

THERMO-PHYSICAL AND MECHANICAL CHARACTERISATION OF NOVEL MATERIALS UNDER DEVELOPMENT FOR HL-LHC BEAM INTERCEPTING DEVICES*

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Abstract

The collimation systems for high energy particle accelerators as High Luminosity LHC (HL-LHC), must be designed to withstand the close interaction with intense and energetic particle beams, safely operating over an extended range of temperatures in harsh environments and extreme conditions (pressure, strain-rate, radiation) [1].

In order to withstand such conditions, the materials must possess, among other properties, outstanding thermal shock resistance and high thermal and electrical conductivity. Such conditions cannot be found in standard materials but are only met by advanced or novel composite materials [2]. Therefore, an extensive R&D program has been launched to develop novel materials capable of replacing or complementing materials used for present collimators. So far, Molybdenum Carbide – Graphite (MoGr) and Copper-Diamond (CuCD) composites have been identified as the most promising materials.

Literature data are scarce or non-existing for the extreme conditions that the beam intercepting devices see in service even in the case of standard materials. Moreover, advanced or novel materials have in general not extensively been characterized. For these reasons, the successive characterisation campaigns constitute a linchpin of the R&D program. This paper reviews the experimental program performed for the thermo-physical and mechanical characterisation of the materials, and discusses the most relevant techniques and results.

INTRODUCTION

The construction of highly energetic particle accelerators such as the Large Hadron Collider (LHC) [1] is of paramount importance for modern high energy physics. This has also made tangible the need for advanced cleaning and protection systems, such as collimators, in order to safely increase the energy and intensity of particle beams, and hence extend further the reach of particle accelerators. The LHC collimators must adopt materials able to withstand the extreme conditions (temperatures, pressures and radiation) induced by the accidental impact of particle beam pulses; in addition to

outstanding thermal shock resistance, these materials are typically required a number of additional relevant properties, such as high electrical conductivity, geometrical stability and resistance to radiation damage [3].

These already stringent requirements are to become even more severe with the machine upgrade foreseen by High Luminosity LHC, which aims at increasing the peak luminosity seen by ATLAS and CMS experiments by a factor of 5, and the integrated luminosity by a factor 10. This is achieved with a twofold increase of the beam intensity. Within this framework, the Carbon-Carbon composites currently installed in primary and secondary collimators might jeopardise the machine performance due to the high impedance induced by the Carbon-Carbon composites low electrical conductivity. In addition, Inermet 180, the tungsten alloy used in tertiary collimators presents very poor robustness in case of beam impacts and could limit the reach in peak luminosity [1].

In order to face these challenges, an intense R&D program has been launched at CERN several years ago aiming to explore or develop a range of novel materials which are to combine the excellent properties of graphite or diamond, specifically their low density, high thermal conductivity, low thermal expansion, with those of metals and transition metal-based ceramics possessing high mechanical strength and good electrical conductivity.

This article presents the outcomes of the characterization campaigns performed over the most promising materials identified so far, namely Molybdenum Carbide – Graphite, Copper-Diamond (CuCD), and Carbon-Carbon Composites.

MATERIALS

MoGr (Fig. 1 left) is a ceramic matrix composite produced by Liquid-Phase Sintering (LPS) at a temperature above the melting point of molybdenum carbide (2589° C) [4]. It features low density, extremely high degradation temperature, good damping properties and excellent thermal conductivity and very low coefficient of thermal expansion (CTE), at least in the direction aligned with the graphite basal planes. A broad range of parameters have been tested in the development phase: composition, powder types and dimensions and processing cycle among others. Currently MoGr is the baseline material for the installation in HL-LHC collimators due to its excellent thermal and electrical properties, its improved radiation resistance [5], and a reasonable industrialization process, which tolerates the

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production of fairly large components that can be easily machined.

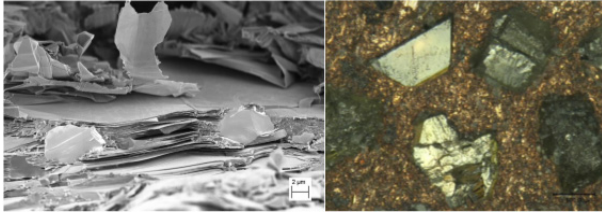


Figure 1: Micrograph of MoGr (left) and CuCD (right)

CuCD (Fig. 1 right) is produced by Solid-State Sintering. Its initial volumetric composition is 60% diamond, 39% copper and 1% boron [6]. The addition of boron promotes the formation of carbides in the diamond copper interface, and partially mitigates the lack of chemical affinity between the two elements which leads to reduced mechanical strength and thermal conductivity.

