# DESIGN DEVELOPMENT AND TECHNOLOGICAL R&D FOR NIOBIUM-CLADDED BEAM PRODUCTION TARGETS

T. Griesemer\*, R. Franqueira Ximenes<sup>†</sup>, G. Arnau-Izquierdo, I. Aviles Santillana, M. Calviani,
 L. Salvatore Esposito, A. Gallifa Terricabras, G. Mazzola, R. Mena Andrade, A. T. Perez Fontenla,
 European Organization for Nuclear Research (CERN), Geneva, Switzerland
 M. Cusworth, T. Dutilleul, W. Kyffin, Nuclear AMRC, Rotherham, United Kingdom

### Abstract

High power particle producing target components in research facilities often consist of refractory metals. They experience challenging thermo-mechanical conditions and therefore require dedicated cooling systems. Employing water-cooling in direct contact with the target materials, especially tungsten (W), induces erosion and corrosion. Cladding the target blocks with erosion/corrosion-compliant materials is also a solution for a reliable heat transfer from the core materials to the coolant. Tantalum (Ta) is used in various facilities as cladding due to its corrosion resistance, outstanding thermo-mechanical properties, and diffusion bonding compatibility. The Beam Dump Facility (BDF) - a new proposed fixed target experiment at CERN - explored at first Ta2.5W cladding for molybdenum-based alloy TZM and pure W blocks. However, Ta presents non-negligible decay heat and high price. In this study, niobium-based materials - pure Nb, Nb1Zr, and Nb10Hf1Ti (C-103 alloy) - are evaluated as an alternative for cladding. The niobium alloys are assessed by their diffusion bonding via Hot Isostatic Pressing (HIP) and by thermo-mechanical characterization of the interfaces. Simulations of the impact with a high-power proton beam complement the study.

## **INTRODUCTION**

The Beam Dump Facility (BDF) is a new proposed fixed target experiment at CERN which is part of the "Search for Hidden Particles (SHIP)" collaboration [1]. A high energy proton beam of 400 GeV/c, with  $4 \times 10^{13}$  protons per pulse and a projected cumulated of  $4 \times 10^{19}$  protons on target (POT) per year is foreseen. The average power deposited in the target is around 305 kW. The physics requirements demand a target material with a high density, a high atomic and mass number, and a short nuclear interaction length. In addition, these materials face challenging thermo-mechanical conditions and thus need superior thermo-mechanical material properties and dedicated water-cooling. As a result, the chosen materials are TZM, a molybdenum alloy with titanium and zirconium, and pure tungsten (W) [2]. However, tungsten undergoes erosion, corrosion, and embrittlement when in direct contact with water [3]. Therefore, tantalum (Ta) has been selected to clad the core materials, as employed in other facilities such as KENS [4], LANSCE [5], and ISIS [6]. Based on the described design, a BDF target

prototype was manufactured and irradiated in 2018 [7, 8]. Loss-of-cooling-accident studies expressed concerns due to the high decay heat created by the Ta cladding [9]. These findings initiated the research of an alternative cladding material despite having a solid baseline design. This paper is assessing niobium (Nb) alloys by thermo-mechanical simulations with a high-energy beam impact. In addition, the diffusion bonding capabilities are studied and compared to a previously done cladding study with Ta [10].

## **CLADDING MATERIALS**

Niobium was selected as an alternative cladding material due to its bonding compatibility with W and molybdenum (Mo) [11]. Ta and Nb show identical diffusivity rates and full solubility towards W and Mo [12]. Besides presenting ductile behavior, both refractory metals have outstanding thermo-mechanical properties. In addition, Nb has under BDF operational conditions less activation as well as lower decay heat than Ta. Three different Nb alloys were assessed: (1) pure Nb, (2) Nb1Zr, and (3) Nb10Hf1Ti (C-103). Nb1Zr consists of 1 % zirconium and its mechanical properties are higher than pure Nb. C-103, 10 % hafnium and 1 % titanium, is known for its remarkable mechanical properties.

# THERMO-MECHANICAL CALCULATIONS

## Finite Element Model (FEM)

Calculations were run to compare the Nb alloys with Ta2.5W, a tantalum alloy with 2.5 % tungsten, cladding for the most critical TZM - Block 4 - and W - Block 14 - blocks of the BDF target. The cladding has a thickness of 1.5 mm, the block diameter is 250 mm, and the thickness is 25 mm for Block 4 and 50 mm for Block 14. The energies deposited on the target blocks were calculated with FLUKA Monte Carlo simulations [13], and imported into ANSYS® Mechanical<sup>™</sup> [14] for thermo-mechanical finite element analyses (FEA). Figure 1 illustrates for all cladding materials the maximum deposited energy density per cycle in longitudinal direction for the BDF target. It is noticeable that the deposited energies in the niobium-cladded targets are similar. The shown energy depositions were used to calculate the steady-state condition followed by a transient thermal simulation of three beam impacts from steady-state temperature. At the end, structural simulations were performed using the computed temperature distribution of the 3rd pulse as an input to calculate the thermal-induced stresses. All

