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Tungsten Alloy Development as Advanced Target Material for High-Power Proton Accelerator

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Tungsten (W) is a principal candidate as target material because of its high density and extremely high melting point. W inherently has a critical disadvantage of its brittleness at around room temperature (low temperature brittleness), recrystallization embrittlement, and irradiation embrittlement. TFGR (Toughened, Fine Grained, Recrystallized) W-1.1%TiC has been considered as a realized solution to the embrittlement problems. We started to fabricate TFGR W-1.1%TiC in 2016 under collaboration between KEK and Metal Technology Co., LTD (MTC). The TFGR W-1.1%TiC samples were successfully fabricated in June, 2018. As a result, the specimen showed slight bend ductility and 2.6 GPa of fracture strength. The TFGR W-1.1%TiC was included in the HRMT-48 PROTAD experiment on September 28th, 2018. After cooling, the post-irradiation-examination will be conducted for the irradiated TFGR W-1.1%TiC.

KEYWORDS: Proton Accelerator Target, Tungsten, Recrystallization embrittlement, Irradiation embrittlement,

1. Introduction

Muon and/or neutron, hereafter muon/neutron, are produced by collision of high-energy proton against the target material at proton accelerator facilities. The muon/neutron have been widely used for various fields of condensed matter physics and fundamental nuclear and particle physics. So far, the muon/neutron facilities were constructed at large proton accelerators to obtain intense muon/neutron beam due to the demands of the physics research. Nevertheless, opportunities to experiment the muon/neutron physics are limited because construction and upgrade of the megawatt-class high-power proton accelerators cost enormous budgets. So, demands of efficient muon/neutron production are increasing to promote the muon/neutron science. Muon/neutron produced on the target at the primary proton beamline are transported to the experimental area. The position of the head component that is a capture magnet in the case of muon beamline or a moderator in the case of neutron beamline dominantly determine the acceptance of muon/neutron transport. Figure 1 demonstrates a schematic view of muon capture and transport to experimental area. Then, the density of the target material should be higher, because a smaller spatial volume of the muon/neutron production on the target is beneficial to more efficient transport to downstream experiments. However, it must be taken into account that higher density leads to denser beam loss and higher radiation damage on the target material.

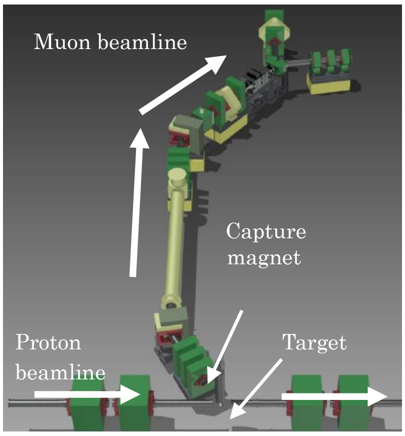

Fig. 1. Schematic view of muon capture and transport to experimental area. The acceptance of muon beam is determined by the position of the capture magnet of the muon beamline against the target in the primary proton beamline.

Tungsten (W) is a principal candidate as target material because of its high density (19.3 g/cm^3) and extremely high melting point (3420 ºC). The use of W leads to provide 10 times higher brightness of muon/neutron than that of the current target materials. Actually, the W target is considered to be used in the upcoming projects such as COMET Phase 2 and MLF 2nd Target Station at J-PARC, Mu2e at Fermilab, SNS 2nd Target Station at ORNL, and neutron target at ESS. While expected as the target material for the proton accelerator, W inherently has a critical disadvantage of its brittleness at around room temperature (RT) (low temperature brittleness). The low temperature brittleness can be avoided by heavy plastic working, although the working can be sufficiently applied only to filaments or hot-rolled thin plates and its effect also depends on the working direction. However, even if the brittleness is alleviated, W exhibits significant embrittlement due to recrystallization that occurs when W is heated at and above the recrystallization temperature, which is almost one-third of the melting point (recrystallization embrittlement). Moreover, it exhibits significant embrittlement by proton irradiation as well (irradiation embrittlement). Extensive efforts have been made to develop W materials that exhibit enhanced resistances to these types of embrittlement all over the world [1]. This article will address our methodology to surmount the shortcomings of the conventional W materials and focus on prospective outcomes from the applications of a TFGR (Toughened, Fine Grained, Recrystallized) tungsten alloy, W-1.1%TiC, to proton-accelerator targets.

