ANÁLISE NUMÉRICA DAS PROPRIEDADES TÉRMICAS NO PROCESSO DE REMOÇÃO DO METAL EM CALCÁRIO POR ABLAÇÃO TÉRMICA AO XXVIII CREEM

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Abstract. *The thermal ablation process is an advanced manufacturing technology that has drawn the attention of modern industry to its ability to perform microprocessing and nanomanufacturing. The fields of medicine, aerospace engineering, polymer manufacturing and welding are some of the main ones that utilize the method. Assuming that when solid parts are sufficiently heated, some materials can undergo a phase transition, changing from solid to gaseous state, this process being called "sublimation". If the material is heated quickly enough, in an environment with controlled boundary conditions, material is removed by sublimation, a process known as "thermal ablation". This phenomenon occurs when focusing a laser on a prototype, in a controlled environment. If the material reaches the sublimation temperature, ablation occurs on a scale proportional to the sublimation factor. In this way, a portion of the material transforms to the vapor state instantly, as a result of the rapid temperature variation, removing mass from the prototype system. This study presents a transient numerical analysis of the thermal behavior of materials during the ablation phenomenon through laser heating. The present work was done by replicating parts of a previously published experiment on the removal of an ore layer in a limestone monument by ablation. It also contains information about the software modeling of the described situation (with the basic programming of the process in the software COMSOL) and the effects/interaction of the laser pulse on the materials. The energy efficiency of the ablation process was calculated, comparing the use of a moderate and stable laser flux to a high peak energy laser pulse in terms of preserving the lower layers of the material. From the analysis of the temperature distribution obtained in the study of the problem of laser heating in thermal ablation and the parameters that influence the process, it was proved that shorter pulses with higher laser intensity have better energy efficiency compared to a long and stable flow.*

Keywords: *thermal ablation, laser pulse, heat conduction, numerical simulation.*

Resumo. *O processo de ablação térmica é uma tecnologia de manufatura avançada, que atraiu a atenção da indústria moderna, pela capacidade de realizar microprocessamentos e nanomanufatura. Os campos de medicina, engenharia aeroespacial, manufatura de polímeros e soldagem são alguns dos principais que empregam o método. Assumindo que quando peças sólidas são suficientemente aquecidas, alguns materiais podem ultrapassar uma fase de transição, mudando do estado sólido para gasoso, sendo esse processo denominado "sublimação". Se o material é aquecido de forma suficientemente rápida, em ambiente com as condições de contorno controladas, ocorre a remoção de material por sublimação, processo conhecido como "ablação térmica". Esse fenômeno ocorre ao focar um laser em um protótipo, em ambiente controlado. Se o material atingir a temperatura de sublimação, a ablação ocorre em escala proporcional ao fator de sublimação. Dessa forma, uma porção do material se transforma para o estado de vapor instantâneamente, como resultado da rápida variação de temperatura, removendo massa do sistema do protótipo. Este estudo apresenta uma análise numérica transiente do comportamento térmico de materiais durante o fenômeno de ablação através de aquecimento por laser. O presente trabalho foi feito replicando partes de um experimento previamente publicado, sobre a remoção de uma camada de minério em um monumento de calcário por ablação. Este também contém informação sobre a modelagem em software da situação descrita (com a programação básica do processo no software COMSOL) e dos efeitos/interação do pulso de laser nos materiais. Foi calculada a eficiência energética do processo de ablação, comparando o uso de um fluxo de laser de potência moderada e estável a um pulso de laser de alto pico de energia quanto a preservação das camadas inferiores do material. A partir da análise da distribuição de temperatura obtida no estudo do problema do aquecimento por laser na ablação térmica e os parâmetros influentes no processo, foi comprovado que pulsos mais curtos com maior intensidade de laser possuem melhor eficiência energética em comparação a um fluxo longo e estável.*

Palavras chave: *Ablação Térmica, Pulso de Laser, Condução de Calor, Simulação Numérica.*

1. INTRODUCTION

Industry attention to laser ablation have increased in recent decades with many new applications of the process in modern industry, such as minimally invasive ablative techniques in medical processes (Marqa *et al.*, 2011) and the manufacture of electronic microdevices (Hu *et al.*, 2020). This is due to the possibility of applying the method on tiny scales, such as nanometers, with high precision, flexibility and considerable resource savings.

