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Laser powder bed fusion of refractory metals: A new way to produce components and devices for nuclear physics

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Summary. — The research conducted by the DIAM laboratory (Development and Innovation on Additive Manufacturing) attempts to produce and characterize materials that are not well specified nor well characterized by state of the art for the LPBF process but are not yet commercially available. The materials chosen were determined by experiments and research carried out by the National Institute for Nuclear Physics (INFN). This contribution focuses on refractory metals produced via additive manufacturing, especially molybdenum, tungsten, tantalum and niobium, from the material characterization point of view to the final product design and fabrication.

1. – Introduction

Additive Manufacturing (AM), also known as 3D printing, is considered the complementary technology to the conventional subtractive process. Metal Additive Manufacturing (MAM) is a production technology that fabricates metallic parts with layer-by-layer procedures, starting from different possible raw material forms (*i.e.*, powder, wire, or sheets). Even though the way to implement the "layer by layer" process can be different (*i.e.*, direct melting, joining, or binder jetting and sintering), as well as the energy source adopted, the main idea behind this technology is that a model made using a three-dimensional computer-aided design (3D CAD) system may be directly built in only one or a few steps. The manufacture of complicated geometries is made possible by AM technology. On the other hand, AM requires a thorough understanding of the limitations and qualities of the materials used to construct the additively manufactured part [1]. In fact, not all metallic materials are suitable to be produced using additive manufacturing production processes. For instance, this contribution illustrates some peculiar characteristics of different refractory metals produced by the Laser Powder Bed Fusion (LPBF) process. In the LPBF process, common research subjects include density measurements, surface morphology analysis, mechanical properties evaluation, and residual stresses analysis [2].

Despite the fact that the LPBF process manufactures functional components with peculiar characteristics and microstructure, more research is required to optimize the process parameters and building strategies to achieve the desired features [3].

Among the AM technologies, the LPBF is the most industrially widespread process used to fabricate additively manufactured metallic components. In the LPBF process, the raw material is metal powder, while the energy source is a laser beam, which melts the previously distributed powder bed by scanning it using a fixed speed (v), power (P), and distance between consecutive scan tracks (h). When the laser finishes executing the designed pattern for the specific layer, the build platform lowers by a predetermined value (t), and the process is repeated until the part is fully manufactured. All the aforementioned parameters make it possible to estimate the volumetric energy density (VED) that the laser supplies to the powder bed in order to melt it (see eq. (1)).

$$VED = \frac{P}{v \cdot h \cdot t}. \tag{1}$$

The contribution is structured in two parts. A brief introduction to the production of refractory metals by the LPBF process (sect. **2**). A summary of the DIAM lab research activity, mainly focused on the material characterization roadmap adopted before producing the final refractory metal parts, is described in sect. **3**. This last section is deepened into three subsections based on the additively manufactured material behavior affinity, molybdenum and tungsten (sect. **3**˙1), tantalum and niobium (sect. **3**˙2), and final product fabrication, particularly with tantalum and molybdenum, respectively for structural or non-structural application (sect. **3**˙3).

2. – LPBF of refractory metals

Refractory metals belong to the group of "transition metals". Differently from the other elements of the group, they have physical, chemical, and mechanical properties specifically useful for high-temperature applications in extreme operating conditions. One of the most important characteristics is the high density (*i.e.*, Os = 22.5 g/cm^3; W = 19.25 g/cm^3), which is combined with outstanding thermal shock resistance (W) and great resistance to electrochemical corrosion (Ta). These metals are therefore suitable for uses related to nuclear energy. Future nuclear fusion devices, like the tokamak at ITER, may use W as a material for their plasma-facing components (PFC) [4]. Mo is an excellent candidate for fusion reactor components due to its high thermal conductivity and temperature strength [5]. Niobium and its alloys are widely used in aerospace industry but also in high-energy and nuclear physics field, especially for the fabrication of next-generation superconducting radiofrequency (SRF) accelerating cavities [6]. One of the research activities conducted at the Development and Innovation on Additive Manufacturing (DIAM) laboratory of the National Institute for Nuclear Physics (INFN) aims to develop and characterize these exotic materials produced with the LPBF process. The materials chosen were determined by the projects and experiments carried out by INFN. Refractory metals, are employed frequently, for instance, in the construction of isotope separation on line (ISOL) facilities that generate radioactive ion beams for nuclear physics research [7,8]. Ta, Mo, and W semi-finished materials are particularly used in the manufacturing of some of the most crucial and significant ISOL components, such as the high-temperature production targets and ion sources.