<sup>\*</sup> tina.griesemer@cern.ch

<sup>&</sup>lt;sup>†</sup> rui.franqueira.ximenes@cern.ch

FEA models considered the same thermal (forced convection) and structural boundary conditions (spatial constraints with no residual stresses). The material properties of the Nb alloys were taken from literature [15] while the material models of Ta2.5W, TZM, and W were built from internal material characterization studies.



Figure 1: Maximum deposited energy density per cycle for different cladding materials in the BDF target.

### FEM Simulation Results

As illustrated in Fig. 1, the energy deposition is lower in niobium than in the tantalum cladding which correlates to the thermal results of Block 4. The Ta2.5W cladding reaches a maximum temperature of 170 °C while Nb, Nb1Zr, and C-103 reach 135 °C. Therefore, the maximum temperature of the Ta2.5W-cladded TZM is slightly higher than the niobium-cladded TZM (see Fig. 2). The equivalent (von-Mises) stresses correlate with the observed maximum temperatures. As a result of the lower energy deposition in the niobium cladding of block 1 to 13, more energy is deposited in the W core material of Block 14. Therefore, the maximum operational temperatures of the niobium-cladded W cores are slightly higher, 10 °C, than the tantalum-cladded W block. This results in higher equivalent (von-Mises) stresses in the niobium-cladded W core. However, the stresses in the Ta2.5W cladding are slightly higher than in the niobium, mostly driven by the higher Young Modulus of tantalum.

The safety margins of Ta2.5W, Nb, Nb1Zr, C-103, and TZM are derived from the von-Mises yield criterion. W is considered brittle during operational temperatures, thus it is advised to use the Christensen criterion [16]. Since W is lacking in compressive strength data under operational conditions, the maximum-normal-stress criterion has been used. Table 1 is showing the highest equivalent (von-Mises) stress  $\sigma_{vm}$ , maximum principal stress  $\sigma_1$ , yield strength  $\sigma_y$ ,



Figure 2: Maximum temperature in the longitudinal (Z) axis for different cladding materials in Block 4.

ultimate tensile strength (UTS), and the resulting safety factors for each material combination. The W core has limited safety margins for all cladding options due to the low UTS of 142 MPa determined from sintered W with high porosity which was also used for the BDF target prototype in 2018. In contrast, higher values of 330 MPa [2] are expected from literature for sintered and HIPed W.

Nb, Nb1Zr, and C-103 cladding, including the core materials, showed similar thermal and structural behavior: temperatures, equivalent (von-Mises) stresses, maximum principal stresses, and minimum principal stresses despite having different FEA input parameters. Nevertheless, the yield strength of the niobium alloys differ significantly. For instance, the safety margin for pure Nb is too small, therefore it is not recommended as a cladding alternative for the BDF target. Nb1Zr and C-103 have sufficient safety margins and the safety factors for C-103 cladding is even higher than Ta2.5W.

Table 1: Safety Factors of Different Cladding Materials for the Most Critical BDF Blocks 4 and 14

Block Nr.	Cladding / Core	$\sigma_{\rm vm}, \frac{\sigma_1}{[{ m MPa}]}$	σ <sub>y</sub> , <u>UTS</u> [MPa]	Safety Factor
4	Ta2.5W / TZM	127 / 123	227 / 460	2/4
	Nb / TZM	79 / 120	149 / 460	2/4
	Nb1Zr / TZM	72 / 121	170 / 460	2.5/4
	C-103 / TZM	63 / 121	254 / 460	4/4
14	Ta2.5W / W	80 / <u>96</u>	227 / <u>142</u>	3/1.5
	Nb / W	70 / <u>120</u>	149 / <u>142</u>	2/1
	Nb1Zr / W	72 / <u>117</u>	170 / <u>142</u>	2.5 / 1
	C-103 / W	71 / <u>116</u>	254 / <u>142</u>	3.5 / 1

## **DIFFUSION BONDING OF PROTOTYPES**

#### Prototype Design

The previous study on Ta cladding was used as a reference to compare the diffusion bonding capabilities of the Nb alloys with TZM and W. Therefore, the same prototype geometry and diffusion bonding procedure was applied in the Nb alloy cladding study. The prototype consists of four components: a cylindrical core material and three cladding parts, a tube and two cylindrical discs. They have a diameter of 26 mm, a height of 50 mm, and a cladding thickness of 1 mm (cylindrical) and 10 mm (top and bottom). The material combination of each prototype is shown in Table 2. During the study, material quality issues occurred in the Nb-cladded prototypes 1 - 4, thus 15 - 18 were added subsequently. In the Ta study, a 50  $\mu$ m thick Ta foil was used as a diffusion interfacial aid which has shown improved diffusion bonding results [10]. Therefore, the same Ta foils were utilized at the bottom of each prototype.