Inside state-of-the-art manufacturing methods, laser ablation can be combined with other manufacturing processes such as additive manufacturing (shown in (Hu *et al.*, 2020)), coating (Cleries *et al.*, 2000) and welding. It can also be used to minimize roughness on surfaces after some fabrication techniques by changing surface properties (Bizi-Bandoki *et al.*, 2011).

In this study, a real situation was reproduced, in which a piece of limestone suffered deposition of iron dust from a industry, forming a tiny layer of this metal on the limestone, as shown in Fig. (1). This context was inspired by the studies of (Cocean *et al.*, 2017), which mentions the damage that iron, in contact with limestone (e.g. deposition of dust from metal industries on a monument, old building and civil constructions made of limestone material), can cause the surface. In this situation it is possible for changes in the appearance and color of the surface to occur due to chemical reactions. This interaction can cause limestone degradation. Thus, in order to clean this surface, following the governing equations, it is possible to remove the iron and achieve less degradation of the limestone. For this, a laser is pointed at the surface of the iron layer, controlled by parameters to promote the ablation of only the metallic material.

The aim of this work is to serve as a baseline for ablation simulations and to demonstrate the improvement in energy efficiency using high-peak-energy laser compared to a steady-state laser. Using the Comsol software, different conditions were tested and compared in order to prove that a short duration and high intensity laser regimen can be used to optimize the conditions of the ablation method, preserving the prototype and decreasing the total energy consumption. For this, specific parameters of laser energy, pulse time and conduction were analyzed in both materials.

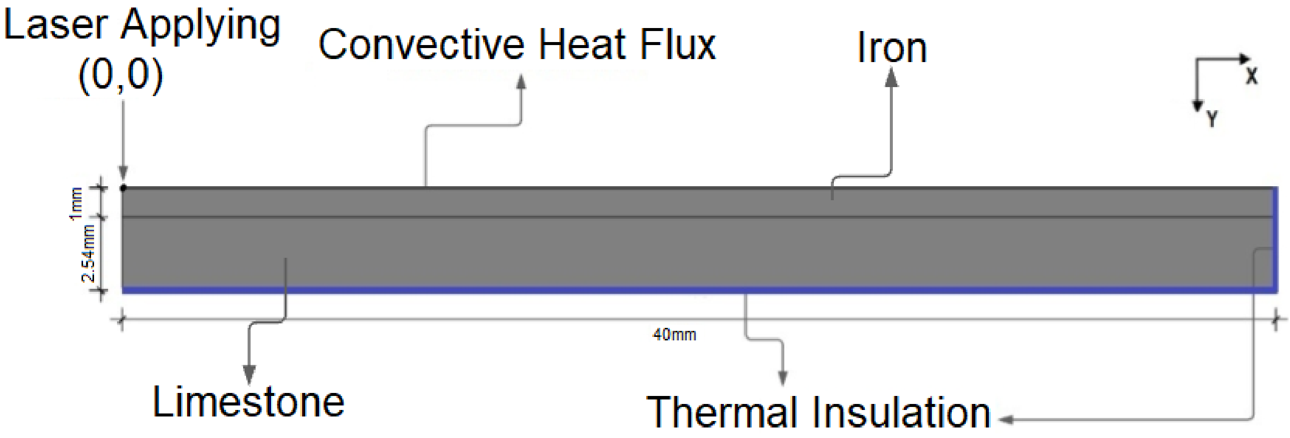

Figura 1. Schematic geometry of a surface metallic layer and a lower limestone layer, developed in COMSOL.

