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LASER ABLATION STUDIES OF CONCRETE*

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Laser Ablation Studies of Concrete

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Abstract

Laser ablation was studied as a means of removing radioactive contaminants from the surface and near-surface regions of concrete. We present the results of ablation tests on cement and concrete samples using a 1.6 kW pulsed Nd:YAG laser with fiber optic beam delivery. The laser-surface interaction was studied using cement and high density concrete as targets. Ablation efficiency and material removal rates were determined as functions of irradiance and pulse overlap. Doped samples were also ablated to determine the efficiency with which surface contaminants were removed and captured in the effluent. The results show that the cement phase of the material melts and vaporizes, but the aggregate portion (sand and rock) fragments. The effluent consists of both micron-size aerosol particles and chunks of fragmented aggregate material. Laser-induced optical emission spectroscopy was used to analyze the surface during ablation. Analysis of the effluent showed that contaminants such as cesium and strontium were strongly segregated into different regions of the particle size distribution of the aerosol.

Introduction

The U.S. Department of Energy's nuclear weapons complex, a nation-wide system of facilities for research and production of nuclear materials and weapons, contains large amounts of radioactively contaminated concrete. This material is part of the legacy of the Cold War, and must be disposed of prior to the decommissioning of the various sites. In many cases the radiation levels emitted by these materials will remain dangerous for thousands of years, so that after the facility has been decommissioned the material from it must be stored and monitored over a very long period. Often the radioactive contaminants in concrete occupy only the surface and near-surface (~3-6 mm deep) regions of the material. Since many of the structures such as walls and floors are 30 cm or more thick, it makes environmental and economic sense to try to remove and store only the thin contaminated layer rather than to treat the entire structure as nuclear waste. Current mechanical removal methods, known as scabbling, are slow and labor intensive, suffer from dust control problems, and expose workers to radiation fields. Improved removal methods are thus in demand.

Prior to decontamination, one must characterize the surface to determine the types and amounts of contaminants present in order to decide on an appropriate cleaning strategy. Current methods involve scanning the surface with a generic radiation sensor to determine whether or not a particular structure has a problem, then removing core samples from many points on the surface and having more sophisticated analyses performed off-site to determine the type and extent of contamination. Thus a system capable of on-line analysis is valuable since operators can determine the type of contaminants in real time and make more efficient use of the costly sampling and characterization process. Likewise, the removed waste itself must be analyzed to insure that proper storage and monitoring techniques are used.

Laser ablation has previously been investigated laser as a means of concrete surface removal[1-6]. Lasers are attractive since the power can be delivered remotely via articulated mirrors or fiber optic cables and the ablation head can be manipulated by robots, thus avoiding exposing workers and the laser system to the radiation field. In addition, lasers can be instrumented with emission spectrometers or effluent sampling devices to provide on-line analysis. In contrast to mechanical scabbling systems, laser beams can penetrate cracks or follow very rough or irregularly shaped surfaces. Finally, a laser ablation system produces the smallest possible waste stream since no cleaning agents such as detergents or grit (from grit blasting systems) are mixed with the effluent.

Multi-kilowatt Nd:YAG and CO_2 lasers are capable of ablating concrete surfaces and effecting decontamination. Both cw[1,2] and pulsed[3,4] systems have been investigated, with the main difference lying in the mechanism of ablation. Pulsed systems rely on the shock wave produced in a small volume of rapidly heated material to disaggregate and explosively remove concrete, while cw systems rely on the differential thermal expansion of the various components of concrete (i.e. cement, sand, and aggregate) to induce thermal stress in a larger volume, which results in the fracture and removal of material.

In this paper we describe the ablation of cement and concrete by a 1.6 kW pulsed Nd:YAG laser. Ablation efficiency and material removal rates were determined as functions of irradiance and pulse overlap. The ablated surfaces and effluent were analyzed to determine ablation mechanisms and optimize the process. Doped samples were ablated to determine the efficiency with which surface contaminants are removed and captured in the effluent. The results show that while the majority of the concrete ablates by shock-wave induced fracture and disaggregation, some of the cement phase of the concrete melts and vaporizes and produces a fine aerosol. Thus the effluent consists of both micron-size cement aerosol particles and larger chunks of fragmented aggregate material. Laser-induced breakdown spectroscopy was used to analyze the material during ablation.

