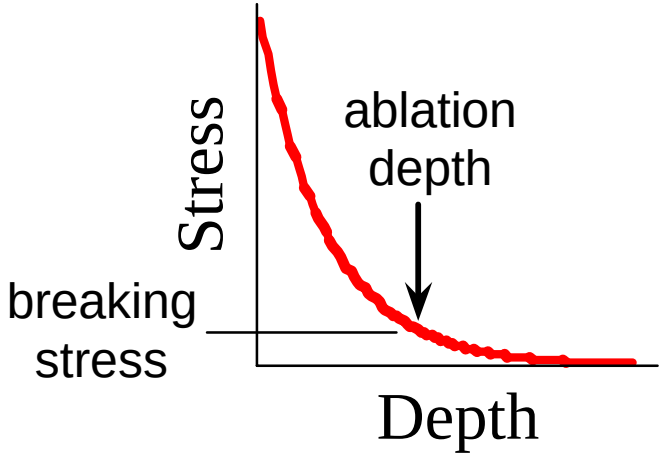


9O% pulse overlap

Above 60% overlap, significant melting leads to reductions in ablation efficiency.

Thermal Shock Ablation Mechanism

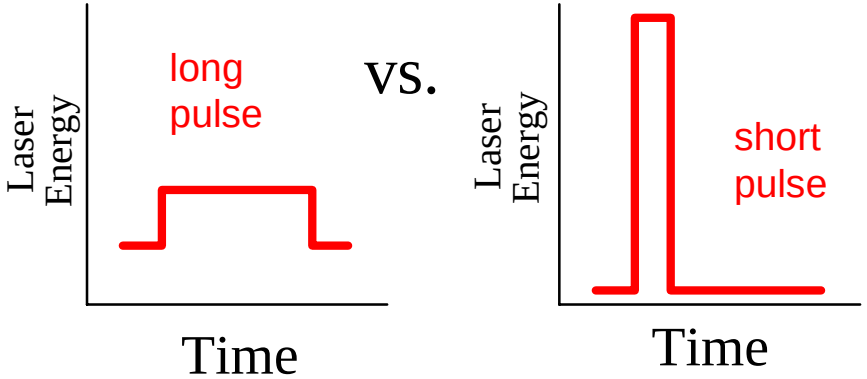




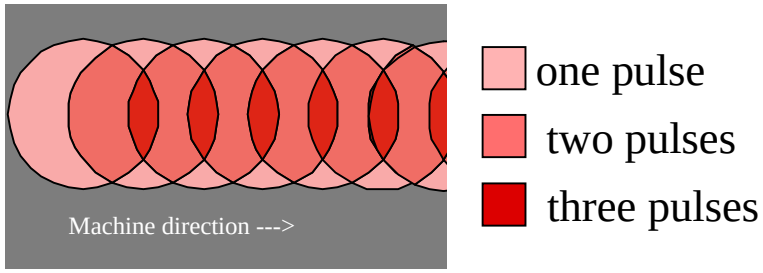
Material ablates when the thermally induced stress exceeds the breaking stress.

Concrete melts when

1) heating rate is low:

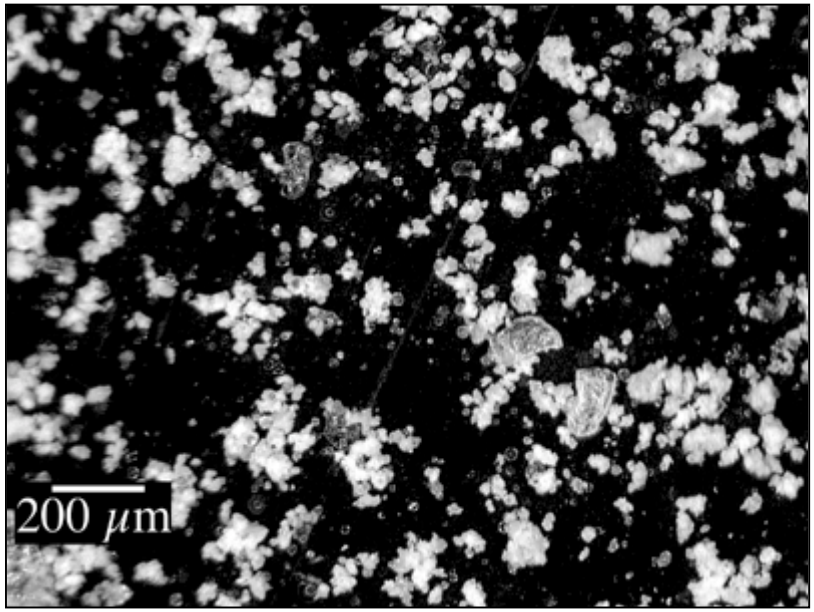


2) pulse overlap is high:

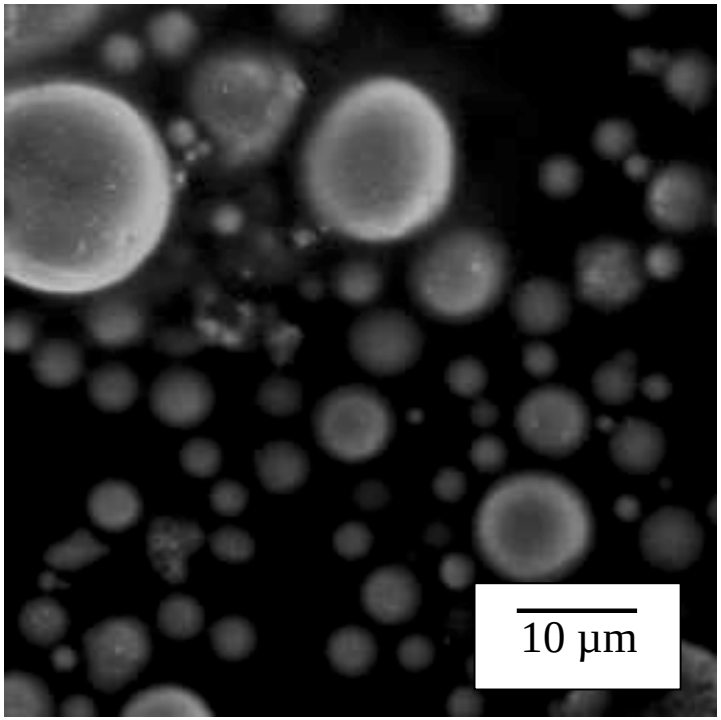


Average exposure = 2.3 pulses @ 60% overlap

Ablation Effluent: Disaggregated Material and Aerosol

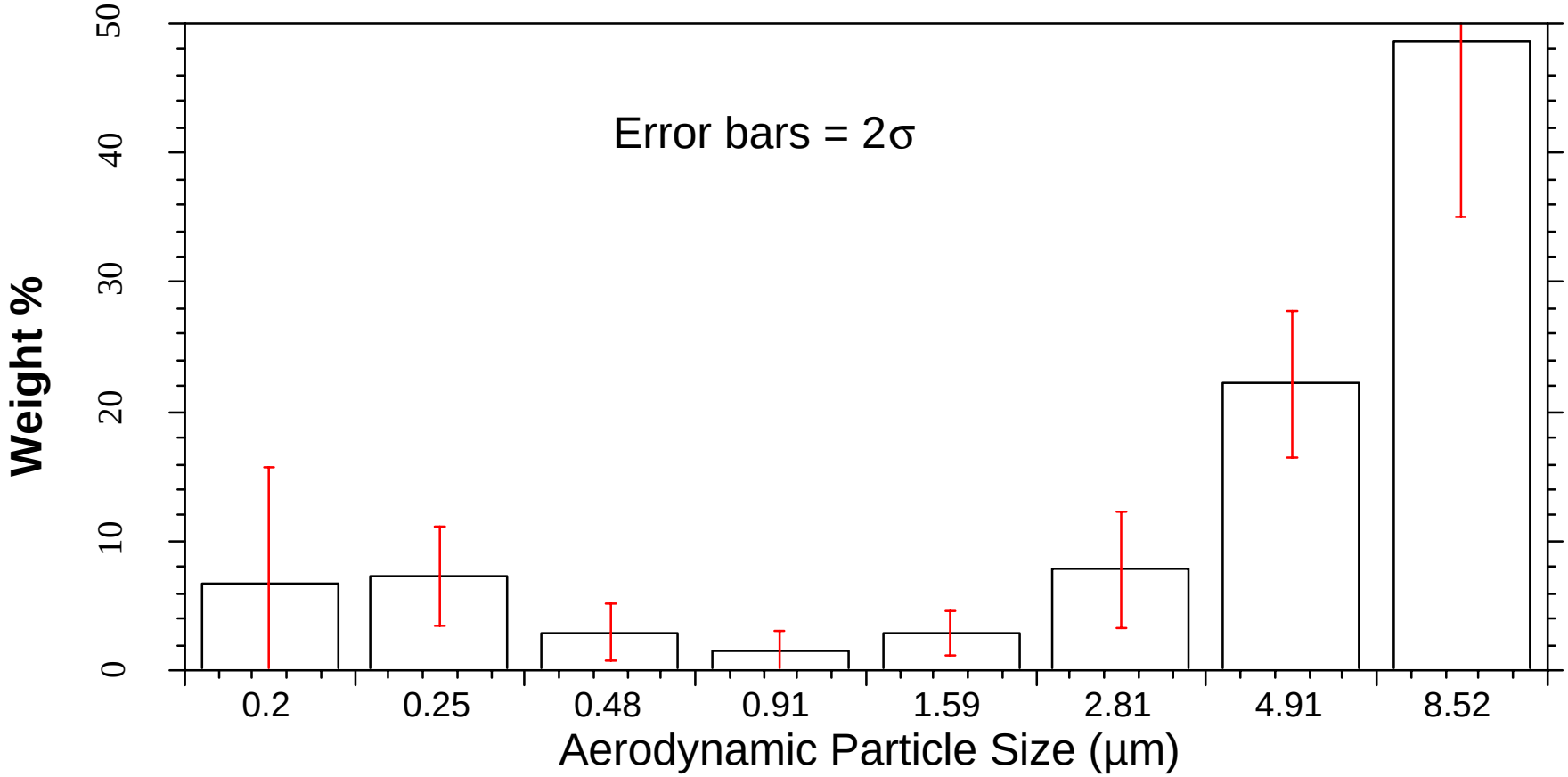


- Most effluent particles consist of shattered sand grains and disaggregated cement matrix material.
