

Applications of Lasers to the Solution of Environmental Problems

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Abstract : This presentation will focus on current work in the Ames Laboratory where laser ablation is being used for both analytical sampling and metal surface cleaning. Examples will be presented demonstrating the utility of optical spectroscopy for monitoring laser ablation processes.

1. Introduction

The public in the United States and, generally, throughout the world is greatly concerned with environmental contamination. This concern, which grew rapidly throughout the 1960's and afterwards, has resulted in extensive regulation of industries that produce hazardous wastes and interest in past releases of such wastes that may have occurred when regulations were more permissive than they are today.

The Ames Laboratory is one of several US Department of Energy (USDOE) laboratories exploiting lasers to understand the impact that USDOE facilities have had on the environment. The USDOE is committed to clean up its former facilities, many of which are radioactively contaminated. The application of conventional technologies during the

clean up could create massive quantities of secondary wastes. Thus, great efforts are being made to develop innovative technologies that minimize secondary wastes and encourage recycling. Such technologies must protect workers and the public from exposure to radiation.

Lasers, which can transport energy to remote locations and can be tuned to wavelengths that permit even isotopic specificity, are uniquely suited to participate in environmental technology development.

This paper will discuss two applications of lasers to USDOE clean up problems. The first example, laser surface cleaning, is the subject of a recent patent application¹. The second study, in which lasers are used to sample materials captured in tree-rings² demonstrates how lasers can be used to study events that occurred long ago but are still of vital interest to the public.

2. Experimental

Information about the lasers referred to in the text of the paper are collected in Table 1.

Table 1. Operating parameters for lasers referred to in paper.

Laser	Excimer Questek Model 2460 vβ	Nd:YAG Continuum Model NY82-30	Nd:YAG US Lasers Model 405Q
Rep. rate (Hz)	150 - 300	30	100 - 10000
Wavelength (nm)	248 (KrF)	1064	1064
Pulse energy (mJ)	250	700, 450*	20 - 45
Pulse width (ns)	25	~8, 150000*	100 - 230

*Laser operated in the "free-running" mode.

2.1 Laser surface decontamination

Ultraviolet wavelengths are expected to be most efficient for laser surface ablation³ but laser surface cleaning efficiency is a complex phenomenon involving laser irradiance, wavelength, and repetition rate. Surface cleaning efficiency has been greatest using the high repetition rate, acousto-optical Q-switched [AOQS], laser described in the last column of Table 1.

Energy from each of the lasers listed in Table 1 could be delivered to surfaces using appropriately coated mirrors and lenses. The AOQS laser could be reliably transmitted using an optical fiber whereas other laser beams, because of wavelength or very short pulse duration, could not.

The material removed from material surfaces could be monitored in real-time by analyzing the light emitted by the plasma created near the metal surface during laser surface cleaning.

2.2 Retrospective environmental analysis by tree-ring assay

Tree cores (~4 mm diameter, variable length) were sanded to reveal the ring pattern. The cores were sampled by laser ablation with the resulting particulates conveyed to a laboratory-constructed, twin-quadrupole inductively coupled plasma — mass spectrometer⁴ for atomization, ionization, and then mass detection. The laser spot size on the sample was ~1 mm in diameter and, due to the non linear distribution of the rings, this spot size produced variable temporal resolution. Laser sampling was done using a Continuum NY82 Nd:YAG laser operating in the free-running mode².