2. Toughened, Fine Grained, Recrystallized Tungsten alloy, W-1.1%TiC

2.1 History of TFGR W-1.1%TiC

TFGR W-1.1%TiC has been considered as a realized solution to the embrittlement problems and had been developed at Tohoku University till 2013 [2, 3]. TFGR W-1.1%TiC exhibits grain boundary reinforced nanostructures containing a high density of effective sinks for irradiation-induced point defects, a DBTT (Ductile-to-Brittle Transition Temperature) down to around RT and enhanced resistances against surface damages by thermal shock/fatigue in the recrystallized state [2, 3]. Thus, while TFGR W-1.1%TiC seems to be an ideal material as the proton-accelerator target, its development is still on the way. The material is fabricated via a powder metallurgical route consisting of MA (Mechanical Alloying), HIP (Hot Isostatic Pressing) and GSMM

(Grain boundary Sliding based Microstructural Modifications) [2]. Currently, the available size is about 30 mm x 30 mm x 10mm. In addition, the entire route is so time-consuming that the available quantity is insufficient for the target applications. After all, the technology to fabricate TFGR W-1.1%TiC was not transferred to the next generations at Tohoku University because a lot of difficulties were anticipated to enlarge the size and to realize an economical way of fabrication in future.

2.2 Present status of development for TFGR W-1.1%TiC under KEK-MTC collaboration

To revive the activities, we have been working on fabrication of TFGR W-1.1%TiC and/or more improved W materials with sufficient dimensions for the target applications and investigation of their feasibility as the target materials since 2016 under collaboration between KEK and MTC (Metal Technology Co., LTD) [4].

Figure 2 shows the process to fabricate TFGR W-1.1%TiC. At first, a glove box for handling with a high-purity level and a three-dimensional ball milling apparatus were specially fabricated for production of TFGR W-1.1%TiC. The raw powders of tungsten and titanium carbide are handled inside the glove box, which can achieve a highly-pure argon atmosphere with less than 1 ppm of oxygen concentration. It can also be employed to exclude gas-impurities such as oxygen and nitrogen from the raw powder materials in a vacuum chamber and a tube furnace for heating up to 1000 ºC. The purified powders and MA balls are loaded in a MA vessel. The raw powders are mechanically alloyed and are ultrafine-grained in the ball-milling apparatus through three-dimensional oscillations. In the past developments at Tohoku University, it was proved that existence of argon bubbles caused the origin of cracks. Therefore, the fabrication process in the glove box and the ball milling apparatus was performed in the hydrogen atmosphere, because hydrogen is diffused and excluded from TFGR W-1.1%TiC in the subsequent HIP and GSMM process. Furthermore, hydrogen avoids the stick of the powders on the MA balls and the MA vessel due to its high thermal conductivity. Thus, while the use of hydrogen is beneficial, the measures against the combustibility of hydrogen must be considered carefully. In the current developments, on the other hand, the powders are mechanically alloyed not in a hydrogen atmosphere but in vacuum to simplify the process and the glove box. To realize the MA process in vacuum, the MA vessel is remotely filled with the raw powders in the vacuum chamber equipped beside the glove box. In addition, the cooling ability of the current ball milling apparatus is reinforced by a refrigerator in the MA process, because thermal conduction in the hydrogen atmosphere in the past development

