### A Laser-Based Solution to Industrial Decontamination Problems

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## **Abstract**

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The ability of lasers to deposit significant amounts of energy on surfaces located at large distances from the laser can be exploited to solve very difficult industrial problems. The Ames Laboratory has been working in partnership with Lockheed Martin Idaho Technologies (LMIT) to apply laser technologies to the decontamination of radioactively contaminated surfaces located in hostile environments. Many such applications exist within former USDOE and nuclear industry facilities. As opposed to laser coating removal systems, which are designed to "strip" relatively soft coatings from a substrate without damage to the substrate, the system being developed by Ames - LMIT is designed to remove contaminants that are embedded within the metal surface itself. The system generates irradiance levels sufficient to remove microns of metal from a surface and an off-gas system that prevents the redeposition of materials removed from the surface. Process control is assisted by monitoring the laser-generated plasma produced during laser surface ablation. Results achieved using this apparatus for various metal types will be presented along with a discussion of other potential industrial applications.

#### Introduction

It has previously been demonstrated that Q-switched Nd:YAG and excimer lasers used at moderate irradiance values (~10<sup>8</sup> - 10<sup>9</sup> W\*cm<sup>-2</sup>) can decontaminate metal surfaces via laser ablation<sup>1,2</sup>. When a focused laser beam irradiates a metal surface, the surface will absorb a fraction of the incoming photons and, when the laser irradiance is sufficiently great, material will be ejected from the surface by a combination of processes that include vaporization and ablation. Decontamination is achieved by removing contaminated surface layers and then capturing the ejected material before redeposition can occur. Previous work determined that, using a laser with appropriate operating characteristics, ejected material could be transported away from the surface by an air stream and successfully captured with a high efficiency

particulate air (HEPA) filter. We note that laser decontamination can be effected in air at ambient pressures, which is an experimental simplification of great benefit to an end user.

Decontaminating a large surface area with a laser in a reasonable amount of time (i.e., competitive with industrial surface decontamination technology) requires that either the laser beam be sufficiently intense to achieve useful irradiance values over a large area or that the laser operate at a very high repetition rate. In either case, the laser beam needs to be rastered quickly across the surface to achieve large area decontamination in reasonable amounts of time.

This paper reports the results of an investigation into the use of a relatively new, high repetition rate (KHz range), high average power acousto-optically Q-switched Nd:YAG laser for metal surface decontamination. The relatively long pulse length of this laser (~100 ns) should reduce physical strain on optical fibers caused by faster pulse (~5 - 8 ns) Q-switched Nd:YAG lasers and allow the reliable, long-term use of such fibers for laser beam delivery. The laser output wavelength (1064 nm) is known to be efficiently transmitted by common optical fibers and the peak power of the laser beam after passing through the fiber should still be sufficient for efficient ablation and unlike continuous wave or long pulse width Nd:YAG lasers, which can be effectively transmitted by optical fibers, the acousto-optically Q-switched Nd:YAG laser has a pulse width sufficiently short to prevent substrate melting that can embed a surface contaminant in the bulk of the material being treated.

# **Experimental**

The laser used in this study (Model 405Q, U.S. Laser Corp.) is a high power, high repetition rate acousto-optically Q-switched Nd:YAG laser. During Q-switched operation, the repetition rate can be varied from 100 Hz to 30 KHz. A maximum output average power of 200 W was obtained when the repetition rate exceeded 9 KHz. The laser beam was delivered to samples located in a room remote to the laser with a ten meter long step index fused silica core fiber with a Kevlar reinforced PVC sheath (Mitsubishi Cable America, ST1000H-FV). This fiber has an 1000 μ m core diameter with a numerical aperture of 0.2. A flexible armored coiled metal jacket was used to protect the fiber. The fiber was usable for extended periods of time (i.e., one fiber has been used continuously for approximately ten months, the duration of this work) with only very occasional end repolishing needed to maintain maximum fiber transmission.

The effect of radiation exposure on fiber optic performance was tested. The laser transmission of three fibers, each one meter long, was measured in Ames and then the fibers were shipped to LMIT where they were exposed to a high energy

gamma radiation source for varying periods of time. After this exposure, the fibers were shipped back to Ames and the laser transmission was remeasured. Each fiber was tested at an input laser power of ~165 W for five minutes before and after exposure to the radiation source. No significant degradation of the fibers was observed.