3. Results and Discussion

3.1 Laser surface decontamination

Various metals, ranging from very soft metals, such as lead and aluminum, to very hard alloys, such as stainless steel and Haynes No. 25 [primarily composed of cobalt], have been successfully decontaminated using laser ablation. It has been long known that a pulsed laser can ablate materials from metal surfaces. Converting this knowledge to a useful cleaning technology has required that some misconceptions about

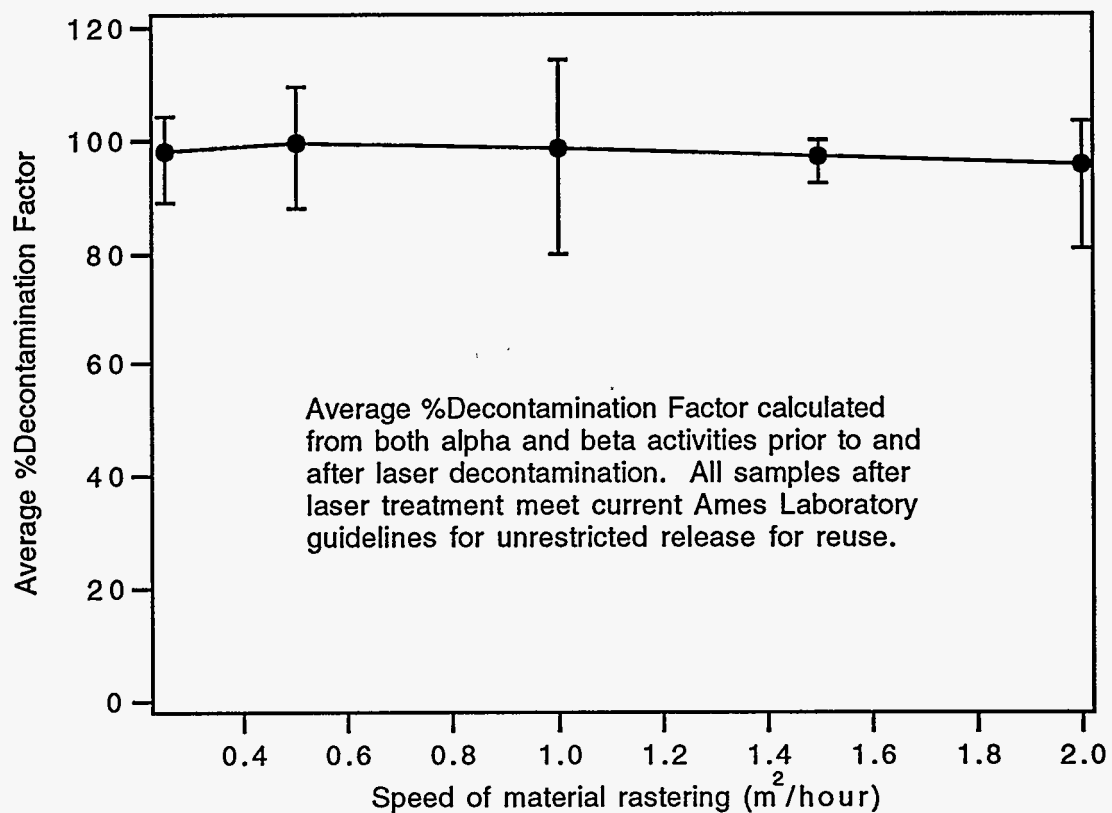


Figure 1. Excimer laser decontamination of aluminum duct contaminated with thorium oxide. The "Average % Decontamination Factor" is defined so that 100% represents zero remaining radioactivity.

lasers be corrected. Modern lasers do not require focusing to a "point" to reach irradiance levels necessary for surface ablation. A modern excimer laser produces an output pulse that exceeds 10^8 W/cm² over an area of 0.12 cm². Given repetition rates achievable with such lasers (~300 Hz), useful surface cleaning rates are possible. Figure 1 demonstrates that radioactively contaminated aluminum ducts can be decontaminated using excimer laser irradiation at rates up to 2 m²/hour

