ISBN: 978-3-95450-208-0 doi:10.18429/JACoW-IPAC2019-MOPTS093

ULTRA-HIGH VACUUM CHARACTERIZATION OF MOLYBDENUM-CARBIDE GRAPHITE FOR HL-LHC COLLIMATORS*

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Abstract

In view of the High-Luminosity upgrade of the Large Hadron Collider (LHC) collimation system, a family of novel molybdenum-carbide graphite (MoGr) composites was developed to meet the challenging requirements of HL-LHC beam-halo collimation, in particular the electrical conductivity and thermo-mechanical performances. The Ultra-High Vacuum (UHV) behaviour of this material was extensively characterized to assess its compatibility with the accelerator's specifications. The results presented in this paper correlate the outgassing behaviour with the microscopic features of MoGr compared to other graphite-based materials. Residual gas analysis (RGA) was exploited to optimize post-production treatments.

INTRODUCTION AND VACUUM REQUIREMENTS FOR LHC COLLIMATORS

In the High-Luminosity upgrade of the Large Hadron Collider (HL-LHC), the energy stored in the beam will increase by a factor of two with respect to the LHC: the absorber materials in the collimation system must maintain their structural robustness in case of accidental impacts, while minimizing the perturbation to the beam induced by RF impedance [1]. As a result of an intense R&D program, MoGr was identified as the optimal composite material to meet both thermo-mechanical and electrical requirements for the primary and secondary collimators [2]: MoGr, in fact, exceeds the electrical conductivity of isotropic graphite by one order of magnitude, while keeping comparable thermo-mechanical performances. The behaviour of this composite material has been structurally assessed under intense proton beam impacts [3], [4]. A prototype collimator with MoGr blocks is presently installed in the LHC. Additionally, the MoGr surface can be coated with pure molybdenum, further increasing surface conductivity by a factor of 20.

A challenge for the collimation system is that its materials must fulfil the UHV requirements of the collider. An adequate vacuum level limits the scattering of the protons against the residual gases and guarantees a longer beam lifetime. Collimators in the LHC must respect a limit for total outgassing of $2 \cdot 10^{-7}$ mbar·l·s^{-1}, which is related to the maximum acceptable pressure to guarantee 100 h beam lifetime [5]. Deducting the outgassing of the stainless steel vacuum tank and of the metallic components of the collimator jaws containing the MoGr blocks, the maximum tolerated outgassing for the absorber material is $5 \cdot 10^{-8}$ mbar·l·s^{-1}. The nuclear scattering cross-section of the proton beam with the gas particles depends on their atomic number, as shown in Table 1 [6]. For this reason, additional UHV requirements are also imposed on the residual gas composition.

Table 1: Expected Proton-Nucleus Cross-Section at LHC for Some Gases

Gases	Cross-Section (mb)
H_2	94
He	130
CH_4	568
H_2O	554
CO	840
CO_2	1300

For a metallic component after bake-out, the dominant outgassing species is H_2; therefore, the RGA and the UHV acceptance limits for molecular mass are usually normalized to this gas, as shown in Figure 1.

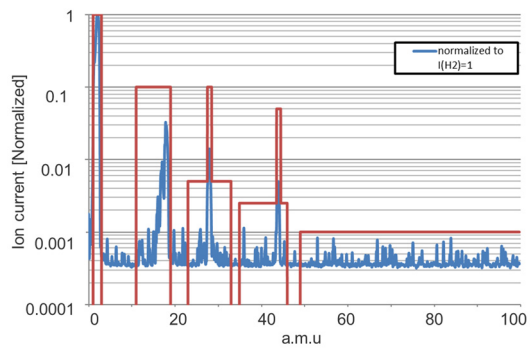

Figure 1: RGA analysis typical of a metallic component (blue) and the LHC acceptance limits (red).

Finally, it is important to note that collimators are installed in the Long Straight Section (LSS) of the LHC, where the neighbouring beam pipes are coated with Non-Evaporable Getters (NEG) to increase the pumping speed of H_2, CO, CO_2, H_2O, N_2 and O_2 [7], and decrease the secondary electron yield of the walls. Except for H_2, the NEG coating can rapidly saturate in case of high absorption of the other species. For this reason, the air flow degassed by a component must be under tight control: the acceptance limit for the so-called virtual leak of a collimator is $5.0 \cdot 10^{-9}$ mbar·l·s^{-1}. This value corresponds to the saturation of one meter of NEG-coated beam pipe in 150 days.