Despite its promising electrical and thermal properties, the complex industrialisation process and machining with precise tolerances hinders its practical application for collimators that require tight alignment tolerances.

Carbon fibre – Carbon (CFC) is another ceramic matrix composite in which randomly dispersed fibres are used as a reinforcement phase. The material has been used in the LHC collimators due to its low density, thermal shock resistance, and low thermal expansion. Its poor electrical conductivity make it less appropriate to be embarked in HL LHC collimators.

Amongst the three mentioned materials, MoGr is transversally isotropic and CFC is orthotropic, while CuCD is isotropic.

THERMAL CHARACTERIZATION

The density of the materials is measured with a class I hydrostatic balance, whenever possible according to the Standard ASTM B311-13 [7], employing ethanol as liquid medium. The density of the latter is determined prior the measurements of the samples by measuring the weight in air and in the alcohol of a reference volume of 10 cm³. The density measurements of the samples are carried out within a half-hour from the determination of ethanol density and the room temperature is monitored to detect excessive excursions. Typically, the density is measured over laser flash analysis (LFA) specimens, before the thermal diffusivity test are carried out. When the sample showed excessive porosity, i.e. it absorbed non-negligible quantity of liquid, it was baked at low temperature to dry it prior the LFA characterization.

The specific heat, $c_p(T)$, is measured by means of the differential scanning calorimeter DSC 404 C Pegasus® by NETZSCH, according to the Standard DIN 51007 [8]. The procedure consists in three consecutive measurements, performed under the same conditions. The first is done with empty pans and aims to establish a baseline for the following measurements, representing the intrinsic behaviour of the instrument, the second one is a measurement over a standard reference sample of known c_p , e.g. POCO-graphite and sapphire, and the latter is the

measurement of the unknown sample. It is possible to determine the c_p , by comparison of the heat fluxes occurred during the two final measurements. The test environment atmosphere is characterized by Ar flowing at 50 ml/min to prevent oxidation. This is especially important when measuring graphitic materials which combust at temperatures above 400°C.

The coefficient of thermal expansion, CTE (T), is evaluated by measuring the change of length of the sample subjected to linear heating/cooling phases, from room temperature to a maximum of 1950°C. The instrument used is a horizontal push-rod dilatometer DIL 402E by NETZSCH. The sample in the shape of a cylinder is housed in a tubular holder and put in contact with one extremity of the push-rod. The sample carrier is enclosed in a water-cooled electrical furnace. A displacement sensor (LVDT), connected to the other extremity of the push rod, acquires the raw displacement data, which is the combination of both the instrument and the unknown sample's change of length. To obtain the sample expansion or contraction from the raw signal, a preliminary measurement over a reference standard material with a well-known behaviour, like. POCO-graphite, is required. The test parameters, e. g. heating/cooling rate, atmosphere and temperature program, are chosen conforming to the Standard ASTM E228 [9].

The laser flash apparatus LFA 427 by NETZSCH is the instrument used to measure the thermal diffusivity, $a(T)$. The disk-shaped test specimen is embedded in a furnace, able to reach 1950°C, and housed on the top of a tubular sample holder. The method relies on a model, initially proposed by Parker et al. [10], for the temperature rise response of the rear surface of a disk when its front face receives an infinitesimally short heat flux. In the LFA, the heat input is performed by a laser pulse (typical duration 0.6 ms) and the temperature rise of the rear surface is sensed by a liquid nitrogen-cooled InSb detector. The simplest relationship between time response and thermal diffusivity is: $a=0.1398 \cdot d^2/t_{0.5}$, where d is the thickness of the sample and $t_{0.5}$ is the time at which the rear surface reaches half of its maximum temperature. The tests parameters are in agreement with the Standard ASTM E2585 [11].

The thermal conductivity is determined using the following relation: $\lambda(T)=\rho(T)c_p(T)a(T)$.

The sample sizes are presented in Table 1:

Table 1: Sample Geometries Summary

Technique	Sample Size [mm]
DSC	ø 5 x 0.75
DIL	ø 6 x 25
LFA	ø 10 x 2.5
IET and Flexural Test*	25-55 x 10 x 4
Compression Test	ø 7.5 x 15

*flexural sample sizes are not compliant with ASTM C651. Wider geometries have been chosen in order to ease the excitation of its torsional modes in IET tests.

