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## Everlasting dark printing on alumina by laser

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### Abstract

Marks or prints are needed in almost every material, mainly for decorative or identification purposes. Despite alumina is widely employed in many different industries, the need of printing directly on its surface is still a complex problem. In this sense, lasers have largely demonstrated their high capacities to mark almost every material including ceramics, but performing dark permanent marks on alumina is still an open challenge.

In this work we present the results of a comprehensive experimental analysis on the process of marking alumina by laser. Four different laser sources were used in this study: a fiber laser (1075 nm) and three diode pumped Nd:YVO<sub>4</sub> lasers emitting at near-infrared (1064 nm), visible (532 nm) and ultraviolet (355 nm) wavelengths, respectively. The results obtained with the four lasers were compared and physical processes involved were explained in detail. Colorimetric analyses allowed to identify the optimal parameters and conditions to produce everlasting and high contrast marks on alumina.

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### 1. Introduction

Printing, painting or marking are some of the oldest crafts in the history of our civilization. From the very first ancient paintings made in the caves of our ancestors to the state-of-the-art printers nowadays, it has been a valuable, demanded and high-priced skill for the human being. Due to the new industrial requirements, there has been a lot of

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research and development on the technologies to produce different kind of marks in a wide range of materials. It can be done either by changing the surface appearance of the material itself or by adding a pigment, and thanks to it, current technology allows to print directly on almost every material by reliable methods.

Nevertheless, ceramics are materials that request for specific methods to print on them as a consequence of their singular nature. They usually need to withstand very high temperatures, so typical paints made by pigments are not useful for them.

Lasers have been largely used to process ceramics in different ways: cutting (Black, 1997, Quintero et al., 2005), drilling (Murray et al., 1999), cladding (Lusquiños et al. 2008), 3D printing (Comesaña et al., 2011, 2015) or surface glazing (Dhineshkumar et al., 2016) are examples of successful application of lasers on ceramics.

Laser marking is a reliable process and suitable for marking complex geometries with a high processing speed. Marks are made by changing the external appearance of materials, for instance by oxidation or by etching, but these approaches are useless in the case of ceramics.

Alumina is one of the most employed ceramic materials in industry. It has high electric and thermal insulating capacities as well as good mechanical behavior (wear resistance and hardness). However, as mentioned before, printing on it is a difficult task because is an oxide (so is not possible to mark it by oxidation) and pigments cannot be employed since the high performance temperatures for this material could easily degrade them. There is a possibility to mark alumina by doping it with any color imparting element such as iron, chromium and cobalt (Gündüz, 2015). However, currently there is no need of using any dopant since the production of high contrast marks on alumina by different lasers without addition of any element was already reported (Penide et al., 2015). They are produced by generating oxygen vacancies in alumina which are also called color centers or F-centers (Evans and Stapelbroek, 1978). However, as reported before, marks can be lightened and even removed in some hours by heating alumina at 650 °C in a oxidizing atmosphere (air) (Penide et al., 2015). Moreover, some authors reported that laser-induced coloring decreases over time (several tens of days) at room temperature for some glasses (Ashkenasi and Lemke, 2011). Therefore, marks made on alumina by laser might lose their coloration over time due to spontaneous filling of oxygen vacancies and this is an actual and important problem for the implementation of this technique. This is an open question that we tried to answer in this work. For that reason, here we report an experimental study on the evolution over time of the contrast of marks produced by four different laser sources emitting at three different wavelengths, as well as the different results of using these lasers for printing on alumina.

## 2. Experimental

The material employed for this study was alumina (97.5 % purity, GoodFellow) as sintered plates of 5 mm × 5 mm × 0.5 mm whose natural color is white.

Experiments were carried out by means of four different laser sources selected in order to compare the effect of wavelength on the so produced marks. First, an Ytterbium-doped fiber laser (SPI, SP-200) emitting in continuous wave at 1075 nm with an output power of 200 W coupled to a scanning head able to provide a processing speed of 300 mm/s. Next are three diode end-pumped Nd:YVO<sub>4</sub> lasers (Powerline E 20, Rofin-Sinar) emitting in their fundamental wavelength (1064 nm), second harmonic (532 nm) and third harmonic (355 nm). Using the last three laser sources, experiments were carried out with 13 W and 1 laser pass in continuous wave, 4.3 W and 10 laser passes at 20 kHz, and 1.35 W and 50 passes respectively and all experiments were performed at a scanning speed of 25 mm/s and at a pulse frequency of 20 kHz. In the case of green and UV lasers, more laser passes were carried out in order to balance the exposure time of the alumina samples to laser radiation. Laser treatments were made with a flow of argon right over the surface of alumina since it was clearly demonstrated to be the better choice so as to obtain the highest contrast marks (Pedraza et al., 1994; Penide et al., 2015).

Images of the surface of the treated and untreated samples were taken by an optical microscope (Nikon, SMZ-1000) coupled to a CCD camera. On the other hand, several colorimetric analyses were carried out with the aim of obtaining a quantitative result of the contrast between marks and untreated alumina. To do so, alumina samples were illuminated at 45° incidence using a white light fluorescent tube lamp (OSRAM L 13W/21-840) and a spectroradiometer (PR-650 Spectra Scan) was employed to obtain the tristimulus values CIEXYZ of the spectrum of the reflected light from 380 to 780 nm at 4 nm steps. Specifically, five measurements were taken for each sample calculating the average of these values. For the analysis of the contrast, the luminance value (“Y” of the CIEXYZ)

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