*Work supported by the European Union HL-LHC Project. Research supported by the HL-LHC project. 453. The research leading to these results has also received funding from the European Union's Horizon 2020 Research and Innovation program under Grant Agreement n. 73087.
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PROCEDURE FOR OUTGASSING MEASUREMENTS

The absorber materials are tested in a vacuum test bench, equipped with a pumping unit, pressure gauges and RGA prior to their installation in the collimator jaw.

Before starting operation, all the collimators in the ring are isolated by closed valves and baked-out at 250°C for 48 hours, to desorb water molecules. For this reason, the outgassing level and gas species of the absorber blocks are evaluated by the gauges and RGA at room temperature, after a bake-out in the same conditions [8]. The virtual leak is assessed by measuring the argon signal in RGA over time without any active pumping in the system, since the argon content in air is well established.

UHV CHARACTERISTICS OF MOLYBDENUM-CARBIDE GRAPHITE

The RGA of a block of MoGr (grade Nb8403Ng, produced by Nanoker) is shown in Figure 2. After the production, this material is treated under vacuum at 950°C for 72 hours. The reduction of the MoGr outgassing as a function of the duration the vacuum firing was studied [9], and after 72 hours all tested blocks are acceptable for installation.

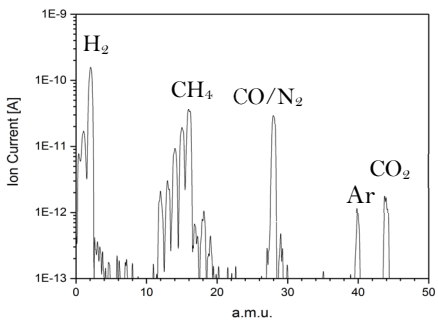

Figure 2: RGA of a MoGr block after vacuum firing.

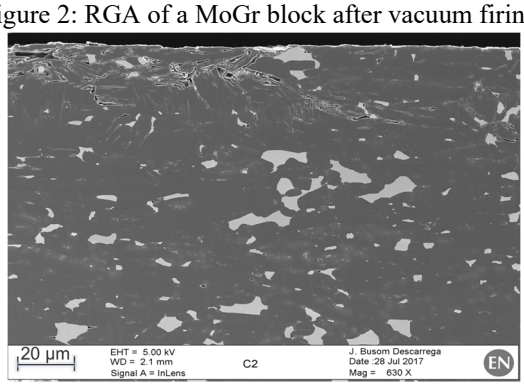

Figure 3: Micrograph of ion-polished MoGr.

The peaks corresponding to atomic mass unit (u) 28 (N_2 or CO) and to u 40 (Ar) indicate the presence of air. Different microscopic investigations (Figure 3) reveal that both superficial and bulk porosities can trap air molecules during the production process and the subsequent exposure to air. Other peaks corresponding to u 15 (CH_4) and u 44 (CO_2) characterize the MoGr spectrum of gases. They can be induced by the reaction of carbon atom with H_2/H_2O during the production process.

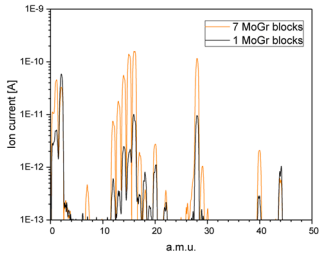

Figure 4: RGA of a MoGr block after vacuum firing with argon or with neon.

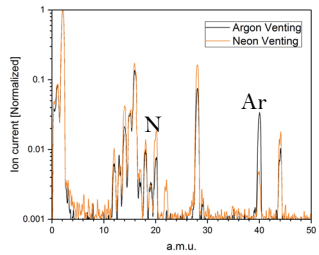

Figure 5: RGA of one and seven MoGr blocks.

Given the characteristic of carbonaceous materials to absorb atmospheric gases in their porosities, the authors decided to use neon as the venting gas in the furnace, after the vacuum firing treatments. The aim is to fill the porosities with neon, mostly replacing air as the absorbed gas. Neon has no impact on the NEG saturation, therefore it is a suitable choice for the LHC. Figure 4 highlights the sensitivity of MoGr to the venting history: the peaks of Ar (u 40) and Ne (u 20) increase after the venting with the corresponding gas. This also indicates that the mechanism of absorption is fast, and it occurs as soon as the material is exposed to a gas at atmospheric pressure, while its desorption is very slow: in fact, the presence of venting gases is still detectable at the end of the vacuum test, almost after 5 days in the test bench, with a pressure of $10^{-10} \div 10^{-6}$ mbar. Further studies to enhance Ne absorption by injecting it at higher temperature, during the cool down in the furnace, are ongoing.