Experimental

Laser ablation experiments were conducted with the 1064 nm fundamental of an Electrox 1.6 kW pulsed Nd:YAG laser. The system produced pulses of 0.5 to 1 ms duration with a nearly square temporal profile. The pulse energies ranged up to 16 J depending on the operating mode and repetition rate, which ranged as high as 800 Hz. The beam was delivered via a 0.5 mm diameter fiber optic cable 10 m in length. Focal plane spot diameters ranged from 0.55 to 0.96 mm. The sample stage was moved in three dimensions under the stationary beam. A modest gas flow was maintained over the focussing lens to keep it free of debris.

Two types of samples were ablated. The first consisted of a 60/40 (wt/wt) mixture of Type I Portland cement cast at a water/cement ratio of 0.5 and allowed to cure for at least thirty days prior to use. Some of these were doped with 0.01 atom fraction of non-radioactive cesium and strontium to simulate some of the more common contaminated concrete found in nuclear facilities. The second type of sample was non-contaminated high density concrete from the Experimental Boiling Water Reactor at Argonne National Laboratory.

Aerosol particle size distribution analysis was done with a seven stage particle impactor. Ablated surfaces and effluent were examined with optical microscopy. Optical spectroscopy of the ablation plume was done with a 0.5 m monochromator equipped with a CCD detector and fiber optic collection. Spectra were accumulated without time gating.

Results

Ablation efficiency, defined here as the mass of material removed per unit energy delivered, was determined under varying conditions. Figure 1 shows the effect of pulse overlap, defined as the linear overlap distance of two consecutive pulses. These experiments were done at 800 Hz with 0.5 ms pulses, with the beam focused to 0.55 mm at the sample surface. Ablation efficiency was constant at about 0.2 mg/J for overlaps from 0 to 60%, but dropped as the overlap increased beyond 60%. As implied by Figure 1, the width and depth of the ablated groove increased as the overlap increased up to about 60% (see below), at which point the increase in groove volume no longer compensated for the increase in overlap and led to the decline in ablation efficiency. Figure 2 is a photograph showing regions of the sample ablated at 0, 60, 80, and 90%. The grooves in the 60% overlap region are clearly deeper than those in the 0% overlap region. Up to 60% overlap

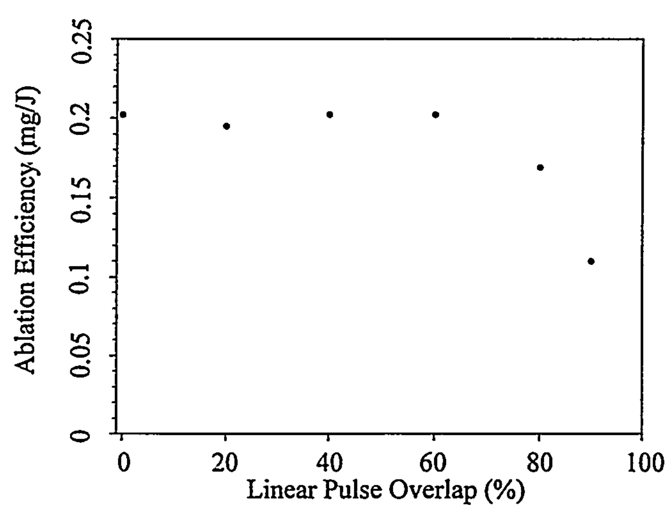

Figure 1: Concrete ablation efficiency as a function of pulse overlap.

the surface shows no significant melting, but at 80% bits of melted material are noted and at 90% a great deal of melting is evident.