#### Methods

To achieve diffusion bonding between the cladding and core materials, Hot Isostatic Pressing (HIP) was used. The following steps were considered for the fabrication of the

Table 2: Material Combinations and Leak Test Results

Nr.	Cladding Material	Core Material	Leak Test
1	Nb	W	Failed
2	Nb	W	Failed
3	Nb	TZM	Passed
4	Nb	TZM	Failed
5	Nb1Zr	W	Passed
6	Nb1Zr	W	Passed
7	Nb1Zr	TZM	Passed
8	Nb1Zr	TZM	Passed
9	C-103	W	Passed
10	C-103	W	Passed
11	C-103	TZM	Passed
12	C-103	TZM	Passed
13	Та	W	Passed
14	Та	W	Passed
15	Nb	W	Passed
16	Nb	W	Passed
17	Nb	TZM	Passed
18	Nb	TZM	Passed

HIPed prototypes: (1) The four prototype components had a tight fit to each other. (2) The cladding tube and discs were joined by electron-beam welding (EBW) with a Pro-beam K25 [17]. The top was welded, cooled down over night, and then the same was repeated at the bottom. The welding chamber had a vacuum below  $6 \times 10^{-4}$  mbar and the parameters were: a voltage of 80 kV, a current of 40 mA, a welding speed of 40 mm/s, and beam deflection was used instead of mechanical movement. (3) To confirm the welding quality, helium leak testing was performed. The procedure is based on the standard BS EN 13185:2001 [18] and the leak detector ASM340 was used. The prototypes were impregnated in a bombing vessel for 20 h with 4 bar. After the removal from the vessel, they rested for 20 min and were tested individually in a vacuum chamber with a leakage sensitivity of  $1 \times 10^{-8}$  mbar l/s. (4) Afterwards, the prototypes were HIPed in two cycles with the same pressure, 200 MPa, and dwell time, 3 h but different temperatures. The first cycle was run with the odd numbered prototypes at 1200 °C and the second with the even numbered ones at 1400 °C. While the heating rate of the second cycle was higher, 6.3 K/min and 7.6 K/min, the cooling rates were almost similar, -8.7 K/min and -8.5 K/min. (5) After HIPing, ultrasonic testing (UT) by immersion was performed from all sides to confirm successful bonding. For each material combination, a reference body was used which was a not welded and not HIPed prototype. (6) Finally, they were cut longitudinally and the bonding interface was investigated by optical microscopy.

## Diffusion Bonding Results

The prototypes were firstly EB welded and afterwards leak tested with helium. The results are presented in Table 2. While only prototypes 1, 2, and 4 failed the leak test, all Nb discs of 1 - 4 showed quality defects originated from the cladding supplier's manufacturing process. Therefore, the prototypes 15 - 18 were added to the study and the mate-

rial defects in the cladding of 1 - 4 were patched by local welding. All prototypes passed the UT test except prototype 1 (see Fig. 3). The UT results show discontinuity as red while blue represents continuity at the bonding interfaces. After being cut in two halves, prototype 1 showed no signs of bonding as cladding and core material fell apart instantly. The failed prototypes 1, 2, and 4 were not leak tested again therefore it is possible that the cracks were not closed during the weld patching. This would allow pressure to enter the area between the core and the cladding material and thus counteract the pressure applied from the HIPing furnace. The other prototypes did not separate after cutting and the bonding interfaces were inspected by optical microscopy. In general, the bonding interfaces showed no signs of detachment between cladding, core, and Ta foil (see Fig. 4) and the Nb alloys demonstrated enough ductility to deform with the chosen HIPing parameters.



Figure 3: UT testing results (C-scans) of the not-HIPed reference body *Ref 1* and the HIPed prototypes 1 and 3.



Figure 4: Optical microscopy of the prototype 15.

## CONCLUSIONS

The thermo-mechanical calculations showed the potential of using Nb alloys as cladding material for the BDF target. The calculated stresses for the cladded Nb, Nb1Zr, and C-103 blocks had alike stress distributions despite having different FEA input parameters. Nevertheless, the Nb alloys have varying yield strengths which results in different safety factors. The safety margins of the Nb1Zr-cladded blocks are comparable and the ones of C-103 are even better than Ta2.5W. Furthermore, the niobium-cladded prototypes showed no signs of detachment when inspected by optical microscopy. Prototype 1 also allowed some interesting conclusions about the applied diffusion bonding process. Firstly, the UT procedure is well suited to identify not bonded interfaces after the HIPing process. Secondly, it is important to achieve a leak tight weld between the cladding components (tube and discs) during the EB welding.