A key feature of the material is that the total H_2 outgassing rate, usually dominant for metallic materials, is lower than the detection limit of the test bench. The peak of u 2, in Figure 2 and Figure 4 is coming in fact from the background of the experimental setup. Comparing the RGA of one MoGr block with that of seven blocks, we observe that H_2 peak remains almost constant, as shown in Figure 5, whereas all the other species (e.g. CH_4, N_2, Ar) increase linearly with the material surface. Therefore, the H_2 signal comes almost totally from the test bench. As a consequence of this, the standard acceptance criteria based on the hydrogen normalization cannot be applied for this material. To evaluate the influence of the CH_4 outgassing, which is the main peak of the RGA, on the beam lifetime, a dedicated simulation was performed, demonstrating that its effects are negligible[10].

The specific outgassing of MoGr averages around $6 \cdot 10^{-12}$ mbar·l·s^{-1}·cm^{-2}. Considering the total surface of MoGr in a collimator, the maximum outgassing is always below the specified limit of $5 \cdot 10^{-8}$ mbar·l·s^{-1}. The internal leak rate ranges from $3.8 \cdot 10^{-9}$ to $1.4 \cdot 10^{-8}$ mbar·l·s^{-1}, in some case exceeding the acceptance threshold. Should this occur, the effect will be mitigated by the installation of NEG cartridges, connected to dedicated ports on the collimator vacuum tank. This mitigation is now planned for future collimators.

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COMPARISON WITH OTHER GRAPHITIC MATERIALS

It is interesting to compare the UHV behaviour of MoGr with other graphitic materials for collimation applications. In particular, the commercial isotropic graphite (grade R7550, produced by SGL) and the Carbon-fibre-carbon (CFC, grade FS140, produced by Tatsuno) are investigated. It is important to underline that the electrical conductivity of these two materials is too low for HL-LHC primary collimators and for other reasons, not discussed in this paper, their application was discarded for the coated secondary collimators. From a chemical point of view, the matrix of the three materials is identical, containing only carbon in its graphitic form. MoGr is reinforced by a small amount of molybdenum carbide particles [11] and CFC by carbon fibres. However, the differences in the production process lead to a different final microstructure, which influences also the UHV behaviour of the materials. The typical values of outgassing rate for specific grades of MoGr, CFC and graphite, are given in Table 2.

Table 2: Outgassing Rate for Different Graphitic Materials

Material	Thermal treatment	Specific outgassing rate [mbar·l·s^{-1}·cm^{-2}]
MoGr Nb8403Ng	72 h vacuum firing at 950°C	$6 \cdot 10^{-12}$
CFC FS140	48 h vacuum firing at 950°C	$2 \cdot 10^{-12}$
Graphite R7550	48 h vacuum firing at 950°C	$8 \cdot 10^{-13}$

Observing the cross-section of a graphite sample in Figure 6, left, we can immediately realize that its porosities are larger compared to MoGr (Figure 3). An overall higher porosity speeds up the air outgassing: once the material is under vacuum, pores are rapidly emptied through big pore/surface channels, and at the end of the bake-out cycle the measured residual outgassing is lower. In MoGr, air is degassed through tinier channels, and therefore the time to evacuate it is longer. CFC FS140 has an even lower density with respect to graphite R7550, and the surface is characterized by macroscopic porosities with extension of hundreds of micrometres (Figure 6, right). The speed of air degassing should thus be even higher. The reason for its worse UHV behaviour with respect to graphite could be related to nano-channels at the interface between the fibres and the matrix, or within the carbon fibres themselves [12], and has to be furtherly investigated.

INFLUENCE OF PRODUCTION PARAMETERS

Previous considerations indicate that for the investigated materials, larger porosities may lead, after pumping, to better UHV performances. It is thus important to limit the material compaction during production, weighing the improvement in the UHV properties with the potential loss in thermophysical properties in the case of too low density.