A 37.5 mm focal length multielement coupling lens mounted in an input coupler was used to precisely focus the laser beam into the fiber core. The input coupler was equipped with a fine focus adjustment. The input end of the fiber was mounted on a X-Y translational stage for precise center line alignment. The output end of the optical fiber was connected to an output coupler assembly that held a 75 mm focal length (f.l.) lens for laser beam collimation and a 50 mm f.l. plano-convex lens that focused the laser beam onto the target surface. Each lens was anti-reflection coated (< 0.25% reflectivity) for 1064 nm operation. The laser spot size on the sample was ~0.9 mm in diameter and, typically, ~92% of the laser output power was delivered to the target through the fiber optics delivery system. A particle collection cell was used to collect material ablated from the surface. For laser testing without a fiber optic delivery system, the laser beam was focused onto the target sample by a plano-convex spherical lens with a 10 cm f.l. and the estimated spot size was 0.5 mm in diameter. The experimental arrangement is depicted in Figure 1.

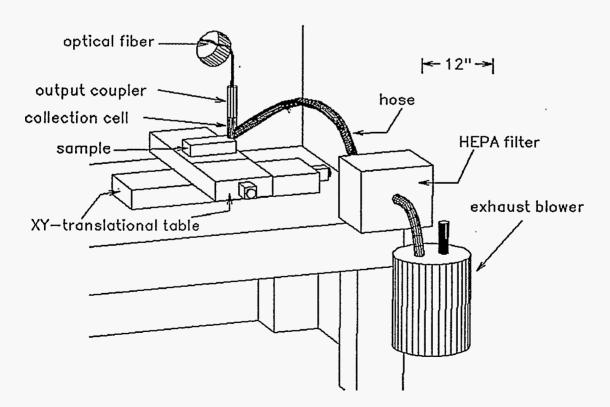


Figure 1. Expanded view of the experimental setup for laser ablation.

Metal samples were mounted on a computer controlled X-Y translational table (Techno, Model HL31SBM24494) and moved beneath the stationary laser beam at a maximum speed of 10 cm/s. The fiber output coupler was positioned so that the sample was at the focal point (± 0.5 mm) of the focused laser beam.

### Results and discussion

A method based on refractive index gradient<sup>3</sup> monitoring was used to optimize laser cleaning efficiency. A probe laser is deflected in the direction of a density gradient and creates a signal in a position-sensitive detector that is proportional to the particle density in a plume. When the probe beam is intercepted by the top of a laser generated plume, the probe beam is deflected downward so that reduced light intensity is registered by a suitably positioned (see Figure 2) photodiode. Once the plume center passes the probe beam, the probe beam is deflected upward and increased light intensity is registered. The angle of deflection of the probe laser beam is linearly proportional to the total number of atoms in the laser-generated plume<sup>3</sup>. Therefore, this technique can be used to monitor relative ablation rates by measuring the deflection angle or the signal change due to the density gradient deflection of a probe laser source.

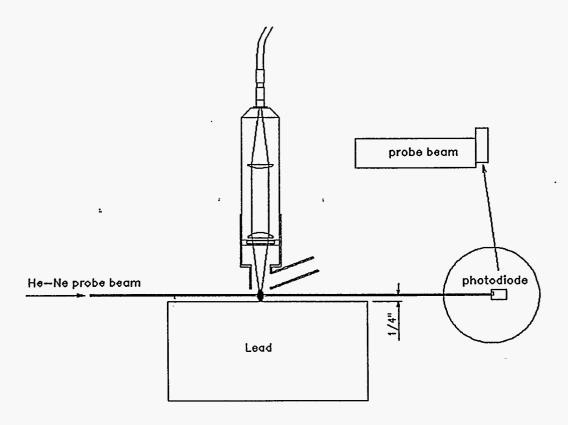


Figure 2. Experimental setup for density gradient measurements.

In our experiment, the photodiode was positioned to intersect ~3/4 of the probe beam, provided by a 1 mW He-Ne laser. The signal from the photodiode was measured and averaged by a digitizing oscilloscope. To validate our measurements of plume density by the refractive index gradient method, the results obtained by this method for ablation from a stainless steel sample were compared against weight loss measurements (no correction for oxidation was made).

Figure 3 shows the probe beam deflection signal for a stainless steel sample when the laser was operated at 5 KHz and a fiber optics delivery system was used.

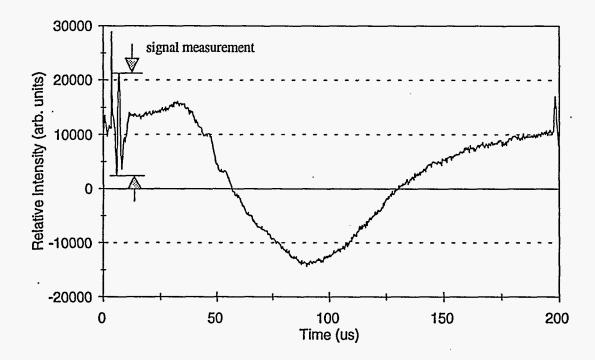


Figure 3. Result of density gradient measurements (stainless steel ablation).