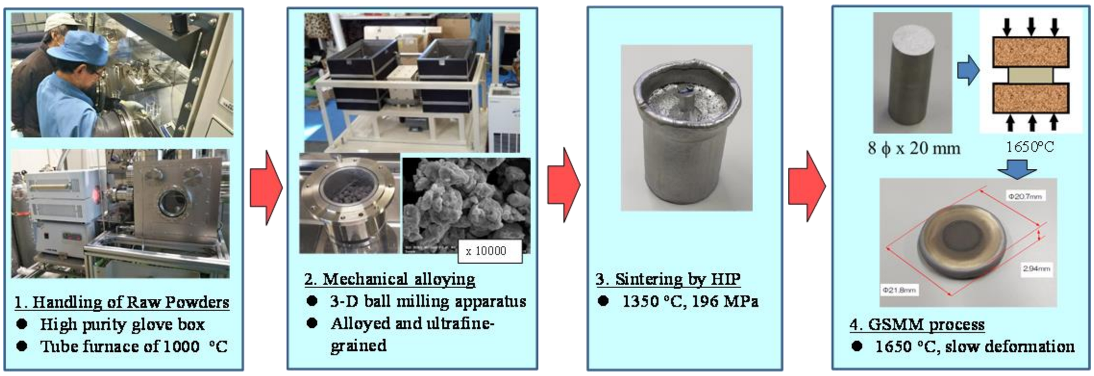

Fig. 2. Process to fabricate TFGR W-1.1%TiC

cannot be expected. Currently the entire MA vessel is included inside the refrigerator at -2 ºC, while the MA vessel was cooled by the supply of air at 5 ºC in the past developments. Consequently, the concentrations of oxygen and nitrogen after the MA process were suppressed to 910 ppm and 90 ppm respectively. They approximately correspond to 10 % and 5 % respectively, compared with ones in a typical glove box without a baking process. Then, the mechanically-alloyed powders are sintered in HIP process at 1350 ºC, 196 MPa for 3 hours. Several specimens, with 8 mm in diameter and 20 mm in thickness, can be obtained from the sintered bulk material through wire-cut electrical discharge machining. Each specimen is slowly deformed at 1650 ºC by a hot press, i.e. with a GSMM (Grain boundary Sliding based Microstructural Modifications) process. Then TFGR W-1.1%TiC samples can be obtained with the size of about 20 mm in diameter and about 3 mm in thickness. TFGR W-1.1%TiC samples were successfully fabricated in June, 2018. The mechanical properties of TFGR W-1.1%TiC were measured through three-points bending tests. As a result, the specimen showed slight ductility and 2.6 GPa of fracture strength. While the result is encouraging, the property is still inferior to the one fabricated in the past development at Tohoku University. The cause of the inferiority will be thoroughly investigated in each of the fabrication processes.

2.3 Thermal shock experiment, HRMT-48 at CERN-HiRadMat Facility

Highly-cyclic and volumetric heating with extremely short-time width is induced by a high-energy and pulsed proton beam. The temperature of the beam spot increases and the heated volume is constrained by the surroundings with low temperature. Then, compressive stress causes propagation of the displacement and consequent tensile stress takes place. The wave propagation gives severe damages against the target material. The HiRadMat Facility at CERN [5] is dedicated to study of the impact of intense pulsed beams on materials. TFGR W-1.1%TiC was included in the HRMT-48 PROTAD experiment on September 28th, 2018. The beam energy, time width of one bunch, and proton number in one bunch were 440 GeV, 25 nanoseconds, and 8.15×10^{11} respectively. The beam radius was $\sigma_x=\sigma_y=1$ mm. 14 bunches irradiated the target materials for 0.4 μsec. After cooling, the post-irradiation-examinations will be conducted for the irradiated TFGR W-1.1%TiC.

3. Conclusion

While tungsten (W) is a principal candidate as target material, W inherently has a critical disadvantage of its brittleness at around room temperature (low temperature brittleness), recrystallization embrittlement, and irradiation embrittlement. TFGR (Toughened Fine Grained, Recrystallized) W-1.1%TiC has been considered as a realized solution to the embrittlement problems. We started to fabricate TFGR W-1.1%TiC in 2016 under collaboration between KEK and MTC (Metal Technology Co., LTD). TFGR W-1.1%TiC fabricated in June, 2018 showed slight bend ductility and 2.6 GPa of fracture strength. Then TFGR W-1.1%TiC was included in the HRMT-48 PROTAD experiment on September 28th, 2018.

Acknowledgment

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