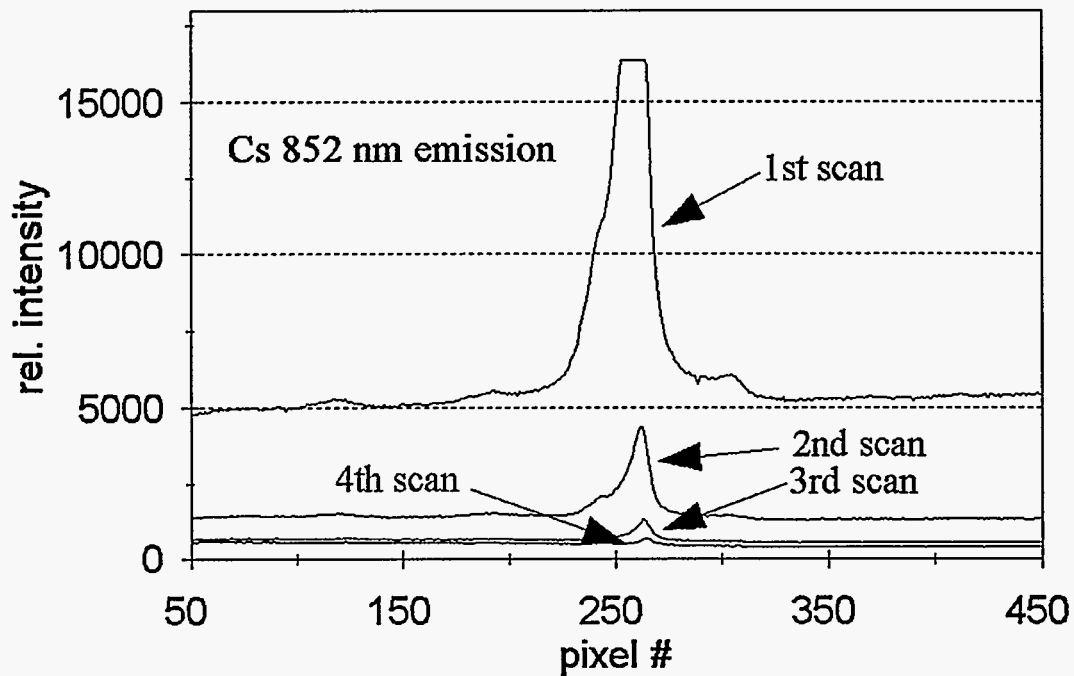


Figure 2. Monitoring the Cs emission signal from the laser-induced plasma produced during Nd:YAG laser surface cleaning. As the surface is cleaned, the Cs signal decreases in magnitude.

and, with improved lasers, this cleaning rate could be increased substantially. This performance is due to the high repetition rate of the excimer laser and to the high energy per pulse from this device. The excimer laser can be focused into a line (~3.5 cm X 0.03 cm) and rapidly swept across the surface.

During the laser decontamination process, a plasma forms at the metal surface that can be used to monitor the progress of decontamination. This is illustrated in Figure 2, where a cesium impurity in a surface coating is monitored during the removal of that coating.

The laser cleaning process has several advantages over competing technologies: the process energy can be delivered remotely via fibers or conventional optics; the laser irradiance can be tailored to suit the cleaning application; no secondary wastes are generated; the technology can be easily automated.

3.2 Retrospective environmental analysis by tree-ring assay

Trees are prodigious consumers of water, with some species taking over 200L/day from the ground through its root system⁵. Dissolved minerals in the ground water may adhere to the xylem walls involved in the conduction of water through the tree and develop a compositional signature that is time stamped within the "tree-rings." Thus, the elemental analysis of tree rings can provide a snap-shot of ground water contamination sampled by the tree during its lifetime.

Cores were taken from trees in the vicinity of "Little Ankeny," a facility on the Iowa State University campus where ~1000 tonnes of ultra-pure uranium was produced in the mid 1940's. The LA-ICP-MS analysis of a core from a sycamore tree located close to the facility is shown in Figure 4. Due to noise events and the relatively low sensitivity of the home-built ICP-MS instrument, signal levels below ~0.006 may be unreliable. This plot shows two U spikes (~1949 and ~1958) that may be related to U releases during operation (~1943-1945), the dismantlement of Little Ankeny (1953), or the construction of an addition to a nearby building (1962). An environmental model where U in soil becomes disturbed during site activity to become available for tree uptake can

explain the U peaks. The discrepancy between the above dates and the U peaks may be due to transannular migration within the tree trunk, which