- At low overlap the ablation rate is dominated by this mechanism.



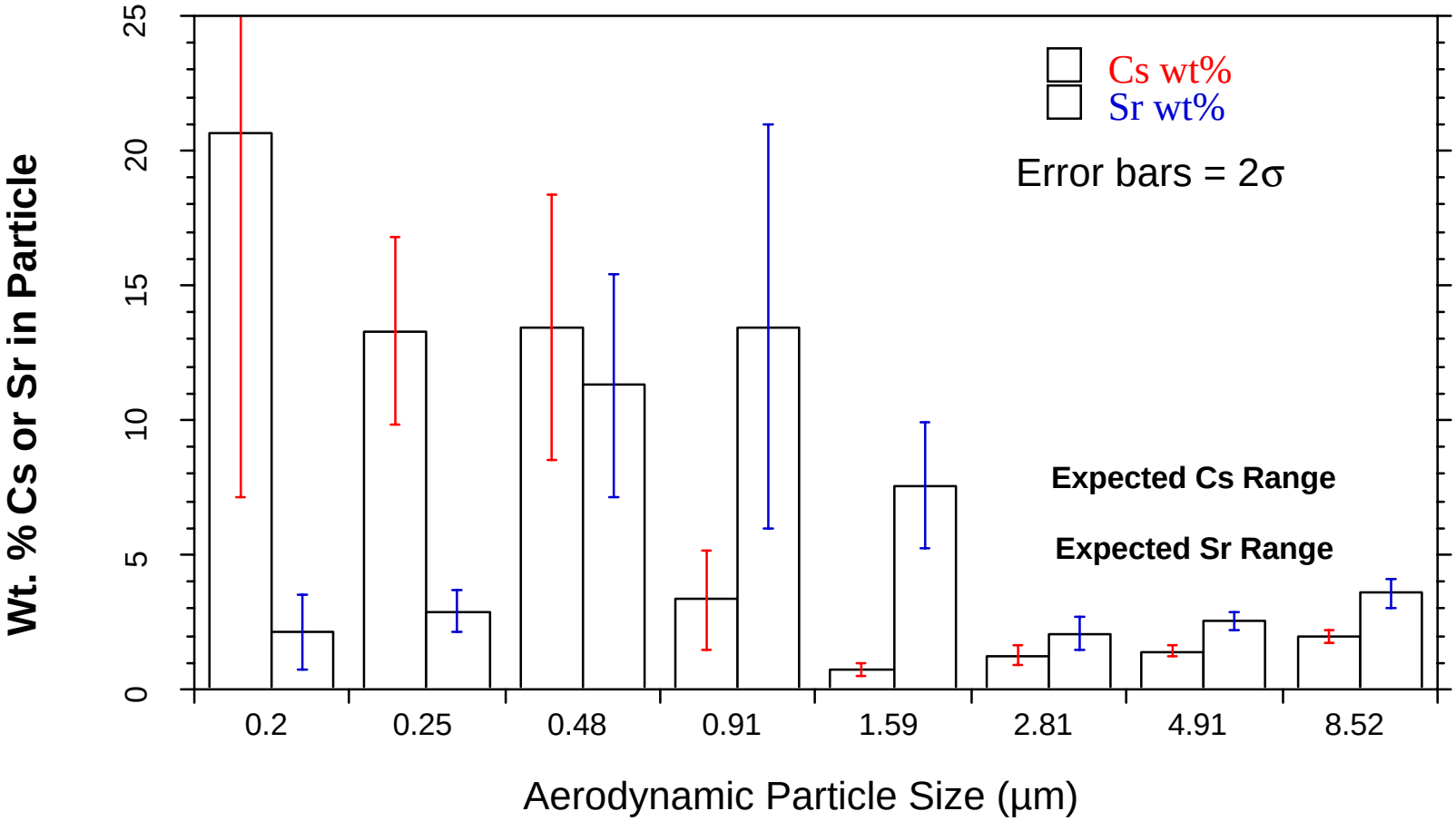
- A few percent of effluent consists of aerosol particles.
- The amount of aerosol increases as pulse overlap increases
- These particles have unusual chemistry.