Figures 3-5 are optical micrographs of the 0, 60, and 90% overlap cases. The major difference between the 0 and 60% cases is that the grooves are deeper at 60% overlap. The holes in the sample surface in Figure 3 are vacancies left behind after the removal of individual sand grains. Figure 4 shows more uniformly deep grooves, with the onset of melting evident from the rounded groove edges. At 90% overlap (Figure 5) the ablated surface is glazed over with melted material. The glazing reduces the ablation efficiency in several ways. First, laser energy that would otherwise have been used to remove material is instead wasted in melting the sample.

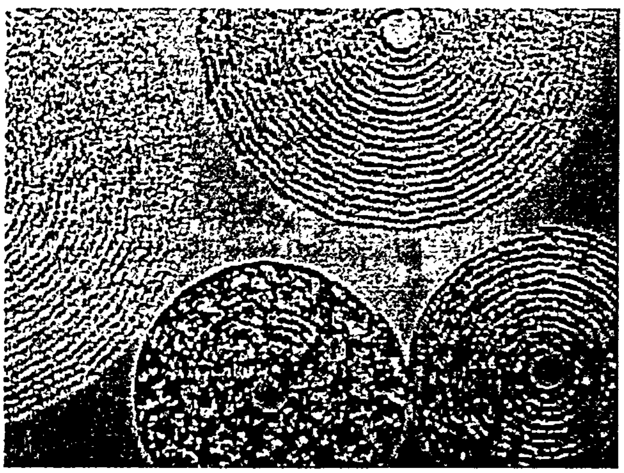

Figure 2: Photograph of an ablated sample of 60/40 sand/cement. The ablated regions had pulse overlaps of (clockwise from left) 0, 60, 80, and 90%. The ablation grooves are ~0.5 mm wide.

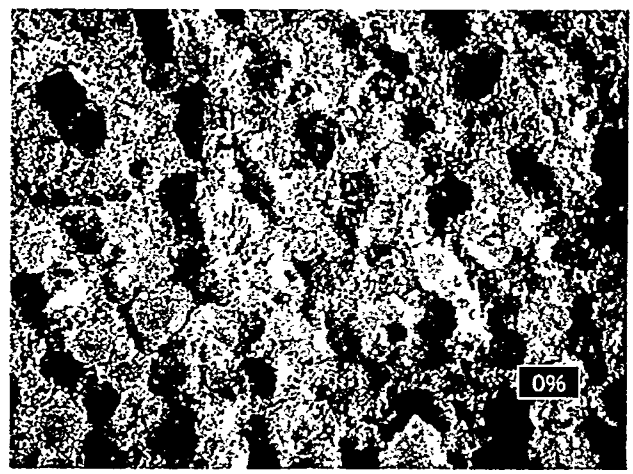

Figure 3: Optical micrograph of the 0% pulse overlap ablation region of Figure 2. The field of view is ~3x4 mm.

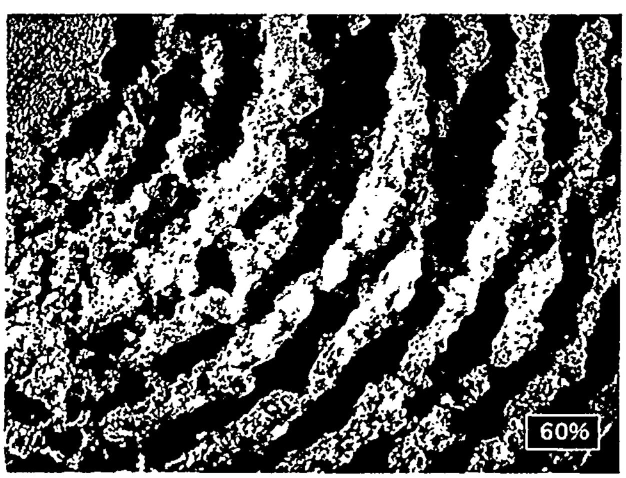

Figure 4: Optical micrograph of the 60% pulse overlap ablation region of Figure 2. The field of view is ~4x5.5 mm.