Depending on the intended uses and performance requirements, refractory metal components are frequently produced using conventional manufacturing techniques. When the intended geometries are complex, as in the case of the aforementioned ISOL targets and ion sources, standard subtractive and joining procedures become troublesome and expensive [8, 9]. Normal production processes are challenging when Ta, Mo, or W are handled at high temperatures, such as in the heat affected zone (HAZ) of welding [10]. In addition, the hardness of Mo and W makes their production even more expensive due to material waste and higher machining tool costs. Sintering is an alternative powder metallurgy-based manufacturing technique that has been investigated and proven for refractory metals components production. As aforementioned, the most advanced manufacturing techniques in the powder metallurgy industry in order to obtain complex geometries and light-weight structures is additive manufacturing. The LPBF process provides a variety of advantages that deal with the main problems in the production of refractory metals. For instance, considering the high cost of powders like Ta, Nb, W, and Mo, the recycle of the unmelted particles is essential. Furthermore, the production process is carried out in a controlled environment chamber, which is an advantage required when producing refractory metals with high density due to their strong tendency to oxidize at high temperatures [11]. Due to their unique characteristics, which include a high melting point, high heat conductivity, and susceptibility to cracking, refractory metals produced by LPBF require special challenges.

3. – From material characterization to product design and manufacturing

The first step in order to obtain almost fully-dense AM refractory metals parts is the process parameters tuning. The fine-tuning of the latest starts from the Single Scan Tracks (SSTs) analysis. Thanks to the first screening of the parameters given by the SSTs analysis it is possible to narrow the parameter window in order to obtain volumetric samples that have densities close to fully dense. High-density specimens are then characterized at room and high temperature. The main goal of high-temperature thermal characterization is to measure emissivity and thermal conductivity in a temperature range as near as possible to 2000°C, the usual higher operating temperature of ISOL ion sources [12]. Assuming that thermal convection is negligible due to the high vacuum environment required for the functioning of ISOL high-temperature devices, these two parameters are in fact sufficient under steady-state conditions to define the temperature field within the ion source. A customized experimental set-up coupling with a consolidate electro-thermal numerical model, both developed at the National Laboratories of Legnaro (LNL) of the INFN, is used to evaluate the thermal properties in the 600–1600°C range [13]. Specifically, sample emissivity is determined using a ratio pyrometer during the radiative and conductive heating, whereas thermal conductivity is calculated using a steady-state high-temperature method based on pyrometric temperature observations and the inverse parameter estimation technique [14]. Tensile or compressive tests, according to the brittleness of the refractory metals tested, are performed at room temperature in order to evaluate the mechanical properties of additively manufactured specimens. Both for thermal and mechanical properties measurement, standard refractory metals are also tested, in order to compare AM with conventionally manufactured samples. The effect of building direction and post-processing machining of AM specimens was also investigated for tensile tests at room temperature. Last but not least, "secondary" process parameters that are related to geometrical performances of the additively manufactured parts were analysed to fine-tune the production process. This characterization