MECHANICAL CHARACTERIZATION

The mechanical tests are aimed to the determination of flexural, compressive and elastic properties of the materials. The tests have been designed as a function of the achievable sample size: usually the material prototype is produced in small volumes, therefore the samples size is limited. In this perspective, the Impact Excitation Technique (IET) is chosen to determine the elastic properties, because it is a non-destructive technique and can be applied on the sample specimens cut for the flexural test. The IET is carried out according to the Standard ASTM C1259 [12] over prismatic specimens supported with wires along the nodal lines. In this configuration, the resonant frequencies are excited by impacting with a small hammer on the antinodes, and sensed with the help of a microphone. After this test, the same specimens are subjected to the 4-point bending technique, performed on a Zwick/Roell Z400 Universal testing machine fitted with a fixture according to the Standard ASTM C651 [13]. The strain of the tensed face is measured with HBM 1-LY11-3/350 strain gauges while the force applied is sensed by a HBM S2M 1kN load cell. The compression tests are performed according to the Standard ASTM C695 [14] by using the compressive plates of the UTM: the force is measured with the load cell integrated in the machine while the axial strain is measured with HBM 1-LY11-3/350 strain gauges.

RESULTS

Since the beginning of the R&D programme more than 30 different grades of novel or advanced materials have been characterized from the thermo-physical and mechanical point of view. The results depicted in Figs. 2 and 3 and in Table 2 show the thermo-physical properties of one of the most representative grades of the three previously mentioned materials.

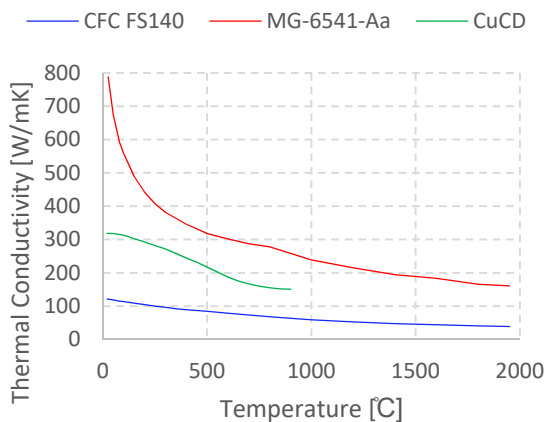


Figure 2: Thermal conductivity of CFC FS140, MG-6541-Aa and CuCD.

MoGr presents outstanding thermal transfer properties, as shown in Fig. 2, its conductivity is twice that of pure

copper in some grades. CFC shows in comparison lower thermal conductivity values.

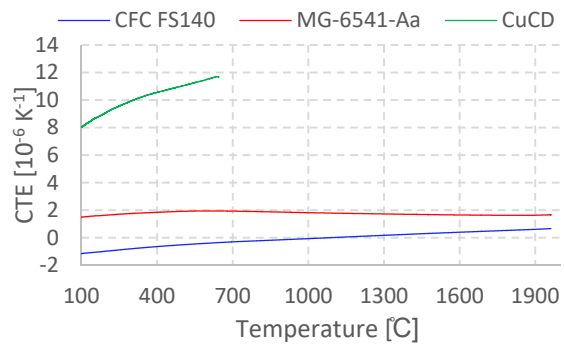


Figure 3: Coefficient of thermal expansion of CFC FS140, MG-6403-Ga and Copper Diamond

The excellent dimensional stability presented by MoGr and CFC up to 2000°C is displayed in Fig. 3. In the case of CuCD, the presence of diamond maintains the CTE under 12·10⁻⁶ K⁻¹.

Table 2 summarizes the thermo-physical and mechanical characteristics for the three materials:

Table 2. CFC FS140, MG-6403-Ga and Copper Diamond Thermo-Physical and Mechanical Properties Summary

Material	CFC FS140	MG-6541-Aa	CuCD
Density ρ [g/cm ³]	1.8	2.5	5
CTE α _L (RT to 1000° C) [10 ⁻⁶ K ⁻¹]	0	1.9	11
CTE α _I (RT to 1000° C) [10 ⁻⁶ K ⁻¹]	10	11	
Thermal Conductivity λ _L (RT) [Wm ⁻¹ K ⁻¹]	124	788	318
Thermal Conductivity λ _I (RT) [Wm ⁻¹ K ⁻¹]	42	50	
Electrical Conductivity ρ _L (RT) [MSm ⁻¹]	0.12	0.975	-
Electrical Conductivity ρ _I (RT) [MSm ⁻¹]	0.009	0.063	
Young's Modulus E (Flexural)[GPa]	74	60	67
Ultimate Strength Rm (Flexural)[MPa]	165	63	100

CONCLUSIONS

The materials characterization constitutes one of the axe of the development and exploration of novel or advanced materials for beam intercepting devices, and calls for broad variety of tests, ranging from the thermo-physical and mechanical tests described in this paper to high strain rate mechanical testing [15], long term irradiation campaigns [16] and tests under direct beam impacts at CERN's HiRadMat Facility [17].