2. Methods and Materials

Aligned to the objective of this study, was developed a 2D geometric model composed by two materials (iron and limestone) in contact. The iron is represented by the 1 millimeter superficial layer, using the pure element Comsol library properties, and the limestone, by the 2.54 millimeters below layer, using the software library properties of the $CaCO_3$ (calcite) solid, polycrystalline. The laser is pointed to the above layer and have x coordinate constant along the trajectory. It decreases in the *y-axis* coordinates, while the material is suffering ablation. This materials were chosen and organized approaching to the conditions of a limestone piece exposed to the deposition of iron dust released by industries over nearby region. Initial ambient thermal properties were considered for both materials, as 293.15 K temperature and 1 atm pressure.

Developing the mathematical model, the first boundary condition of this simulation was the thermal insulation in edges that are not directly affected by heating or ablative effects. Then, considering the material vaporization, a second boundary condition was developed enforcing that the solid material cannot exceed the sublimation temperature. Finishing, was considered the heat loss by ablation, as described in (Frei, 2016).

This first boundary condition is initially defaulted by the software as isolation on all edges. As the other boundary conditions were applied, the affected edges receive new properties. Considering the physics applied (laser heating) to the geometry in the software at the point $x=0$ and $y=0$ (where the laser is pointed), these affected edges receives new heating parameters (general inward heat flux and convective heat flux, due to the laser incidence and heat loss caused by the material ablation, respectively).

The laser is represented by a general inward heat flux, modeled by the Gaussian Laser Beam distribution in 2D:

$$Q(x,y) = Q_0 exp(-(\frac{x}{0.5[cm]})^2) \tag{1}$$

where Q(x, y) is the energy distribution, Q_0 is the total laser power input.

For the last boundary condition, was considered a convection regime over the upper surface. The thermal condition used is an ablative heat flux modeled by the Equation (2).

$$q_a = h_a(T - T_a) \tag{2}$$

where q_a is the convective heat flux caused by the material ablation, T_a is the ablation temperature and T is the actual temperature of the point. The h_a coefficient is the temperature-dependent heat transfer, inputted as a linear ramped function, added in Comsol definitions using a 10^9 slope value, as have demonstrated (Frei, 2016).

Applying the described conditions and physics, when the material reaches the sublimation temperature it triggers off the material removal, that in the case of the element iron at ambient pattern properties, starts around 3134 K (Cocean *et al.*, 2017). The material removal is modeled by the physics "Deformed Geometry", using "Free Deformation" condition over both domains, "Prescribed Mesh Displacement" over the r axis (where *x=0*) and "Prescribed Normal Mesh Velocity" quantifying the material removal, with the Equation (3), as (Frei, 2016) have demonstrated.

$$v_n = \frac{q_a}{\rho * T_s} \tag{3}$$

where v_n is the material removal in m/s, ρ is the density and T_s is the sublimation temperature.

Tabela 1. Laser pulse time width, iron removal quantity and heat flux density parameters used to analyze the energy efficiency of a laser pulse

Time *s*	**Iron removal %**	**Thermal Flux Density** MW/m^2
60	33	89.80
	66	90.65
	100	91.30
30	33	108.50
	66	110.30
	100	111.30
10	33	151.35
	66	155.70
	100	159.00

Results taken from Comsol.

Following the aim of removing only the iron and preserving the limestone substrate, the laser power and incidence laser time were controlled, in order to achieve specific ablation levels. In this work, strategic parameters, shown in Table 1, were tested to analyze the total energy consumption at each of these laser pulse time width and heat flux density.

However, the ablation process involves some other physical phenomenons that were not considered in this study. A more elaborated investigation must include gas dynamics properties, plasma formation, plasma shielding effect (Aghaei *et al.*, 2008), Marangoni convection, among other physic phenomenons.

