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Original Article

Carbon Contained Ammonium Diuranate Gel Particles Preparation in Mid-process of High-temperature Gas-cooled Reactor Fuel Fabrication

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A B S T R A C T

This study investigates the dispersibility of carbon in carbon contained ammonium diuranate (C-ADU) gel particles and the characteristics of C-ADU gel liquid droplets produced by the vibrating nozzle and integrated aging–washing–drying equipment. It was noted that the excellent stability of carbon dispersion was only observed in the C-ADU gel particle that contained carbon black named CB 10. ADU gel liquid droplets containing carbon particles with the excellent sphericity of approximately 1,950 μm were then obtained using an 80–100-Hz vibrating nozzle system. Dried C-ADU gel particles obtained by the aging–washing–drying equipment were thermal decomposed until 500°C at a rate of 1°C/min in an air or in 4% H_2 gas atmosphere. The thermally decomposed C-ADU gel particles showed 24% weight loss and a more complicated profile than that of ADU gel particles.

1. Introduction

Nuclear fuel used in a high temperature gas-cooled reactor is tristructural isotropic (TRISO)-coated particle fuel. Spherical fissile material (UO_2 or UCO) is coated by trilayers such as inner-pyrolytic carbon, SiC, and outer-pyrolytic carbon [1,2] to prevent the release of fission products during the operation of the reactor. Depending on the type of reactor, hundreds of TRISO particles are contained in a pebble or prismatic shaped graphite element loading in the core of a reactor.

The initial form of the spherical fissile material is called ammonium diuranate (ADU) gel particle fabricated using the gel-supported precipitation method modified from the sol–gel processes [3,4]. This method involved the following sequence. First, organic alcohol is added to a uranyl nitrate (UN) solution to produce a broth solution. Second, the broth solution passes

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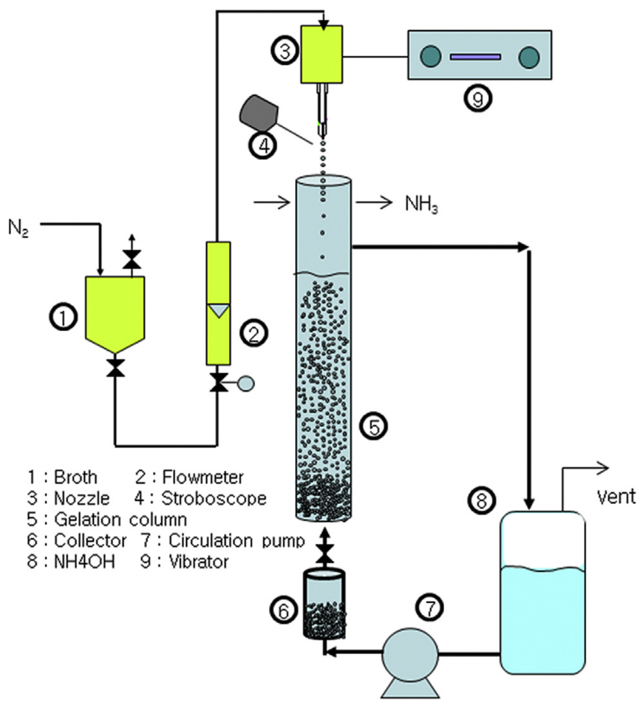

Fig. 1 – Material flow diagram for C-ADU gel particles fabrication.

through the vibrating nozzle system forming spherical ADU gel liquid droplets. Third, uranyl ions existing in the droplets chemically react with ammonium ions in the gelation solution at room temperature. Last, intermediate ADU gel particles after the reaction are formed into spherical-shaped particles and go through aging, washing, drying, heat treatment, reduction, and sintering processes. The particles are then turned into high-density UO_2 microsphere particles.

It is known that CO or CO_2 gas is created by the chemical reaction between excessive oxygen in spherical UO_2 (actually, UO_{2+x}) particles and pyrolytic carbon during the coating process. The formed gaseous phase increases the core pressure of TRISO particles leading to an unstable factor of nuclear fuel [5,6]. The production of the gaseous phase during the coating process can be suppressed by the addition of carbon particles previously in the raw material solution when the spherical UO_2 particle is produced [7]. Excessive oxygen is eliminated in the carbon contained UO_2 called UCO (uranium oxycarbide; $UO_2 + UC_2$) particle.

The external gelation process is developed to replace the internal gelation process for the spherical carbon contained ADU (C-ADU) gel particles. The fabrication method for the spherical UCO particles in the United States is the internal gelation process [8]. Whereas the internal gelation process uses carbon dispersed acid-deficient uranyl nitrate solution, the UN solution contained carbon particles with tetrahydrofurfuryl alcohol (THFA), and polvinyl alcohol (PVA) as broth solution preparation and ammonia solution as gelation medium are utilized in the external gelation process [9]. To obtain this solution, it is important to select the appropriate carbon substance with excellent dispersibility in advance. The current study points to the capability of the external gelation process to yield C-ADU gel particles [10,11] and the selection of a suitable carbon substance to disperse carbon in the UN solution.

2. Materials and methods

2.1. *Selection of carbon substance by simulating solution*

Several simulating solution tests were conducted to select the appropriate carbon substance in advance of producing spherical C-ADU gel particles. A raw material solution with a concentration of 1.97 mol Ce/L was created by dissolving $Ce(NO_3)_3 \cdot 6H_2O$ (99.99%; Kanto Chemical, Tokyo, Japan) powder in distilled water at room temperature to replace the uranium solution utilized in the general ADU gel particle fabrication process. Appropriate amounts of THFA and PVA solutions are then added in the simulating solution. The viscosity of the solution is controlled by adding distilled water. A total of 10 different carbon substances—six substances from Cabot Co. (Cabot Co., Louisiana, USA) and four substances from Colombian Chemicals Co. (Colombian Chemicals Co. Adityabirla, India)—were examined in the current study through the following sequence. First, the carbon substance is added to the simulating solution. Second, carbon particles

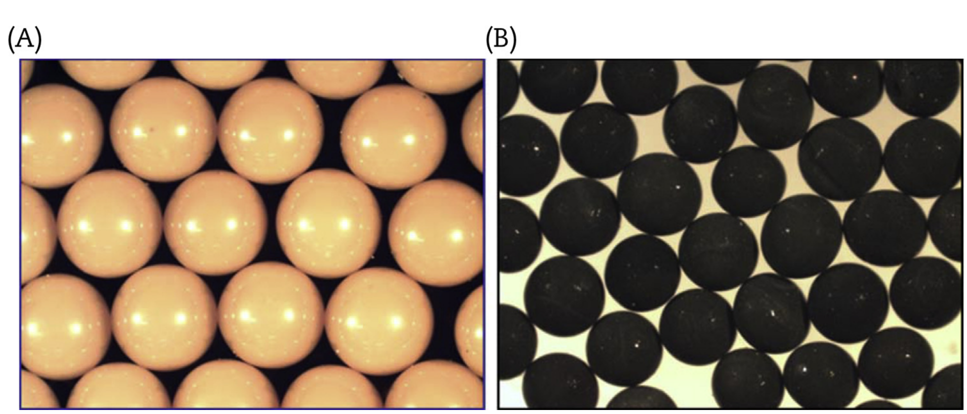

Fig. 2 – ADU gel particles. (A) Without carbon; (B) with carbon.