

Fabrication of VHTR Nuclear Fuel Compacts Using Double-Action Compaction for Particle Damage Reduction and Anisotropy Improvement

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1. Introduction

Graphite is currently used as a structural and fuel component material in High Temperature Gas-Cooled Reactors (HTGR) due to its excellent thermal-mechanical properties and neutron moderation characteristics [1]. Graphite can effectively contain fission products for several hundred hours at temperatures up to approximately 1600 °C [2]. HTGR uses tri-structural-isotropic (TRISO) particle fuel, where UO₂ or UCO kernels, each about 500 μm in diameter, are coated with multiple layers: a PyC buffer, inner pyrolytic carbon (IPyC), silicon carbide (SiC), and outer pyrolytic carbon (OPyC). Thousands of these TRISO particles are embedded within a fuel element called a graphite compact, which serves to efficiently transfer heat to the coolant while preventing the release of fission products during reactor operation. The graphite powder mixed with TRISO particles undergoes the heat treatment processes of carbonization and purification as part of the preparation process before being loaded into the reactor as nuclear fuel. In the compact manufacturing process, uniaxial compaction often fails to fully transmit pressure to the bottom of the powder bed due to friction between the powder and the mold wall. Consequently, it is known that applying pressure from both ends during the compaction process can achieve a higher green compact density at relatively lower pressures compared to uniaxial compaction. In this study, a double-action compression mold was fabricated to perform warm compacting of graphite compacts. The density, hardness, and microstructure of these double-action compressed compacts were then compared with those of traditionally uniaxially compressed compacts.

2. Methods and Results

2.1 Graphite powder preparation

Natural and synthetic graphite powders were acquired from Graphit Kropfmuhl AG and SGL, Germany. The BET surface areas measured using gas adsorption techniques of the natural graphite powder was 4.8 m²/g and that of the synthetic graphite powder was 1.15 m²/g, respectively. These powders were classified as high-

purity nuclear grade graphite, with a purity level exceeding 99%. The particle size distribution of both types of graphite was determined by sieving with vibratory sifters for one hour, showing that around 90% of the particles, regardless of type, were within the 32-63 μm range. The phenolic resin binder used in this research is a type of resole, produced by a base-catalyzed reaction between formaldehyde and phenol, with the formaldehyde to phenol molar ratio being greater than one. The matrix graphite powder was a blend of natural and synthetic graphite mixed with the phenolic resin binder. The natural and synthetic graphite powders were combined in a 4:1 weight ratio and placed in a glass container with five ceramic balls, mixed for two hours using a Turbula® shaker mixer. Meanwhile, solid phenol, equating to a quarter of the weight of the mixed graphite powders, was dissolved in methanol (CH₃OH) using a magnetic stirrer to form the binder solution. The weight ratio of natural graphite, synthetic graphite, and phenolic resin was then set to 16:4:5, respectively. The graphite powders and the binder solution were manually mixed and kneaded using a laboratory kneader until a cohesive, sticky mass was formed. This mixture was then passed through a 2 mm metal sieve to break it into smaller fragments. These fragments were dried in an oven at 100 °C for 15 hours under a nitrogen (N₂) atmosphere and then milled in a paddled attrition metal vial with 20 stainless steel balls at 280 rpm for 8 hours, resulting in the matrix graphite powder, which predominantly consisted of particles smaller than 63 μm.

2.2 Double-action compaction, carbonization, and purification

The double-action compaction was carried out using the equipment shown in Figure. 1. Heating bands were wrapped around the mold, and the process was conducted for 3 minutes at room temperature, 100 °C, 140 °C, and 180 °C, with pressures of 10 MPa, 13 MPa, and 18 MPa. Compared to the uniaxial compression specimens previously fabricated by KAERI (Korea atomic energy research institute), which were produced at 100 °C using 98 MPa, this process was performed at significantly lower pressures [3]. After the compacts were formed, compacts underwent heat treatment in a

vacuum furnace at 800 °C for 2 hours. The heating and cooling rates were set to 0.71 °C/min. During this heat treatment, the phenolic resin within the graphite compact was carbonized, thereby improving the mechanical properties of the graphite compact. The slow ramp-up and ramp-down rates were carefully controlled to manage gas production and prevent the stress levels from exceeding a critical threshold, which could cause cracks during the carbonization process of the phenolic resin. Following carbonization, the graphite compacts were purified in a vacuum furnace at 1800 °C for 2 hours under a vacuum of approximately 5×10^{-4} Pa. The heating and cooling rates were adjusted to 5.92 °C/min and 7.4 °C/min, respectively.