Cement Aerosol Particle Size Distribution



Bimodal distribution indicates multiple particle formation mechanisms, i.e. vaporization/condensation & melt/spatter

Neutron Activation Analysis of Aerosol Fraction

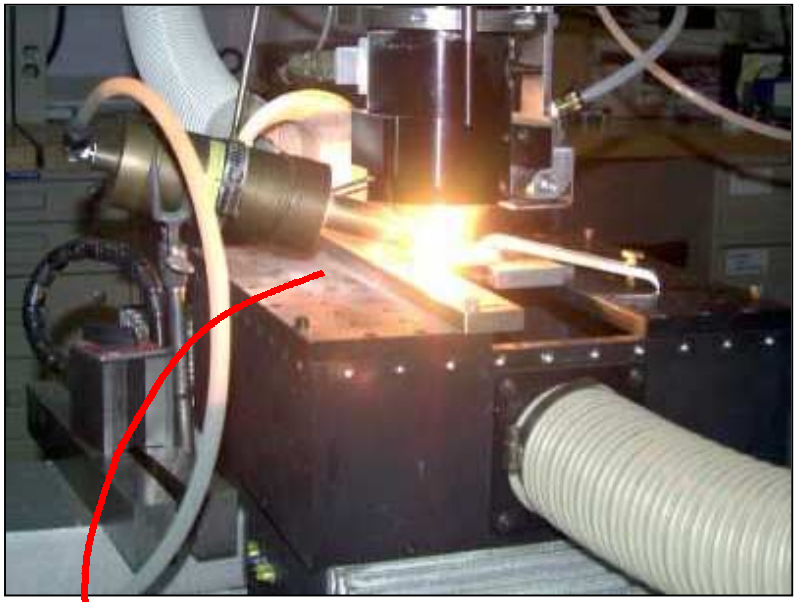


Cesium and strontium segregate into different portions of the PSD

- *Volatile cesium collects in the smallest particles ⇒ vaporization / condensation*
- *Strontium collects in intermediate-sized particles ⇒ ?*

