Production of Zircaloy-4 Powder using Scraps and its 3D Printing Application

Joo-Young Ryu*, Han-Gil Woo, Dae-Woon Choi, Chae-Young Nam, Jin-Seok Lee, Sang-Youn Jeon
KEPCO Nuclear Fuel Co., 242, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, 34057, Republic of Korea
**Corresponding author: jyryu@knfc.co.kr*

1. Introduction

KEPCO Nuclear Fuel (KNF) is considering the possibility of applying 3D printing technology to nuclear fuel components. It is officially called as additive manufacturing (AM) [1]. This technique allows to make desired shape with complex geometries using feed materials. Most raw materials such as zirconium powder or bars currently depend on the overseas market. Moreover, additional costs are incurred in the manufacturing unit price due to the addition of transportation costs and periods from abroad. On the other hand, if scraps generated in the process of manufacturing nuclear fuel components are recycled to zirconium alloy bars, it can be useful as a raw material for 3D printing metal powder or end plugs for fuel rod. It might have effects of import substitution and cost reduction. In particular, since KNF is carrying out development of a spacer grid using 3D printing technology, the demand for zirconium powder is expected to continue from now on. Therefore, the possibility of manufacturing Zircaloy-4 (Zry-4) bar using scraps is confirmed and there is no abnormality in quality of produced powder, economical competitiveness of raw materials and components of KNF can be strengthened.

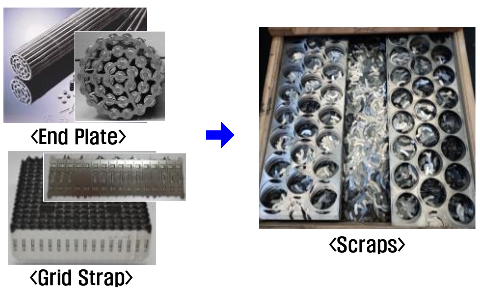


Fig. 1. Zirconium alloy scraps after sheet metal working

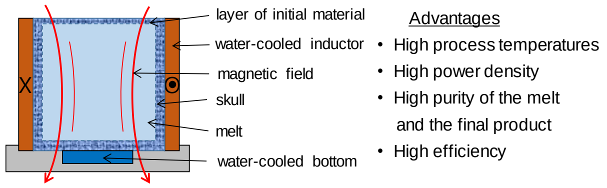
Fig. 1 shows an example of scraps generated in the manufacturing process of pressurized water reactor and heavy-water reactor fuel components. The amount of scraps generated during nuclear fuel manufacturing in 2011 was reported to be about 50 tons[2].

In this study, KNF casted Zircaloy-4 bars using scraps, and then produced Zircaloy-4 powder for powder bed fusion (PBF) [3] of 3D printing. Finally, small tensile specimens in each direction (X, Y, 45º, Z) were manufactured. These specimens will be tested for mechanical properties after neutron irradiation in the near future. The overall flow of manufacturing from scrap to specimens is summarized as follows.

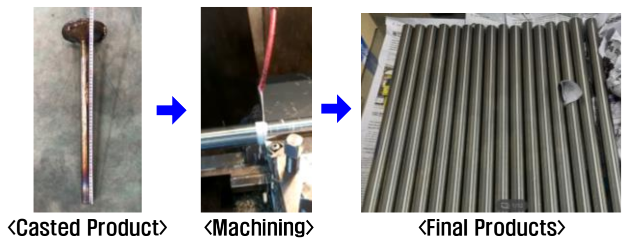
- Scrap → Bar → Powder → 3D printed bulks with each direction → Cutting into small tensile specimens

2. Production of Zircaloy-4 Bar

The Zircaloy-4 material belongs to the reactive metal group and has a melting point of about 1850ºC, which is not suitable for general melting facilities, but it is possible to work with the Induction Skull Melting (ISM) method process. The ISM technique uses a water-cooled crucible woven with copper tubes in a vacuum chamber, and a coil is installed to form high-frequency induced current so that the metal can be melted by enough heat from the induced current. The technology has been developed to dissolve metals with high melting points, and it has the advantage of obtaining a high purity metal. After the scraps are melted, the molten metal is solidified in a graphite mold to form a round bar shape. Fig. 2(a) is a schematic of the ISM process, and Fig. 2(b) shows the result of products of about 160 kg of Zircaloy-4 bars with Φ30mm, L500mm size that are made from scraps through ISM equipment.



(a) Schematic of ISM process [4]



(b) Zry-4 bars

Fig. 2. Zry-4 bars produced through ISM process

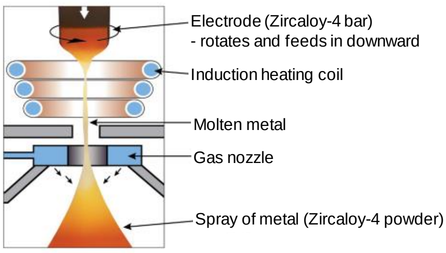
Sampling inspection for chemical composition was performed on scraps as raw materials and bars produced after casting based on ASTM B532 standard. The methods used in the inspections that are X-Ray Fluorescence spectrometer (XRF), Inductively Coupled Plasma-atomic emission spectroscopy (ICP), and inorganic contents test (C, H, N, O) were applied. Table I indicates the test results. It is clearly seen that the chemical composition is within the ASTM standard.