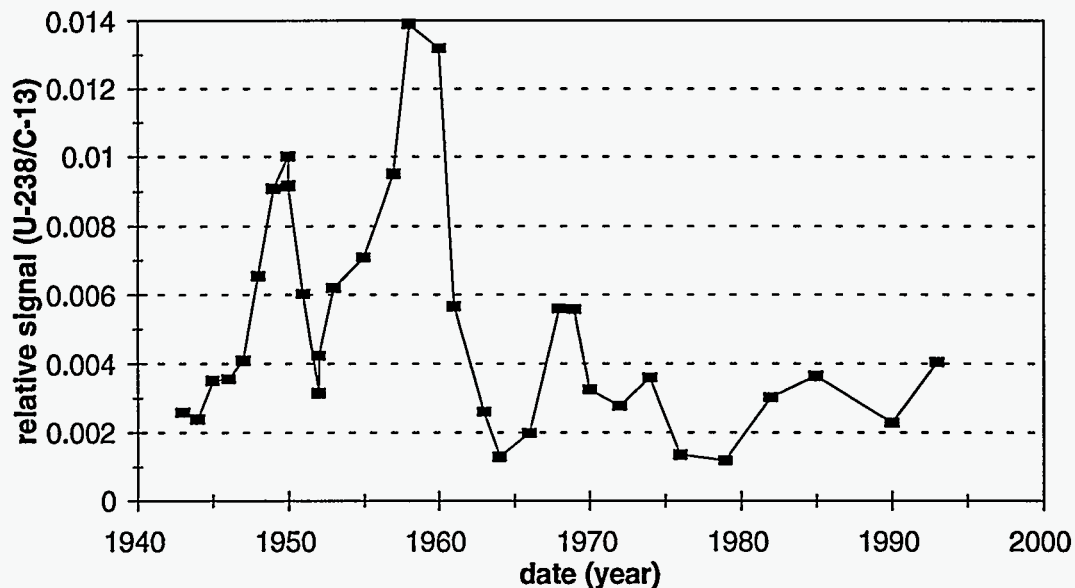


Figure 4. Laser ablation ICP-MS determination of U in core from tree close to the Little Ankeny site on the ISU campus. Solution assays of this tree showed average U concentration of 125-250 ppb in the wood.

has been noted to limit the accuracy of correlations between tree ring location and measurements of chemical species within the tree to approximately 5 years in some species, or to U transport times through soil. Subsequent soil analysis showed U to be present at above expected concentration; the concentration increased with sampling depth and U at 60 cm below the surface was about ten-fold higher than U in near surface soils.

This study was inspired by comments from a local citizen in Ames who was concerned about a discharge of radioactive liquids that occurred over forty years ago. It represents one possible mechanism for developing information about past insults to our environment.

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References

1. R.L. Ferguson, M.C. Edelson, and H-m. Pang, "Laser Ablation System, and Method of Decontaminating Surfaces," US Patent Appl. No. 08/506,585, filed July 25, 1995.
2. E. Hoffman, C. Ludke, H. Scholze, and H. Stephanowitz, *Fresenius J. Anal. Chem.*, (1994) **350**, 253 - 259.
3. C. Geertsens, A. Briand, F. Chartier, J-L. Lacur, P. Mauchien, S. Sjostrom, and J-M. Mermet, *J. Anal. Atomic Spectrom.* (1994) **9**, 17 - 22.
4. A.R. Warren, L.A. Allen, H-m. Pang, R.S. Houk, and M. Janghorbani, *Applied Spectroscopy* (1994) **48**, 1360 - 1366.
5. T.M. Hinckley, J.P. Lassoie, and S.W. Running, "Temporal and Spatial Variations in the Water Status of Forest Trees," 1978, *U.S. Forest Service Monograph #20*, 72 pp.