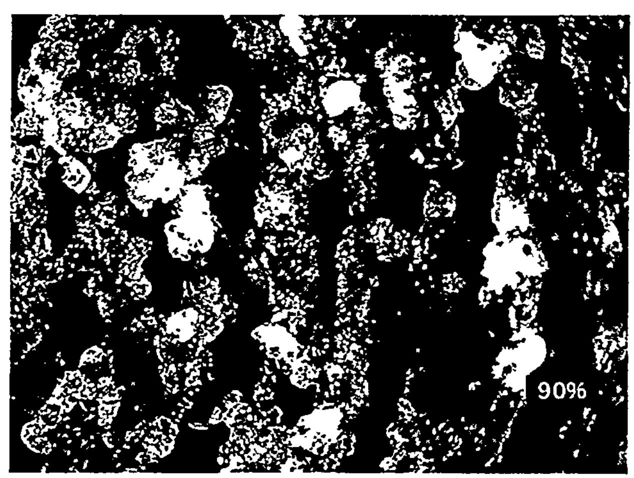

Figure 5: Optical micrograph of the 90% pulse overlap ablation region of Figure 2. The field of view is ~3x4 mm..

Ablation at low overlap is more efficient since it is dominated by a shock mechanism which fractures and dislocates chunks of material, and no energy-intensive phase transition is involved. Second, the glazed surface is more reflective, resulting in reduced absorption of the laser energy in the overlapped region. Finally, residual heat in the substrate reduces the thermal gradient induced by the overlapping pulse, thereby reducing the magnitude of the shock wave produced in the material. This effect is probably minor compared to the other two since, to first order, it should be apparent at overlaps less than 60%. That fact that it is not implies that all or nearly all of the heated volume is removed at overlaps less than 60%. Figures 3 and 4 show that there is very little heat affected zone at pulse overlaps less than 60%.

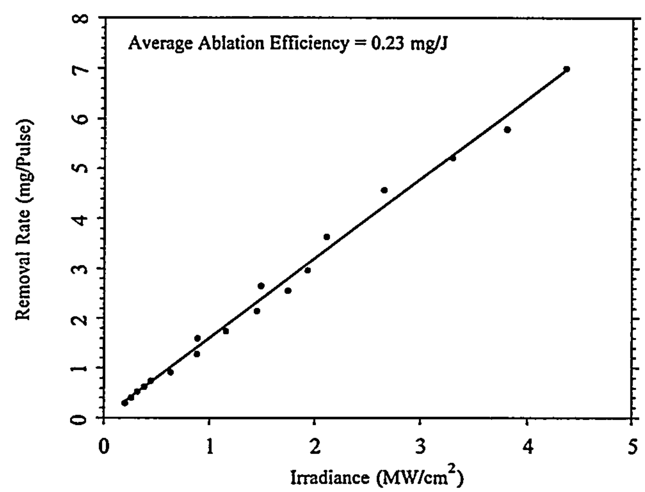

Figure 6: Concrete removal rate as a function of irradiance.

Figure 6 shows the effect of increasing irradiance on the ablation rate. The sample was ablated with either 0.5 of 1 ms pulses at a spot size of 0.95 mm and pulse overlap of 0%. The beam focal plane was at the surface of the sample. The irradiance was varied by changing the pulse peak power from 1.4 to 30.2 kW. The repetition rate was either 50, 100, or 200 Hz depending on the peak power. The relationship is linear, indicating that ablation efficiency is constant over this range of irradiances. The slope of a linear fit to the data yields (with appropriate unit conversion) an ablation efficiency of 0.23 mg/J. From this value the concrete removal rate for any pulsed Nd:YAG with a similar temporal pulse profile can be calculated directly from the average laser power. For example, with the beam focused on the sample surface, our 1.6 kW system is capable of a maximum ablation rate of 370 mg/s. The caveat that the beam be focused on the surface is necessary because changing the focal position with respect to the surface can affect the ablation efficiency[7].