Table I: Chemical composition of Zry-4 bar [weight %]

Elements	Scrap(raw mat.)	Bar(as-cast)	Standard
Zirconium	98.3	98.1	Bal.
Tin	1.31	1.44	1.20 ~ 1.70
Iron	0.18	0.21	0.18 ~ 0.24
Chromium	0.11	0.09	0.07 ~ 0.13
Oxygen	0.12	0.12	0.09 ~ 0.16
Carbon	0.018	0.025	< 0.027
Silicon	-	0.005	0.005 ~ 0.012
Hydrogen	0.002	0.0005	< 0.0025
Nitrogen	0.001	0.002	< 0.0080

3. Production of Zircaloy-4 Powder

Zircaloy-4 powder is thermodynamically highly reactive and is highly likely to ignite or explode by reacting well with oxygen and hydrogen in the air. Thus, it is very hard to produce the powder, moreover, Zircaloy-4 powder for nuclear components should meet ASTM B352 specifications in respect of chemical composition and impurity limits. It is also required that the particle size shall be between 15~50μm and the particle shape shall be spherical to be used with PBF 3D printer. Among the producing powder technologies, electrode induction-melt inert gas atomization (EIGA) method is actually reasonable process to produce Zircaloy-4 powder. Fig. 3(a) shows the schematic of EIGA process. The bar is inserted into a circular coil and melted by heat generated by the induced current. After that, it is made into powder by high-pressure inert gas injection. Since the process is a non-contact method, it is suitable for the production of high-purity powder. Fig. 3(b) shows the amount of produced powder that is about 19 kg. The yield rate of powder production for PBF is at around 25%.



(a) Schematic of EIGA process [6]

(b) Zry-4 powder (for PBF)

Fig. 3. Zry-4 powder produced through EIGA process

Sampling inspection for powder properties was performed in terms of PBF powder standard. As a result of inspection, most of the evaluations of powder properties, chemical composition and powder characteristic data were satisfied as shown in Fig 4 and Table II, respectively. But silicon content is abnormal so that it is required to investigate why the content has exceeded the limit. However, since this study is in the experimental stage to explore producing powder from scraps, the overall results can be acceptable.

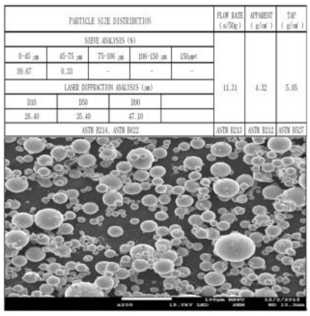


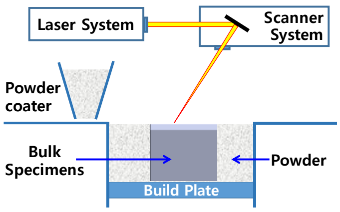
Fig. 4. Test results of Zry-4 powder (for PBF) property

Table II: Chemical composition of Zry-4 powder [weight %]

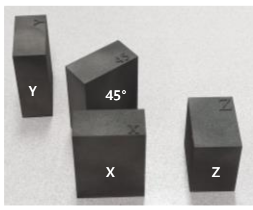
Elements	Powder	Standard
Zirconium	Bal.	Bal.
Tin	1.17	1.20 ~ 1.70
Iron	0.19	0.18 ~ 0.24
Chromium	0.084	0.07 ~ 0.13
Oxygen	0.11	0.09 ~ 0.16
Carbon	0.018	< 0.027
Silicon	0.096	0.005 ~ 0.012
Hydrogen	0.001	< 0.0025
Nitrogen	0.0002	< 0.0080

4. Production of Zircaloy-4 Tensile Specimen

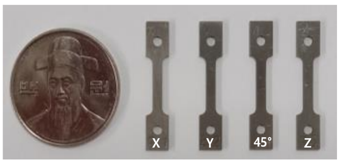
A tensile test plan was established to verify the mechanical properties of 3D printed products that were made from scraps. For this purpose, as shown in Fig. 5(a) and 5(b), Zircaloy-4 bulk specimens in each build-up direction were made by PBF method, and finally individual specimens were cut through electric discharge machining. The size of the tensile specimen is smaller than that of the general specimen as shown in Fig. 5(c), which is for the purpose of neutron irradiation test in the future.



(a) PBF process concept [3]



(b) Bulk specimens



(c) Small size of tensile specimens in each direction

Fig. 5. Zry-4 tensile specimens through 3D printing

5. Future Work

In the second half of this year, a neutron irradiation test with these specimens will be conducted at a research reactor of HANARO in Korea Atomic Energy Research Institute [7]. The number of specimens according to the build-up direction will be determined and these will be loaded into the capsule for irradiation.

In the future, after finishing neutron irradiation, these 3D printed specimens will be subjected to tensile test in the hot cell, and the results will be compared and analyzed with general wrought specimens.

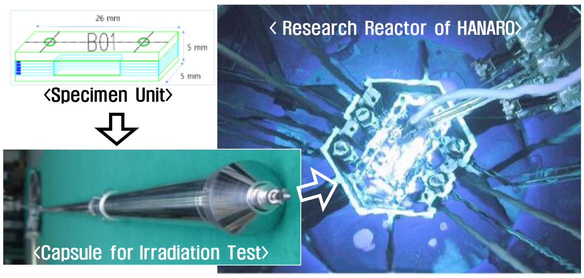


Fig. 6. Specimen unit and capsule for irradiation test [7]

6. Conclusions

KNF investigated a study to confirm the possibility of making Zircaloy-4 3D printing powder and bars to recycle scraps generated in the process of manufacturing of nuclear fuel components. For this purpose, Zircaloy-4 bars were casted using the ISM process, and Zircaloy-4 powder was produced using the EIGA process. The chemical composition and powder property inspection results show most of the data are almost satisfied. In the future, KNF plans to continue works that a neutron irradiation test for the specimen made from scraps will be conducted at a research reactor of HANARO and mechanical property experiments will be performed.

ACKNOWLEDGEMENTS

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