3. Results and Discussion

A numerical and graphical analysis were done in order to clarify the greater efficiency of the energy applied to the system, in terms of energy input intensity, laser incidence time, ablation effect on metallic material and laser power density.

A study over the total energy input was developed, with Tab. 1 data, to confirm that an optimal use of laser energy can be achieved, opting for a high-peak-energy laser pulse. It is obtained minimizing the pulse time width and maximizing the thermal flux density, as shown in Table 2. The total energy input can be measured multiplying the thermal flux density by the laser incidence time, obtaining the laser power density or laser fluence, unit used in (Yan *et al.*, 2019), where's exposed a nanosecond laser precision fabrication. This numerical analysis presented in Tab. 1 prove one of the advantages of the short time pulse parameters.

Other notable advantage of high-peak-energy laser pulse is the preservation of the substrate. The images in Fig. 2 obtained in Comsol simulations show the ablation process in iron according to the parameters defined in Table 1. Using this simulations, it's possible to analyze the heating conduction through the materials. A shorter and more intensely

Tabela 2. Comparison of total energy input on system between pulse time width parameters

Time *s*	**Iron removal %**	**Thermal flux density** MW/m^2	**Laser power density** MJ/m^2
60	33	89.80	5338
	66	90.65	5439
	100	91.30	5478
30	33	108.50	3225
	66	110.30	3309
	100	111.30	3339
10	33	151.35	1513
	66	155.70	1557
	100	159.00	1590

Data obtained from Table 1 analysis

repeated laser exposure implies lower conduction in the material, causing less impact on the substrate. It is very favorable to the ablation, considering that it limits the thermal conduction in materials. This simulation can be used to compare the heat conduction in materials used on different parameters of time. It is important to mention that in this simulation, the decomposition temperature for the limestone was not considered. As demonstrated (Do and Specht, 2011), the limestone decomposes in relevant rates at 850ºC.

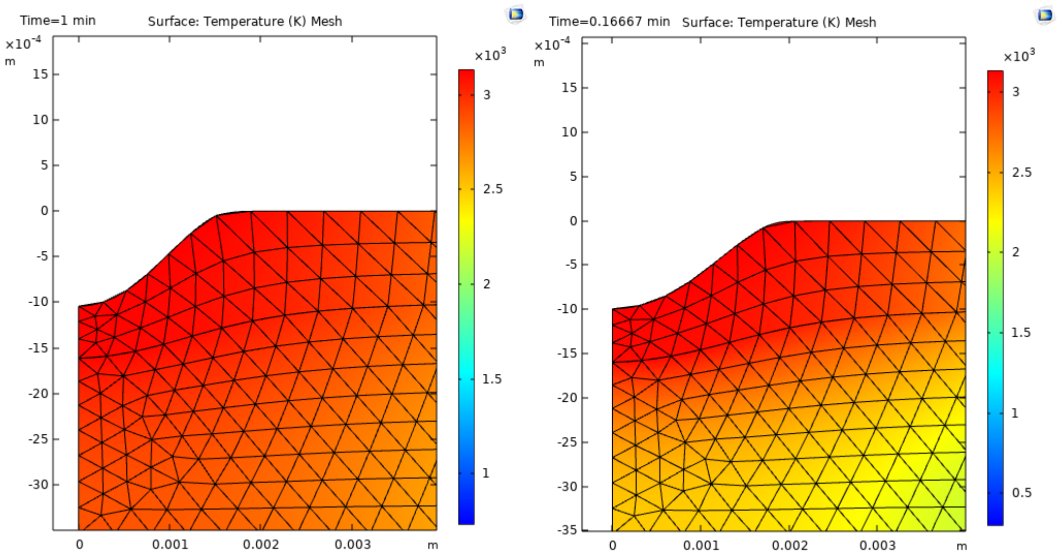

Figura 2. 60 and 10 seconds Comsol simulations of ablation process affecting 100% of iron layer

To achieve a better precision to reproduce the ablation phenomenon, considering the geometric model used in this simulation, an extremely fine mesh was used to define the element size. This configuration on this geometric dimensions divided this solid in 2252 elements in the mesh.