When the probe beam is "off," no signal is observed even when the ablation beam is "on." This indicates that the photodiode is indeed measuring the deflection of the probe beam and not scattered light from the more powerful laser source. The first peak in Fig. 3 is related to the firing of the laser. Even when the probe beam is "off," this peak is observed whenever the Nd:YAG laser is fired. The second peak results from a negative-going signal and is followed by a positive-going peak as predicted in Ref. 3.

The peak height difference between the first negative and positive peak, as shown in Figure 3, was used to compare relative ablation rates at different laser repetition rates. Figure 4 shows a plot of deflection measurements versus laser

repetition rate for stainless steel ablation. Weight loss measurement results are included in Figure 4 and both measurements indicate that the ablation rates are similar when repetition rates vary from 1 to 4 KHz but start to drop off above 5 KHz.

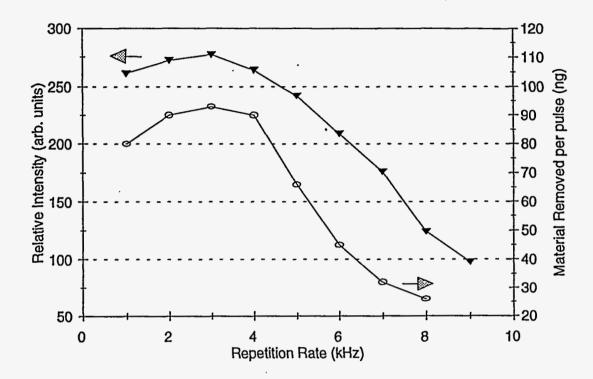


Figure 4. Comparison of density gradient (black triangle) and weight loss measurements (stainless steel). Both exhibit same repetition rate dependence.

# Determination of cleaning efficiency using simulated contaminants

Figure 5 shows how cleaning efficiency, as measured on SIMCON II coupons<sup>4</sup>, varies with laser repetition rate when not using fiber optic beam delivery. The laser was rastered over the surface twice at a scanning speed of 10 cm/s. Over 95% of Zr and ~100% of Cs was removed when the repetition rate was increased to 7 KHz. When the repetition rate was increased further, the removal efficiency of Zr dropped slightly but still exceeded 90%. For Cs, cleaning efficiencies near 100% were obtained even at the higher repetition rates. Several scanning speeds were used to determine whether increasing the exposure time would improve surface cleaning efficiency.

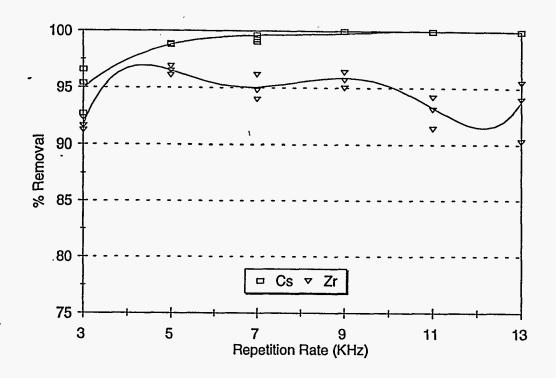


Figure 5. Cleaning efficiency of Cs and Zr on "SIMCON II" coupons as a function of laser repetition rate (without fiber optics delivery system).

Table 1 lists the average cleaning efficiency for Zr at three different scanning speeds (100% cleaning efficiency was obtained for Cs in all cases). In each case, the laser scanned the surface twice. No significant improvement was obtained even when the exposure time was doubled. When only one pass of scanning was used on SIMCON II coupons, the total cleaning time was 10 seconds at 5 KHz at a 10 cm/s scanning speed and the cleaning efficiency for Cs and Zr was 93% and 91%, respectively. This indicates that most of the contaminants were removed in the first pass.

Table 1. Zr cleaning efficiency as a function of scanning speed.

Scanning Speed (cm/s)	Removal Efficiency	Total time
10	93.1%	20 s
7.5	93.2%	30 s
5	95.5%	41 s

## Laser Cleaning of Radioactively contaminated Materials

1. Haynes 25 The ability of the high repetition rate, high output power Nd:YAG laser with a fiber optics delivery system to decontaminate a very hard alloy was tested. An excimer laser had been used in an attempt to clean this sample without any success. Due to the irregular shape of this sample, only one surface, which was flat, was treated with the Nd:YAG laser system. A hand held Geiger counter was used to monitor the radiation. The Geiger counter head (2" diameter) was positioned ~2 mm away from the flat surface during the radioactivity survey measurement. Since the contaminants emit penetrating radiation, the instrument reading comprised activity from the flat surface and also from other parts of the sample, even deep within the bulk of the material.