The efficiency value of 0.23 mg/J is higher than that obtained in the overlap data of Figure 1. This is most likely due to differences in the samples and day to day differences in laser performance. The data of Figure 6 were obtained from three different samples on three different days, and therefore constitute a more representative sampling than do the data of Figure 1.

Samples of high density concrete ablated with lower efficiency than did the laboratory samples of 60/40 sand/cement. Efficiencies obtained from high density concrete were 0.16 to 0.18 mg/J depending on the surface smoothness of the sample. The lower overall values are probably due to the presence of large aggregate particles (i.e. rocks) which tended to melt more readily than sand or cement. These findings are preliminary, coming from ablation runs with relatively high overlap (~50%), and suggest that for high density concrete the overlap must be kept lower than for the laboratory samples. We are currently investigating this further.

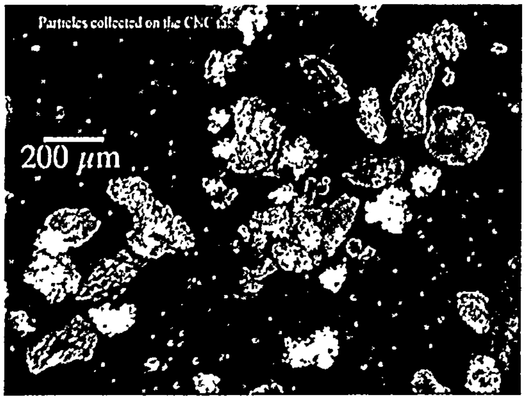

Figure 7: Optical micrograph of the effluent from concrete ablation.

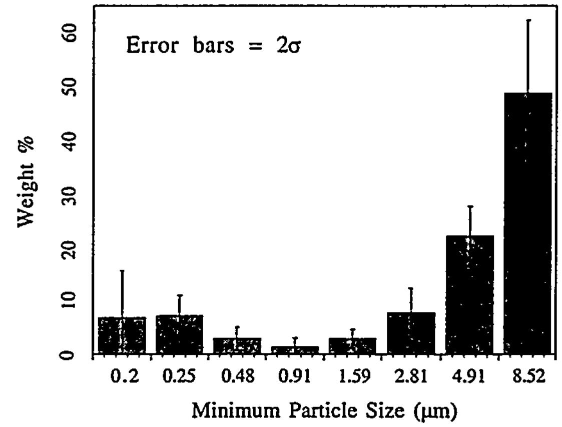

Figure 8: Aerodynamic particle size distribution of the aerosol portion of the concrete ablation effluent.

Our previous studies showed that cement and aggregate behave differently under high power pulsed irradiation[4]. Sand particles, which are generally on the order the same size as the focused beam, i.e. about 0.5-1 mm, tend to fracture or dislodge without melting. Figure 7 shows a portion of the effluent from the ablation of a 60/40 cement/sand sample. The larger, smoother particles are fractured sand grains. The bulk of the stored nuclear waste from an ablation process would take this form, that is small, relatively unprocessed particles. Cement, which is a grainy material composed of particles up to a few tens of microns in size, reacts to the optical energy in several ways. While most of it tends to disaggregate into grainy clumps such as those seen in Figure 7, some of it melts or vaporizes. Much of the melted material spatters off the surface and forms aerosol particles along with the condensate from the vaporized material. Though the aerosol portion makes up a small fraction of the total effluent, is important for several reasons. It is derived from the cement portion of the concrete, which is the phase that contains the radioactive contaminants, and is therefore a concentrated form of the contaminated concrete. Second, it constitutes the worst health hazard generated from the process, because the fine particles remain airborne much longer than the larger grains and represent a greater inhalation hazard. Any vacuum shroud/filtration system must be designed to efficiently collect these particles. Finally, the aerosol phase and the vapor from which it is derived are the most amenable portions of the effluent for sampling and on-line analysis. Figure 8 shows the aerosol aerodynamic particle size distribution obtained from the impactor. The distribution is bimodal, with a minimum at about one micron. Previous studies[4] have shown that the smaller particles are preferentially enriched in aluminum, which is a minor constituent of Portland cement[8]. Together with the bimodal size distribution, this suggests that the smaller particles are formed primarily from the vapor phase (nucleated by an aluminum-rich chemical phase) and the larger particles, many of which are hollow, are formed primarily from the melt/spatter process.

