Transactions of the Korean Nuclear Society Autumn Meeting
Gyeongju, Korea, October 24-25, 2013

Study on Enhanced Thermal Conductivity of HfO_2/xGnP Composites

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1. Introduction

Graphene has high thermal conductivity that is good characteristic for heat transfer. The thermal conductivity of nuclear fuel is one of the most important issues in nuclear safety. In this paper, the object of the study is confirming that graphene can be applied as an advanced-material in nuclear reactors.

The commercial nuclear power industry is interested in advanced fuels. Requirements of advanced fuels are as follows: high thermal conductivity, chemical stability (low reactivity) and low manufacturing cost. Thermal conductivity of UO_2 fuel is very low. It is important parameter for optimum design and safety analysis of a nuclear system. This is because the thermal conductivity of fuels significantly affects the internal energy of the nuclear fuel by heat transfer through the coolant. [1]

Therefore, the safety analysis using fuel thermal conductivity is necessary for the prediction of thermal-hydraulic behavior [2]. If exfoliated graphene nanoplate (xGnP) is mixed in UO_2 fuel, it is chemically stable and its thermal conductivity will be enhanced. Advantages of xGnP are a high thermal conductivity and a low absorption cross section. In present work, HfO_2 is selected as a substitute of UO_2 due to the similar density of HfO_2 and UO_2. HfO_2/xGnP composites were made by adding 10 vol. % of xGnP having a high thermal conductivity and the thermal conductivity of HfO_2/xGnP composites were measured by LFA 447 NanoFlash.

2. Enhanced thermal conductivity and Peak cladding temperature

The thermal conductivity of UO_2 and graphene composites was predicted by MARS-KS (Multi-dimensional Analysis of Reactor Safety-Korean Standard). And the Maxwell model was used for theoretical thermal conductivity of UO_2 and graphene composites.

The Maxwell model is as follows:

$$k_{UO_2+Graphene} = k_{UO_2}\left[1+\frac{3(\frac{k_{Graphene}}{k_{UO_2}}-1)\times \text{Volume Fraction}}{(\frac{k_{Graphene}}{k_{UO_2}}+2)-(\frac{k_{Graphene}}{k_{UO_2}}-1)\times \text{Volume Fraction}}\right] \quad (1)$$

The materials for experiment were HfO_2 and xGnP powder instead of UO_2 and graphene. In this paper, the performace of experimental result performed in university laboratory level. The expected peak cladding temperature according to enhanced thermal conductivity was as shown in Fig. 1.

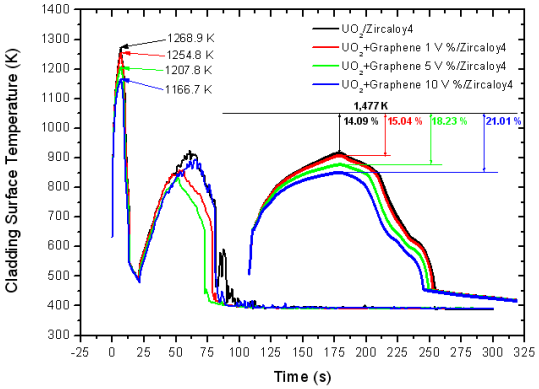


Fig. 1. Expected peak cladding temperature according to volume fraction of UO_2/Graphene.

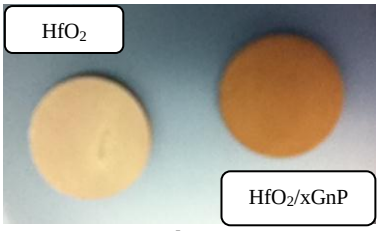
3. Experimental procedure

A total of 2 different powders (HfO_2, 44 microns, alfa aesar; xGnP, 15 microns, XG sciences) was prepared to make two composites that were pure HfO_2 and HfO_2/xGnP composite (10 vol. %). The composites was made by milling and mixing process.

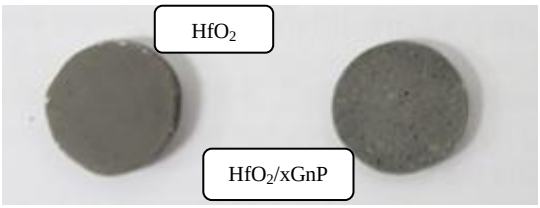
Two methods were used for fabricating the composite. Firstly, oxidative sintering was required to compact process for making pellet structure. And it needed to sintering process at high temperature. Sintered HfO_2 and HfO_2/xGnP composite pellet was fabricated as shown in Fig. 3. The other method for sintering is HIP (Hot Isostatic Pressing) that is pressure assisted method. It is possible to sinter ceramic powders. And the condition of HIP process was at high pressure and temperature during sintering process. Inner side of sintered can was vacuum condition and the material of can was stainless steel. Sintered can and sintered HfO_2 and HfO_2/xGnP composite pellet by HIP sintering were as shown in Fig. 2 and Fig. 3.