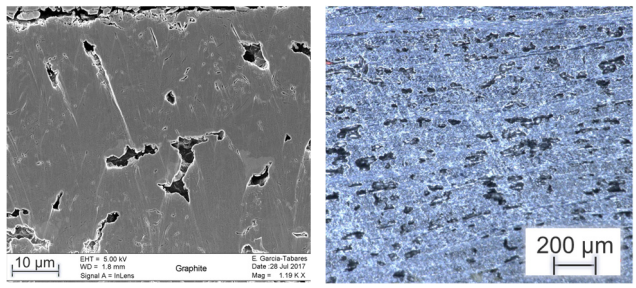

Figure 6: Micrographs of ion-polished graphite (left) and CFC (right).

The final density of the material must consequently be tuned by acting on the parameters (temperature, pressure, duration) of the production cycle. In Table 3, the density of two different MoGr grades is correlated to electrical conductivity and total outgassing. Notably, these results seem to indicate that there is a threshold compaction above which the rate of air outgassing is too low to empty the porosities in a reasonable time. We observe, for a decrease in density of 2%, a reduction in the electrical conductivity of 16%, coming with a significant decrease (factor of 30) of the outgassing rate. The fact that a small variation in density leads to these results suggests that both bulk and surface contribute to the outgassing of this material.

Table 3: Comparison between Different MoGr Grades

Grade	Density [g·cm^{-3}]	Outgassing [mbar·l·s^{-1}·cm^{-2}]	Electrical conductivity [MS/m]
Nb8304Ke	2.60	$1.7 \cdot 10^{-10}$	1.02
Nb8304Ng	2.55	$6 \cdot 10^{-12}$	0.85

CONCLUSIONS

The UHV compatibility of a new composite for HL-LHC low-impedance collimators was studied and optimized. In this paper, the UHV behaviour of the material was detailed and compared to other graphitic materials for collimation application. We observed a qualitative correlation between the UHV performance and the material porosity, which can be tailored acting on the production parameters. The findings highlighted in this paper suggest promising routes to further improve the UHV performance of similar graphitic materials for future applications in particle accelerators and other high-quality vacuum domains.

ACKNOWLEDGEMENTS

The authors want to thank E. Garcia-Tabares and J. Busom-Descarrega for the micrographs and C. Vazquez Pelaez for some of the outgassing measurements.

REFERENCES

[1] G. Apollinari, I. Béjar Alonso, O. Brüning, M. Lamont, L. Rossi, "High-Luminosity Large Hadron Collider (HL-LHC): Technical Design Report V.0.1", CERN, Geneva, Switzerland, 2017.

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[2] A. Bertarelli *et al.*, "Novel materials for collimators at LHC and its upgrades", in *Proc. HB'14*, East-Lansing, MI, USA, Nov. 2014, paper THO4AB03, pp. 438-442.
[3] F. Carra *et al.*, "Mechanical engineering and design of novel collimators for HL-LHC", in *Proc. IPAC'14*, Dresden, Germany, Jun. 2014, paper MOPR0116, pp. 369–372.
[4] A. Bertarelli *et al.*, "Dynamic testing and characterization of advanced materials in a new experiment at CERN HIRADMAT facility", IOP Conf. Series: *Journal of Physics*: Conf. Series 1067, 082021, 2018.
[5] G. Bregliozzi *et al.*, "Description of criteria for vacuum acceptance test", EDMS 1753872, CERN, Geneva, Switzerland, 2017.
[6] P. Grafström, "Lifetime, cross-sections and activation", CAS, Platja d'Aro, Spain, 2016, pp. 226-231.
[7] G. Bregliozzi *et al.*, "Evaluation of the NEG coating saturation level after 3 years of LHC beam operation", in *Proc. IPAC'13*, Shanghai, China, 2013, paper THPF1049, pp. 3397-3399.
[8] G. Cattenoz *et al.*, "Outgassing tests of LHC components in VSC vacuum laboratory of building 113", EDMS 1312756, CERN, Geneva, Switzerland, 2013.
[9] C. Accettura, "Ultra-high vacuum characterization of advanced material for future particle accelerator", Master Thesis, Politecnico di Milano, Milan, Italy, 2017.
[10] P. Chiggiato, "Final statement about collimators equipped with MoGr jaws", EDMS 2061004, CERN, Geneva, Switzerland, 2018.
[11] G. Guardia *et al.*, "Development and properties of high thermal conductivity molybdenum carbide-graphite composites", *Carbon*, vol. 135, pp. 1219–1221, 2018.
[12] J. S. Marcuzzo *et al.*, "Influence of Thermal Treatment on Porosity Formation on Carbon Fiber from Textile PAN", *Materials Research*, vol. 16, pp. 137-144, 2013.