Figure 9 shows emission spectra of the bright plume generated during the emission process. The three spectra were obtained from either the neat 60/40 sand/cement samples or from samples doped with 1 atom% of either cesium or strontium to simulate contaminated concrete. The spectra were obtained by simply positioning the tip of the spectrometer's fiber optic collection cable near the sample and opening the shutter prior to ablating a line across the sample. Typical acquisition times were on the order of a few tens of milliseconds, encompassing up to a few tens of laser pulses depending on the pulse repetition rate. All samples show emission from sodium, calcium, and potassium, which are native to the cement, plus other unidentified lines, most notably broad features around 550 to 650 nm which are probably due to molecular species. The doped samples also show a number of lines easily assigned to cesium and strontium. In all, 25 calcium, 2 potassium, 2 sodium, 18 strontium and 14 cesium lines have been identified, all of which originate from relatively low-lying states (less than about 40,000 cm^{-1}) of the neutral species. The fact that the spectra are dominated by low-lying neutral states of the more volatile elements implies that the plasma is relatively cool. These data were obtained at low irradiance and 0% pulse overlap. These conditions suppress substrate heating and should lead to little heating of the plasma by the beam. Thus it appears that analysis and cleaning are somewhat at odds, since Figure 1 shows that the high overlap that is presumably necessary to generate large quantities of molten, and therefore also vaporized, material is detrimental to ablation efficiency. We are currently investigating means of producing hotter plasmas without high overlap, i.e. by increasing the irradiance. Nonetheless, the simplicity and ease with which spectra can be obtained augers well for the incorporation of an on-line analysis system into the pulsed laser ablation system.

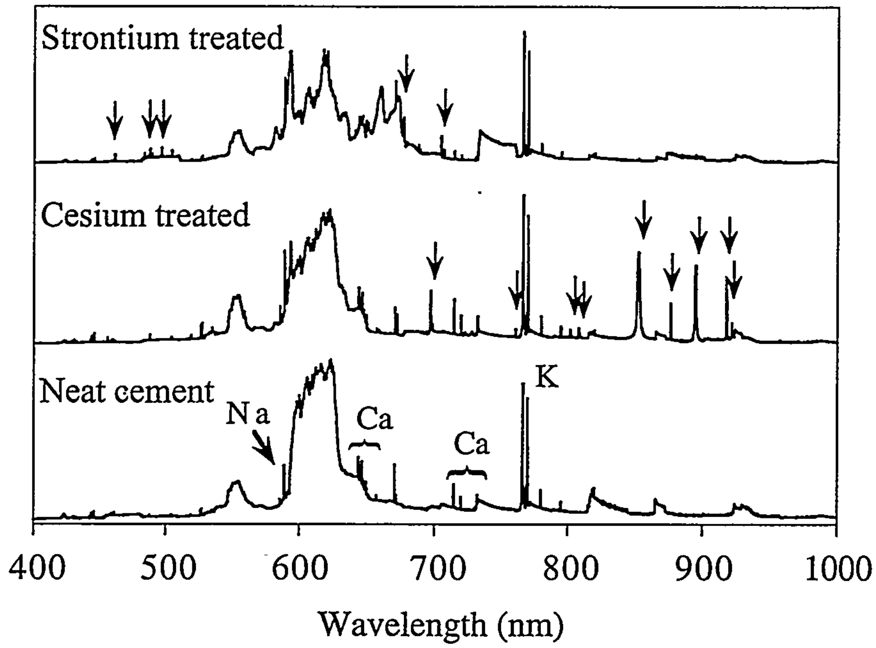

Figure 9: Optical emission spectra of neat, cesium- and strontium-doped samples of 60/40 sand/cement. The arrows indicate some of the cesium and strontium lines.

Acknowledgements

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