LFA 447 (Laser Flash Apparatus) for measurement of the thermal conductivity [3] of sintered composites was used in this paper so it was necessary to make proper disk pellet size (12.5~12.9 mm diameter × 2~4 mm thickness) for decreasing errors.

Fig. 2. Sintered can by HIP sintering



a. Oxidative sintering



b. HIP sintering

Fig. 3. Sintered HfO_2 and HfO_2/xGnP composite pellet by oxidative sintering and HIP sintering

4. Results and discussion

In this section, the results of thermal conductivity are presented. The data of each pellet was as shown in table 1 and 2. The density of composite was between 7.1~8.65 g/cm^3 and pure HfO_2 was more dense than HfO_2/xGnP composite pellet. It means that sintered HfO_2 was denser than HfO_2/xGnP composite.

Table. 1. The data of oxidative sintering pellets.

Oxidative sintering	Diameter (mm)	Height (mm)	Mass (g)	Density (g/cm^3)
HfO_2_a	12.68	2.04	2.149	8.342
HfO_2_b	12.69	2.03	2.152	8.381
HfO_2/xGnP_a	12.6	2.05	1.875	7.333
HfO2/xGnP_b	12.7	2.09	1.895	7.156

Table. 2. The data of HIP sintering pellets.

HIP sintering	Diameter (mm)	Height (mm)	Mass (g)	Density (g/cm^3)
HfO_2_c	12.62	2.25	2.433	8.65
HfO_2_d	12.6	3.3	3.104	7.54
HfO_2/xGnP_c	12.73	2.26	2.085	7.249
HfO2/xGnP_d	12.75	2.29	2.105	7.198

Although HfO_2 was compacter than HfO_2/xGnP in both sintering process, the result of thermal conductivity appeared particular difference. During the oxidative sintering, the composite reacted with oxygen. But the composite of HIP sintering was not reacted with oxygen in air. As a result, the volume ratio of xGnP of oxidative sintering in pellet was decrease but the volume ratio of xGnP of HIP sintering was maintained its ratio.

The thermal conductivity of sintered HfO_2 had similar values about 85 ~ 115 % regardless of sintering method. But HfO_2/xGnP pellet of oxidative sintering was different from pellet of HIP sintering. HIP pellet had the range of value 95 ~ 138 % and oxidative pellet had the range of value 40 ~ 50 %. Even though the range of HIP pellet overlapped 100℃, it showed the tendency of thermal conductivity of xGnP by HIP sintering was higher than sintered HfO_2. Fig. 4 showed the measured thermal conductivity result in this paper.

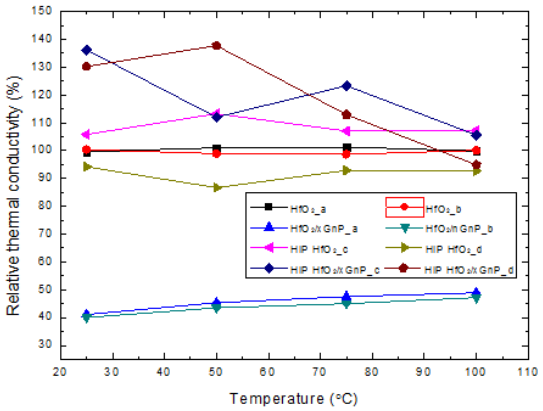


Fig. 4. The measured thermal conductivity result of HfO_2 and HfO_2/xGnP (10 vol. %) composite pellet

5. Conclusions

The hafnium oxide (HfO_2) – 10 vol. % exfoliated graphite nanoplate (xGnP) composite ceramics was produced by hot isostatic pressing (HIP) and oxidative sintering at high temperature. The result of thermal conductivity of HIP sintering was increased by adding xGnP powder. Oxidative sintering was decreased the thermal conductivity because of oxidation between oxygen and carbon in xGnP. But HIP sintering was not effect of oxidation due to vacuum condition in can. In this paper the result of thermal conductivity showed the possibility of enhanced fuel pellet.

REFERENCES

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