So far CuCD (for lower temperature applications) and MoGr showed the most promising results. This good behaviour was confirmed by HiRadMat-23 experiment in 2015 [18].

REFERENCES

- [1] E. Quaranta *et al.*, “Towards Optimum Material Choices for the HL-LHC Collimator Upgrade”, in *Proc. IPAC'2016*, Busan, Korea, 2016, WEPMW031.
- [2] F. Carra *et al.*, “Mechanical Engineering and Design of Novel Collimators for HL-LHC”, in *Proc. IPAC'14*, Dresden, Germany, 2014, MOPRO116.
- [3] S. Redaelli *et al.*, “Collimation Upgrades for HL-LHC”, LHC Performance Workshop, Chamonix, France, 2015
https://indico.cern.ch/event/448109/contributions/1942040/attachments/1218992/1781341/SRedaelli_2016-01-28_print.pdf
- [4] A. Bertarelli and S. Bizzarro, “A Molybdenum Carbide / Carbon Composite and Manufacturing Method”, International Patent WO/2015/062657, 08-May-2015.
- [5] M. Tomut, “Update on irradiation test results at GSI and correlations to material damage simulation activity”, presented at the EuCARD2-WP11 Annual Meeting, CERN, 31-Jan-2017.
https://indico.cern.ch/event/606734/contributions/2447021/attachments/1404149/2147923/Tomut_EuCARD2_ann-meeting_2017-CERN.pdf
- [6] M. Kitzmantel and E. Neubauer, “Innovative hybrid heat sink materials with high thermal conductivities and tailored CTE” in *Proc. SPIE 9346*, vol. 9346, 2015, pp. 934606-934606–11.
- [7] ASTM International, *ASTM B311-13. Standard Test Method for Density of Powder Metallurgy (PM) Materials Containing Less Than Two Percent Porosity*.
- [8] Deutsches Institut für Normung, *DIN 51007:1994-06. Thermal analysis; differential thermal analysis; principles*.
- [9] ASTM International, *E228-11(2016), Standard Test Method for Linear Thermal Expansion of Solid Materials with a Push-Rod Dilatometer*.
- [10] “Flash Method of Determining Thermal Diffusivity, Heat Capacity, and Thermal Conductivity,” *J. Appl. Phys.*, vol. 32, no. 9, pp. 1679–1684, Sep. 1961.
- [11] ASTM International, *ASTM E2585-09(2015), Standard Practice for Thermal Diffusivity by the Flash Method*.
- [12] ASTM International, *ASTM C1259-15, Standard Test Method for Dynamic Young's Modulus, Shear Modulus, and Poisson's Ratio for Advanced Ceramics by Impulse Excitation of Vibration*.
- [13] ASTM International, *ASTM C651-15, Standard Test Method for Flexural Strength of Manufactured Carbon and Graphite Articles Using Four-Point Loading at Room Temperature*.
- [14] ASTM International, *ASTM C695-15, Standard Test Method for Compressive Strength of Carbon and Graphite*.
- [15] M. Scapin, C. Fichera, F. Carra, and L. Peroni, “Experimental investigation of the behaviour of tungsten and molybdenum alloys at high strain-rate and temperature” *EPJ Web Conf.*, vol. 94, p. 01021, 2015.
- [16] M. Tomut *et al.*, “Heavy ion induced radiation effects in novel molybdenum-carbide graphite composite materials”, GSI scientific report APPA-MML-MR-14, Darmstadt, Germany, 2014.
- [17] A. Bertarelli *et al.*, “An experiment to test advanced materials impacted by intense proton pulses at CERN HiRadMat facility” *Nucl.Instrum.Meth.*, vol. B308, pp. 88–99, Aug. 2013.
- [18] F. Carra, “The HiRadMat 23 Experiments: results and analysis” presented at the EuCARD-2 WP11 Topical Meeting - Collimator Materials for Fast High Energy Deposition, University of Malta, 28-Apr-2016. Contribution 6.