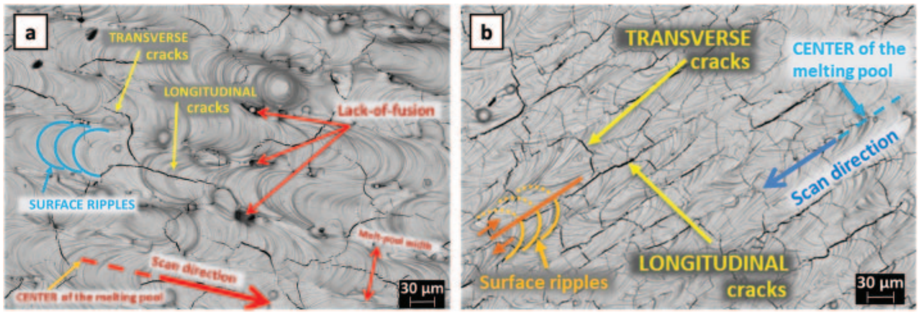

Fig. 1. – The crack network homogenously distributed on W [16] (a) and Mo [15] (b) manufactured by LPBF.

stage enables the investigation of the overhang angles and the evaluation of geometrical integrity as thickness changes, leading to the fabrication of complex geometry.

3·1. *Molybdenum and tungsten.* – Thanks to the fine-tuning of the process parameters, samples of both Mo and W were successfully produced. The best specimens have a high density, which is close to the bulk material produced with conventional production techniques. In the Mo characterization study published by Rebesan *et al.* [15], the density of the samples was particularly affected by the laser power parameter, reaching its maximum value at 150 W (99.9%). On the other hand, in the W production, Rebesan *et al.* [16] have demonstrated how the process parameter involving the distance between consecutive laser scan tracks (h) is decisive in order to raise the density of the manufactured part. In fact, using a low h (0.04 mm) in conjunction with scan speed values in the 400–600 mm/s range allows for density values greater than 99%, with a maximum of 99.61% for a scan speed of 600 mm/s.

The common feature of both additively produced Mo and W is the tendency to crack during the manufacturing process (see fig. 1). This phenomenon is widely described in the literature [15, 16].

The extensive presence of a homogeneously distributed network of cracks leads to a strong reduction of the thermal and mechanical properties, as demonstrated in the results highlighted by the characterization of AM Mo, both at room temperature and at high temperature [15]. This result leads to the final consideration that pure W and Mo are not suitable to be produced with this production process, guaranteeing the absence of defects and with properties close to standard W and Mo produced with traditional technologies. Kaserer *et al.* [17,18], demonstrated that for Mo the only possible solution to eliminate the presence of cracks and to obtain material properties comparable to standard Mo consists of using Mo alloys and a building platform heated up to 800°C. However, their use for the production of non-structural components is currently under investigation in order to verify whether the degree of freedom given by AM in the production of more performing geometries can equally guarantee a functional improvement at high temperatures.

3·2. *Tantalum and niobium.* – a completely different behavior with respect to Mo and W is shown by the Ta and Nb produced with the LPBF process. Indeed, as shown in fig. 2(b) there are no specific surface defects, such as cracks or lacks of fusion, and the melt pools appear to be continuous and homogeneous. This suitability to be produced by laser melting is confirmed by the high density value we obtain in the samples, 99.8% for Ta and 99.7% for Nb.

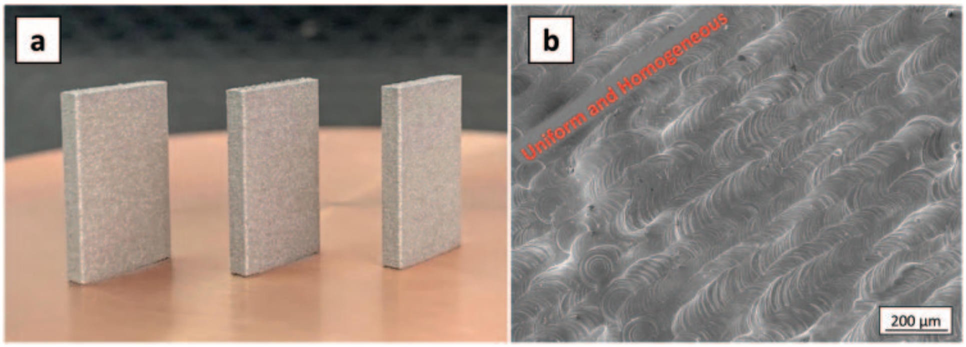

Fig. 2. – (a) The additively manufactured tantalum sample for process parameters tuning; (b) top surface morphology of the tantalum tag.