Optical Emission Spectroscopy of Ablation Plume

Laser-Induced Breakdown Spectroscopy (LIBS)



fiber optic cable

spectrometer

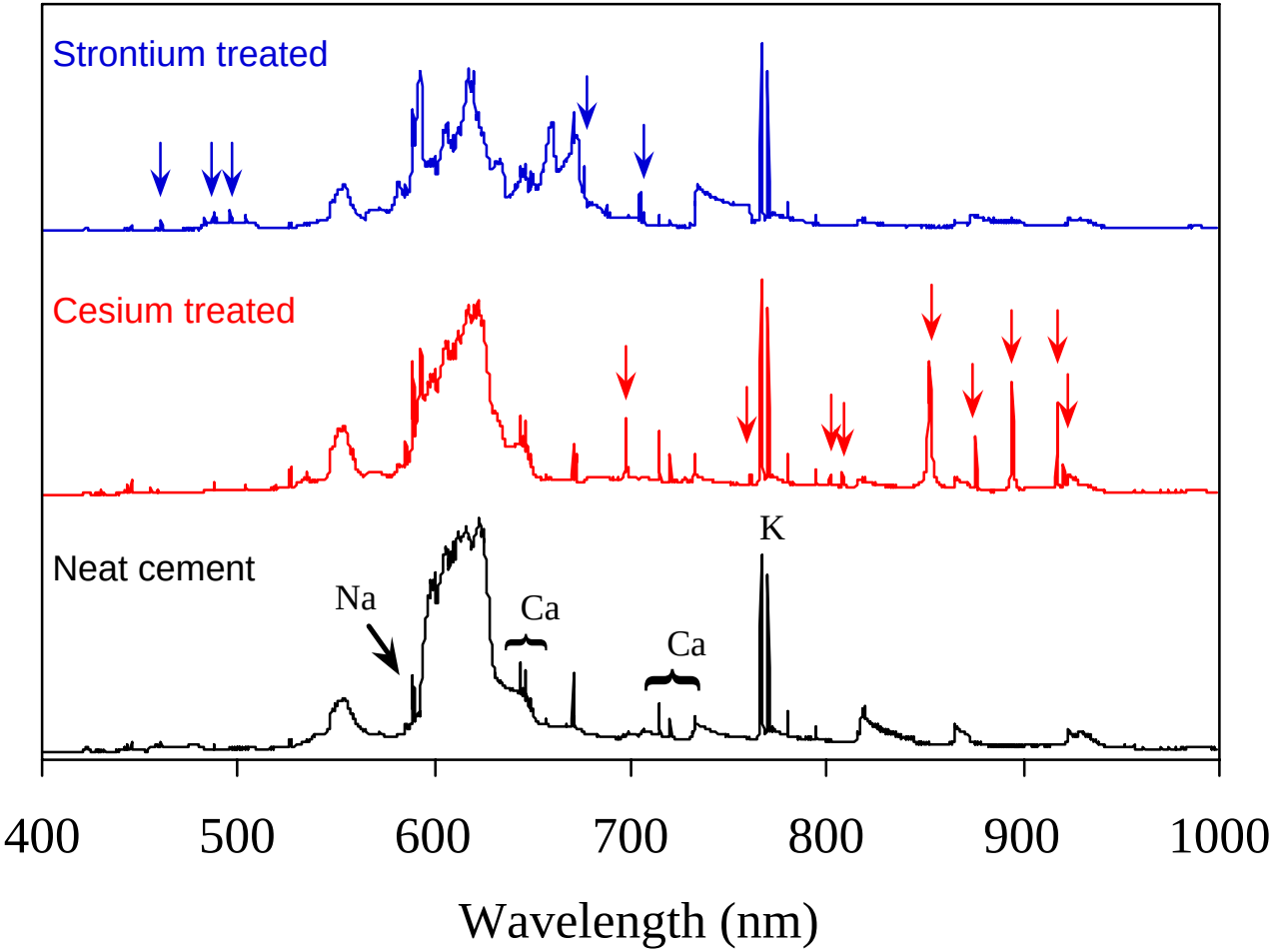
Unique features of this system:

- Large emission volume
  mg of material vs. ng
- Long-lived pulses
  ms vs. ns
- High duty cycle
  40% vs. $\leq 10^{-3}$%

Spectrograph: computer-controlled 0.5 m ARC spectrometer with 16-bit CCD detector and 5 m x 0.5 mm fiber optic cable - *no collection optics or time gating*

Optical Emission Spectra of the Ablation Plume

Sample: 60/40 sand/cement with 1 atom% dopant
Emission collection aperture: 0.5 mm