Analyzing these images is possible to conclude that the shorter the energy peak, lower is the conduction in the materials. Considering that the decomposition of the limestone achieve relevant rates at 850ºC, in this first analysis the bottom material would be modified, ruining the monument, civil construction or artifact made from limestone. Therefore, in order to prevent damaging the useful material, a extremely short peak of energy must be used, controlling the conduction through the material. The Figure 3 shows the great decrease of conduction in the substrate using a much shorter pulse time.

As observed, specially with the intention of separating materials with high vaporization temperature difference, a shorter peak time is a possible parametrization to achieve lower conduction rates, making it possible to control the heating and injuries caused by high temperature in the preserved material. The Figure 3 shows a shorter laser pulse time and a greater difference in convection ratio in material, achieving better preservation of the limestone. This simulation was parametrized with a 0.01 second laser pulse and 2000 MW/m^2 of laser flux density.

4. Conclusion

By the software simulations results, is possible to conclude that as much as the laser pulse width decrease the thermal flux density must increase to obtain the same ablation results. Although the energy input increase, the exposure laser time

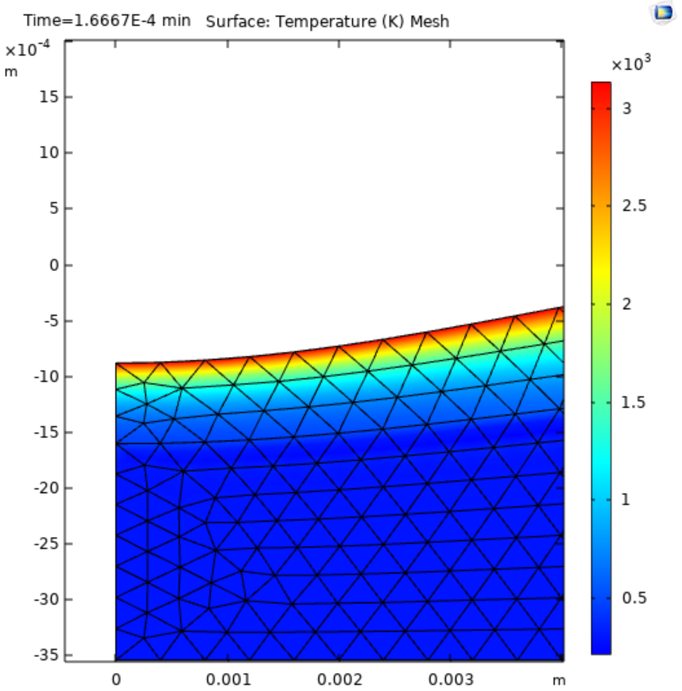

Figura 3. 0.01 seconds Comsol simulation of ablation process

reduction presents a major economy of total energy as shows Table 2.

Analyzing Table 2 we can compare the difference in laser power density to achieve the same ablative results, using 1 minute of laser pulse time width and 10 seconds. For example, to achieve 1/3 of total iron removed, in 60 seconds is necessary 5338 MJ/m^2, while in 10 seconds, only 1513 MJ/m^2. By this analysis is possible to understand the modern using of nanosecond-pulse laser in ablation process.

This work can also be used to simulate a 3D ablation phenomenon on solids. The presented geometry can be considered a 2D revolution solid, that using this same configurations exports this mathematical model to a 3D dimension problem.

This simulations does not consider some physical and chemical phenomenons that also influences the laser heating, as the plasma formation and plasma shielding (Vadillo *et al.*, 1999). It serves as basis for numerical analysis and presents useful information about laser ablation modeling of laser pulse interaction with materials.

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