The laser was operated at 150 W at a 5 KHz repetition rate. An sample was translated along the X-axis beneath the laser at 10 cm/s; between the X-direction scans, the Y-table was stepped 0.5 mm to expose a new area for laser irradiation. Figure 6 shows the results of this experiment. The initial activity of the sample was

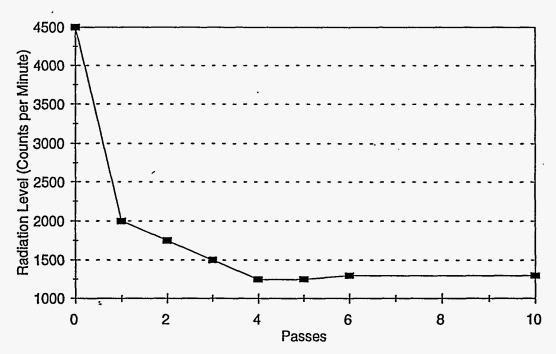


Figure 6. Decrease in radioactivity of the flat surface of a "REALCON I" sample as a function of the number of laser passes across that surface.

4500 cpm. After the first pass, the activity dropped to 2000 cpm. The activity continued to decrease until it reached ~1300 cpm after the fourth pass. The activity remained at this level even after several additional scans. Because only one surface was irradiated by the laser, it is possible that the residual radioactivity was due to contaminants on other surfaces or in the bulk of the sample. If we assume that the residual activity

(~1300 cpm) is extrinsic to the surface activity, then we calculate that the first pass of laser beam removed nearly 80% of the surface contamination ([4500-2000]/[4500-1300]  $\times$  100% = 78.1%). Thus, the acousto-optically Q-switched Nd:YAG laser with fiber optics delivery efficiently removed contamination from the surface of one of hardest alloys.

2. <u>Lead brick decontamination</u> A laser surface decontamination demonstration on a lead brick with fixed contamination was also performed. The smearable contaminants on the lead brick surface were removed at LMIT using the Pentek 604 Self-Stripping strippable coating prior to shipment to Ames. A hand held Geiger counter (Ludlum Measurements, Model 2) with a 2" diameter probe head (Ludlum Measurements, Model 44-9) was used to measure the remaining radioactivity. The results of these measurements at different locations on the brick indicated a range of contamination from ~2500 cpm on one section of a 4" x 8" face to 200 cpm on a section of another 4" x 8" face. The probe head was placed on the lead brick surface for these measurements and since the contamination was fixed, there was little chance for measurement error due to cross contamination.

The laser was operated at 4 KHz with an output power of 148 W (the estimated power delivered to the lead brick surface through the optical fiber system was ~133 W). The X-Y translational table was operated at a 10 cm/s scanning speed and there was a 0.5 mm step in the Y-direction between X-axis scans. Each brick surface was scanned once. The height of the fiber coupler was adjusted so that each surface was in the focal plane prior to laser irradiation.

After one pass on each surface, the radioactivity was measured to be less than 50 cpm on each surface except for one 2" x 8" surface. (This reading only slightly exceeded the background count in the laboratory, which varied from 30 to 50 cpm.) The activity of the left section of this 2" x 8" surface dropped from 600 cpm ~300 cpm after one laser scan. A second scan on this section reduced the activity to ~200 cpm but, even after sixteen scans, the radioactivity remained at ~200 cpm. This suggests that the contaminants in this section of the brick were deep within the lead brick. Nevertheless, all of the other surfaces were effectively decontaminated after only one laser pass at maximum translational speed. The radioactivity on the inside surfaces of the collection cell and the hose used to connect the cell to the HEPA filter were surveyed. It was found that there was no elevated radioactivity on these surfaces.

## **Conclusions**

It is clear from this work that a high power acousto-optically Q-switched Nd:YAG laser can be highly effective in removing fixed metal surface contaminants. The ability to successfully couple this laser to a fiber optic delivery system will facilitate the application of laser technology to surface decontamination even when these surfaces are located in remote and hostile locations without significantly reducing decontamination efficiency. A patent application covering the system described here has been submitted by this paper's authors. Inquiries concerning the commercialization of this technology can be addressed to any of this paper's authors.

A fieldable decontamination system based upon this device for remote applications awaits the development of a robotic deployment system but, given the flexibility attainable with fiber optics laser beam delivery, such a system should be easily assembled. The current system is capable of workstation cleaning applications and can be easily operated in standard industrial environments.

### References

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#### **Meet the Author**

Martin Edelson received a Ph. D. in Physical Chemistry at the U. of Oregon and has been at the Ames Laboratory since 1978. He is currently the Director of the Environmental Technologies Development Program and has been interested in novel applications of lasers to problems associated with the Department of Energy Environmental Management Program. He is married and once owned 12 cats.