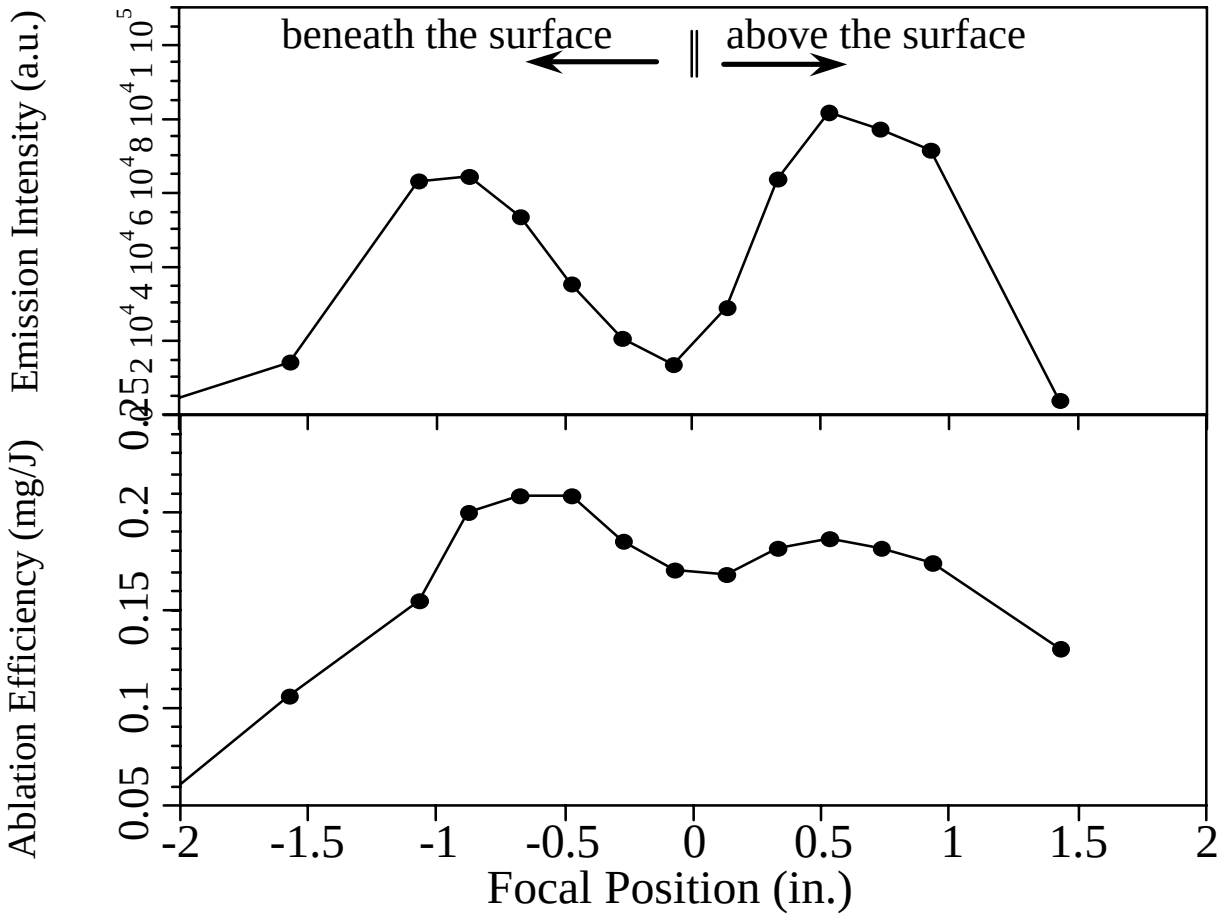


- No continuum background
- All lines are due to neutrals
- Volatile species dominate
- No emission from high-lying (> ~40,000 cm^{-1}) emissive states (e.g. Si, Cl)

Assigned Peaks:

Ca I (25)
Sr I (18)
Cs I (14)
K I (2)
Na I (2)

LIBS as a Diagnostic for Laser Ablation



Emission taken from the brightest part of the spectrum (600-630 nm) correlates well with ablation efficiency.

Conclusions

- Efficient high power laser ablation of concrete proceeds via a thermal shock mechanism
- High power laser ablation of concrete produces several different types of effluent including fractured particles, disaggregated material, and aerosol particles
- Bimodal PSD and fractionation of Cs and Sr within the aerosol fraction indicate multiple particle formation mechanisms
- Optical emission spectroscopy (LIBS) of the ablation plume is simple with this laser system, shows mostly volatile elements, and can be used as a real-time ablation diagnostic tool