An Experiment on Preparation of Matrix Graphite Powder for HTR fuel element

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1. Introduction

Nuclear fuel for HTGR (High Temperature Gas Cooled Reactor) to produce nuclear hydrogen is called TRISO coated fuel, which consists of 500-㎛ spherical UO_2 particles coated with Pyrolytic Carbon(PyC) and SiC in four layers. The coated TRISO particles are mixed with matrix graphite powder and pressed into a spherical shape of about 60 mm in diameter or a cylindrical compact and heat-treated at about 1900oC. These fuel elements have a variety of sizes and forms depending on the types of nuclear reactors.

Basic steps of manufacturing the fuel element include preparation of graphite matrix powder, over-coating fuel particles, mixing fuel particles with matrix, carbonating green compact and the final high-temperature heat processing of the carbonated fuel compact. In order to develop fuel compact fabrication technology, it is important to develop a basic technology for matrix graphite powder (MGP) preparation which has strong influence on the material properties of fuel element. [1,2] In this work, an experiment was attempted by mixing natural and artificial graphite powders, kneading with binder in methanol medium, drying and milling to prepare a simulated matrix graphite powder with proper characteristics for further steps, i.e., further mixing with coated particles, compaction and heat treatment.

2. Experimental

In this experiment, with reference to the composition of A3 matrix powder used in NUKEM process, natural graphite powder, electro (artificial) graphite powder and resin binder are mixed in the ratio of 64:16:20 to produce the matrix powder. To gain data necessary to develop technology for processing the matrix graphite powder including mixing, kneading and milling, an experiment was conducted. The process consists largely of 5 steps as follows. (1) resin dissolution: the process to make the binder by dissolving resin; (2) MGP mixing : the process to mix two different graphite powders; (3) MGP kneading and drying : the process to mix and knead the mixed graphite powder with binder and to dry the mixed matrix graphite cake; (4) MGP milling : the process to grind the matrix graphite cake into fine particle powder in order to press the matrix graphite powder into proper green compacts and to utilize the finer portion of MGP for overcoating the coated particles. In order to find the graphite powder and resin suitable for preparing MGP with intended particle sizes, a total of 4 kinds of graphite powder and two types of resins were used.

2.1 Development of manufacturing method for binder.

To examine any change in the quality of MGP depending on binders, PVA (poly-vinyl alcohol) and phenolic resin were selected. PVA is a water-soluble polymeric material and its quality does not change in water solution. Accordingly, a PVA resin of 14.29 wt.% dissolved in water at 90℃ was prepared and used for MGP preparation. Also, a phenolic resin suggested by NUKEN was used for binder, for which methanol was used as a solvent to dissolve the resin. A binder of 35.2 wt% was yielded by approximately 2-hour dissolution in methanol at room temperature.

2.2 Analysis of graphite powder quality

To choose appropriate graphite powder for the purpose of the present study, Aldrich and GK powder were compared and used as natural graphite powder; and KRB and SGL powder were compared as electro graphite powder. Each raw material was analyzed in terms of attributes followed by the analysis of graphite powder after mixing 60g of natural and 16g of electro graphite powder.

2.3 Optimization of condition for each process

An attempt was conducted to find the fittest condition for the purpose in each step of MGP preparation process including the trial to find conditions for resin dissolution in the first step, powder mixing, MG paste kneading and drying and MGP milling.

3. Results

3.1 Comparison of binder quality

MGP made from PVA and phenol resins and applied with the milling step were characterized with particle size analyses.

In order to compare two resins, MGP particles applied with milling for the most optimal duration of time were compared with NUKEM's particle size distribution. As in the Figure 1 below, PVA and phenol-based MGP particles are not significantly different, but phenol-based MGP, compared to PVA-based MGP, is closer to the particle size distribution suggested by NUKEM.

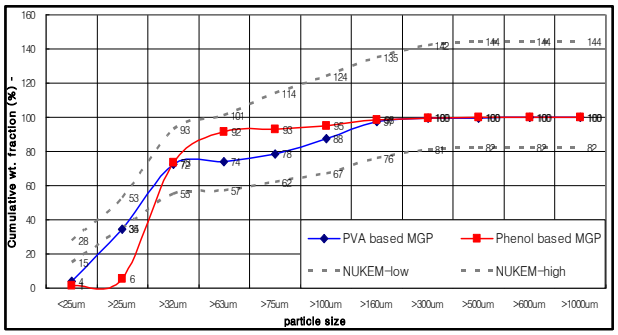

Fig. 1. Variation of particle size distribution on binder (MGP as 12 hours milling, in comparison with NUKEM spec.)

3.2 Analyzing the quality of graphite powder and mixtures

To analyze 4 different kinds of powders, that is Aldrich and GK powder for natural graphite powder and KRB and SGL powder for electro graphite powder, heat analysis, SEM measurement and particle size analysis before and after mixing particles were conducted. Aldrich and KRB powder were mixed in the ratio of 60:16 and GK and SGL powder were mixed to analyze particle sizes in the two groups to find that GK and SGL powder mixture showed a mixed state closest to the theoretical values.

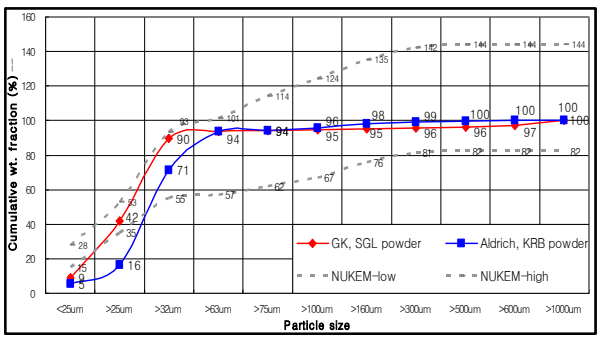

Fig. 2. Variation of particle size distribution on graphite powder (MGP as 12 hours milling, in comparison with NUKEM spec.)

Figure 2 presents the particle size distribution in MGP manufactured using the mixed powder of two groups compared with that of NUKEM. As in Figure 2, GK-SGL mixed powder demonstrates much more precise particle sizes than Aldrich-KRB mixed powder.

3.3 Optimizing process conditions for matrix graphite powder

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A basic experiment was performed to find the most optimal condition for the milling process applied with attrition mill. Particle size distribution was examined by randomly choosing ball sizes, milling time and rpm. Proceeding with the milling trial, samples were extracted at 4-hour intervals to yield particle distribution results through the Sieving test. As a result, the most optimal condition included 8-mm ball, 280 rpm and over 8-hour milling, which fell within the range of particle distribution suggested by NUKEM.

4. Conclusions

When manufacturing MGP using PVA and phenol resins to select a binder, particle size distribution between the two materials was not significantly different. Among the different binders used, phenol resin was found easier to use in the kneading process with more precise particle sizes. In the analysis of the quality to choose the raw material graphite powder, the mixing degree of GK and SGL powder mixed was found closer to the theoretical value with more precise particle size distribution compared to Aldrich and KRB mixed powder.

REFERENCES

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