The preliminary results of the characterization of Ta AM performed so far have shown that the thermal and electrical properties at room temperature and at high temperature are close to those of the reference standard material, whose properties have been measured by the same method. The measurements of the mechanical properties of the horizontally produced as-built specimens show an increase in strength and a decrease in elongation compared to the conventionally produced standard material. The characterization of the electro-thermal-structural properties of Nb is now under development.

3˙3. *Components production and validation.* – As a result of the material characterization phase, from the physical, thermal, and mechanical properties measurements to the evaluation of geometrical performances of the additively manufactured and analysed refractory metals, the first AM molybdenum and tantalum components, shown respectively in fig. 3(a) and fig. 3(b), were successfully produced.

These components are the anode (Mo) and the cathode (Ta) of the FEBIAD (Forced Electron Beam-Induced Arc Discharge) Ion Source used in the production of extremely pure radioactive ion beams in the context of the Selective Production of Exotic Species (SPES) project. Currently, both the anode and the cathode of the traditional FEBIAD type ion source are made of three components, which are subsequently joined by TIG. This welding process strongly compromises the alignment of the components and compli-

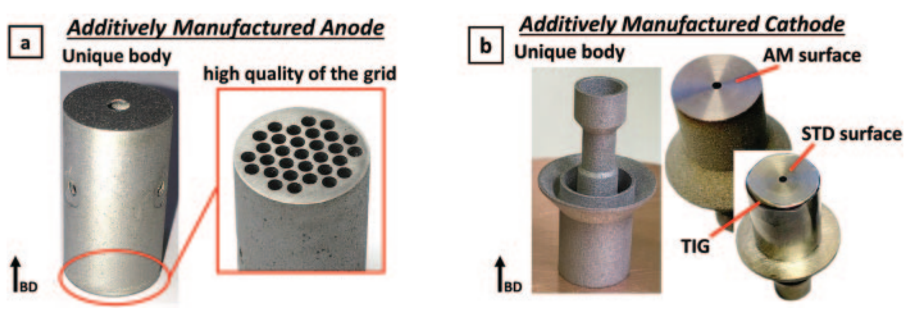

Fig. 3. – (a) The additively manufactured molydenum anode and (b) the additively manufactured tantalum cathode.

ance with the required tolerances, in particular for the distance between the anode and the cathode, a fundamental parameter for the correct functioning of the beam ionization process. Firstly, the main ion source components, produced entirely by AM, do not show the aforementioned issues caused by the welding process; secondly, AM allows for the investigation of new forms of anode-cathode interface and therefore for the evaluation of their effect on the variation of the physical performance of the source. The physical performance of the traditional geometry of the anode and the cathode reproduced by the LPBF process is now under investigation in preliminary high-temperature and off-line ionization tests carried out at the SPES and ISOLDE facility systems at INFN-LNL and CERN.

4. – Conclusion and further development

This study described the production process and the main characterizations performed by the INFN DIAM laboratory in order to be able to produce refractory metal components using Additive Manufacturing technology. Recent research has confirmed that pure W and Mo are not suitable for processing with LPBF technology on commercially available machines. The widespread presence of cracks in manufactured articles leads to a strong reduction in the thermal and mechanical properties of the material. In the near future, the presence of alloy elements that make them processable will be investigated. On the other hand, the LPBF technology is particularly suitable for additively producing Ta and Nb. These metals, even if pure, have densities close to Ta and Nb bulk. The next step is to finalize the evaluation of tantalum and niobium electrical, mechanical, and thermal properties. Finally, following all the steps reported, two components of the ionization source for the SPES project were produced: the anode, a non-structural component made in Mo, and the cathode, a structural component made in Ta. Both samples are currently undergoing high-temperature characterization and non-radioactive beam production in order to be able to compare these additively manufactured components with the traditional anode and cathode used historically.

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