It is foreseen to assess the bonding interfaces of the prototypes further by extracting specimens in the interface area. The specimens will be used for tensile and thermal diffusivity tests to characterize the different material combinations. Moreover, if the BDF facility is approved a new reducedscale BDF target prototype with new cladding materials would be designed, manufactured, and tested with beam during 2024 - 2026.

# REFERENCES

- C. Ahdida *et al.*, "SPS Beam Dump Facility-comprehensive design study," *arXiv preprint*, 2019. doi:10.48550/arXiv.1912.06356
- [2] E. Lopez Sola *et al.*, "Design of a high power production target for the Beam Dump Facility at CERN," *Physical Review Accelerators and Beams*, vol. 22, no. 11, p. 113 001, 2019. doi:10.1103/PhysRevAccelBeams.22.113001
- [3] R. Lillard, D. Pile, and D. Butt, "The corrosion of materials in water irradiated by 800 MeV protons," *Journal of Nuclear Materials*, vol. 278, no. 2-3, pp. 277–289, 2000. doi:10.1016/S0022-3115(99)00248-2
- [4] M. Kawai, K. Kikuchi, H. Kurishita, J.-F. Li, and M. Furusaka, "Fabrication of a Tantalum-clad Tungsten target for KENS," *Journal of Nuclear Materials*, vol. 296, no. 1-3, pp. 312–320, 2001.
  - doi:10.1016/S0022-3115(01)00533-5
- [5] A. Nelson, J. O'Toole, R. Valicenti, and S. Maloy, "Fabrication of a Tantalum-clad Tungsten target for LANSCE," *Journal of Nuclear Materials*, vol. 431, no. 1-3, pp. 172–184, 2012. doi:10.1016/j.jnucmat.2011.11.041
- [6] A. Dey and L. Jones, "Strategies to improve ISIS TS2 target life," *Journal of Nuclear Materials*, vol. 506, pp. 63–70, 2018. doi:10.1016/j.jnucmat.2017.12.044
- [7] E. Lopez Sola *et al.*, "Beam impact tests of a prototype target for the beam dump facility at CERN: Experimental setup and preliminary analysis of the online results," *Physical Review Accelerators and Beams*, vol. 22, no. 12, p. 123 001, 2019. doi:10.1103/PhysRevAccelBeams.22.123001
- [8] R. F. Ximenes *et al.*, "CERN BDF Prototype Target Operation, Removal and Autopsy Steps," in *Proc. IPAC'21*, Campinas, Brazil, May 2021, pp. 3559–3562. doi:10.18429/JACoW-IPAC2021-WEPAB365

- [9] R. Mena Andrade, R. Franqueira Ximenes, and M. Calviani, "Loss-of-coolant-accident-study for the Beam Dump Facility at CERN," NURETH-19 Conference, Brussels, Belgium, 2022.
- [10] J. Busom Descarrega *et al.*, "Application of hot isostatic pressing (HIP) technology to diffusion bond refractory metals for proton beam targets and absorbers at CERN," *Material Design & Processing Communications*, vol. 2, no. 1, e101, 2020. doi:10.1002/mdp2.101
- [11] W. Basuki and J. Aktaa, "Investigation of Tungsten/EUROFER97 diffusion bonding using Nb interlayer," *Fusion Engineering and Design*, vol. 86, no. 9-11, pp. 2585– 2588, 2011. doi:10.1016/j.fusengdes.2011.03.017
- [12] R. Franqueira Ximenes *et al.*, "Beam Dump Facility production target at CERN and advanced cladding technological R&D," 15th International Workshop on Spallation Materials Technology, Santa Fe, USA, 2023.
- [13] C. Ahdida *et al.*, "New capabilities of the FLUKA multipurpose code," *Frontiers in Physics*, p. 705, 2022. doi:10.3389/fphy.2021.788253
- [14] Ansys<sup>®</sup>, Ansys Mechanical Enterprise, version 2020 R2.
- [15] JAHM Software, Inc., *Material Property Database (MPDB)*, version 8.87.
- [16] R. M. Christensen, "A comprehensive theory of yielding and failure for isotropic materials," *Journal of Engineering Materials and Technology*, vol. 129, no. 2, pp. 173–181, 2007. doi:10.1115/1.2712847
- [17] Nuclear AMRC, Welding and materials R&D, https:// namrc.co.uk/capabilities/innovation/welding/, Last accessed on 2023-05-02, 2023.
- [18] Non-destructive testing. Leak testing. Tracer gas method, BS EN 13185:2001, 2001. doi:10.3403/02218833