Figure. 1 Double-action compaction equipment with heating band.

2.3 Characterization

The density of the graphite compacts after each process was calculated by measuring the height, radius, and mass of the cylindrical samples. Vickers hardness was measured on both the surface parallel to the compaction force (H surface) and the surface perpendicular to compaction force (V surface) using a 300 g load. The microstructure was analyzed using a scanning electron microscope. Table 1 shows the density of the compacts according to compaction pressure and temperature. Compared to the previous uniaxial compaction performed by KAERI at 100 °C and 98 MPa, which achieved a density of 1.6 g/cm³,

higher densities were achieved at significantly lower pressures in this study [3]. This is particularly encouraging because reports indicate that when the compaction pressure exceeds 15 MPa, the damage rate of TRISO nuclear fuel particles within the compact exceeds 10^{-3} [4]. Therefore, achieving higher density at lower pressures is a significant improvement.

Table 1. The density of green compacts according to compaction pressure and temperature.

Pressure [MPa]	RT	100 °C	140 °C	180 °C
10	1.20 g/cm ³	1.55 g/cm ³	1.71 g/cm ³	1.71 g/cm ³
13	1.32 g/cm ³	1.67 g/cm ³	1.77 g/cm ³	1.75 g/cm ³
18	1.34 g/cm ³	1.75 g/cm ³	1.78 g/cm ³	1.76 g/cm ³

Figure 2 shows the Vickers hardness of the uniaxial and double-action compaction specimens after each process. Compared to double-action compaction, uniaxial compaction exhibits a significant difference in hardness between the V surface and H surface after purification. This is likely due to the excessively high pressure applied during uniaxial compaction, which may have resulted in pronounced anisotropy between the axial and radial directions.

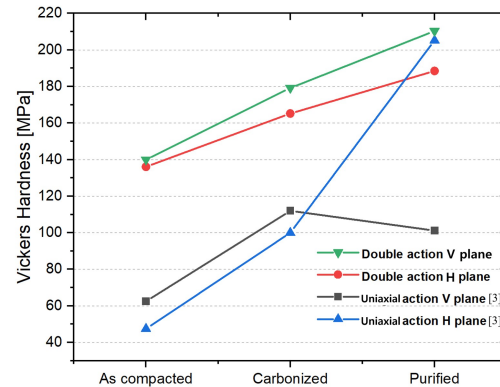


Figure. 2 Vickers hardness of the uniaxial and double-action compaction specimens after each process [3].

Figure 3 shows the SEM (Scanning electron microscope) microstructure of the H surface of the uniaxial and double-action compaction specimens after purification. In the H surface of the uniaxial compact, the phenolic resin-derived coke can be observed lying along the radial direction, whereas in the H surface of the double-action compact, the phenolic resin-derived coke appears relatively isotropic. The observed microstructural anisotropy suggests that the lower pressure used during the compaction process

significantly contributed to improving the anisotropy of the compact.

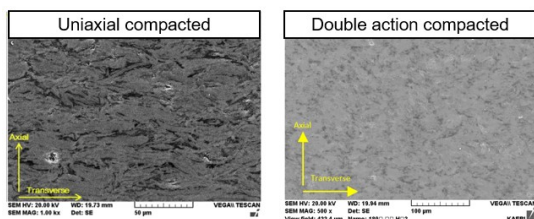


Figure. 3 SEM microstructure of the H surface of the uniaxial [3] and double-action compaction specimens after purification.

Graphite compacts were fabricated using double-action compaction. The graphite compacts produced at lower pressures using double-action compaction exhibited higher green compact density compared to those formed by conventional uniaxial compaction. Additionally, the anisotropy between the axial and radial directions was significantly improved. This study is expected to greatly contribute to the future commercialization of HTGRs and the in-house production of nuclear fuel.

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