# Spherical Fuel Element Deconsolidation System in INET

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## Abstract

For High-Temperature Gas-cooled Reactor in China, coated fuel particles are bonded into spherical fuel elements by graphite matrix. In conjunction with the development of a system to determine fuel failure fractions through γ-analysis of individual and undamaged fuel particles, an automated spherical fuel element deconsolidation (SFED) system was developed in this study to separate the particles from graphite matrix by disintegrating the matrix into fine graphite powder, based on the anodic oxidation of graphite in an electrolyte containing nitric acid.

The apparatus in this work contains electrolyzing cell, rotator, anode, Pt cathode and $HNO_3$ electrolyte, DC electrolytic power supply and control system based on the anodic oxidation. During the deconsolidation, the spherical fuel element is treated by DC power in electrolyte, and the graphite matrix is intercalated, oxidized and integrated to powder. Afterwards, the mixture of electrolyte and graphite powder is treated by vacuum filtration, and coated fuel particles are sampled from outside to inside in sequence. This system is compact, easy to operate and remote monitoring. Also, the position information can be maintained during the deconsolidation treatment, not only in favor of coated fuel particle breakage research and analysis of fission products in PIE, but also avoiding cross contamination among different samples.

## 1. Introduction

High-temperature gas-cooled reactor (HTR) is considered as one of most promising candidates for the next generation of nuclear energy because of a series of features like large thermal inertia, high heat capacity, structural stability, and the advanced inherent safety [1]. The fuel elements of HTR in China are spherical and have an overall diameter of 60 mm. A 5 mm thick unfueled matrix graphite shell comprises the outer portion of the sphere. The central, fueled region of the fuel element is about 50 mm in diameter and is composed of 0.92 mm diameter tri-structural isotropic (TRISO) coated fuel particles that are overcoated and pressed into a 50 mm diameter sphere [2]. The graphite matrix covers more than 95% weight of the fuel element. To study the irradiation behaviors of the spent fuel element, coated fuel particles need to be separated and sampled from graphite matrix from the inside out for further inspections of fission products distribution. Therefore, the structural integrity of coated particles and the complete separation of coated particles from graphite matrix are two key issues for exact measurements. Intact coated particles consisting of intact silicon carbide (SiC) and outer/inner pyrolytic carbon (PyC) layers should be maintained, with graphite matrix and overcoating layers removed.

Deconsolidation is an important sample preparation process that aims to obtain loose coated

fuel particles after irradiation or heat treatment in the Küfa installation for further study of failure mechanisms at the coated particle level [3]. In scaled-up form, deconsolidation techniques may become important in reprocessing of spent fuel and diminishing the amount of graphite requiring disposal. In deconsolidation process, HTR fuel elements are transferred to hot cell installations to perform detailed studies in particular on coated particles, and post-irradiation examination (PIE) work on coated particles starts with deconsolidation of fuel elements in order to separate embedded coated particles from graphite matrix. The principal aim of the deconsolidation process is to identify broken or damaged particles by means of the irradiated microsphere gamma analyser (IMGA) procedure. Some requirements for deconsolidation apparatus are mandatory: avoiding destruction or damage of the particle coating; easy adaptation of the process to remote handing conditions; simple and reliable mode of operation and a reasonable total process time [4].

The new hot cell laboratory in Institute of Nuclear and New Energy Technology (INET) is the first facility in China designed for HTR spherical spent fuel element post irradiation examination test. It is designed in 2011, and constructed in this year. Considering the exactly different shape of spherical fuel elements from PWR and AGR fuel elements, facilities in INET hot cell are specially designed and modified for spherical elements and TRISO coated fuel particles. In this work, an automated spherical fuel element deconsolidation (SFED) system prototype was developed to separate the particles from the graphite matrix by disintegrating the matrix into fine graphite powder, based on the anodic oxidation of graphite in nitric acid electrolyte. Meanwhile, the position information of coated particles in spherical fuel element can be maintained, which aids to better understand coated particle breakage mechanism. Besides, this apparatus can also be easily adapted by remote handling, and the total process can be controlled by electric current intensity without operation-induced damage of the coated particles.

## 2. Experimental and Facilities

The apparatus in this work contains both electric motor system and electro-deconsolidation system. The electric motor system rotates the spherical fuel element and achieves the position transformation; the electro-deconsolidation system disintegrates the graphite matrix to expose the coated fuel particles based on the anodic oxidation of graphite matrix. The apparatus diagram of SFED is shown in Figure 1. The sample sphere is connected to a direct current source as well as a gear motor using a motor rod linker. The motor rod linker is only inserted about 4 mm into the unfueled portion of the sphere to avoid particles' damage as a result of the deconsolidation process. The deconsolidation process involved two steps. In the first step, the sample sphere rotates (1 rpm) with the outer circumference emerged in the electrolyzing solution, leaving a 2 cm-cylinder in diameter at the end. In the second step, the remaining cylinder is rotated by $90^{o}$ and lowered into the electrolyte in discrete 10 steps (Figure 2).

To study the performance of the equipment, un-irradiated spherical graphite balls with $ZrO_2$ kernels produced by the similar processing of $UO_2$ fuel elements were used as the test samples in this work. The tested electric current in this work were 2A (2.4 V), 7 A (6.4 V), 10 A (8.5 V) and 13 A (10.7 V). Each step yields a sample of electrolyte solution, graphite matrix, and associated coated particles after a series of separation method. Then un-bonded particles were collected and packaged separately, providing a profile of fuel from the edge to the center of the sphere.
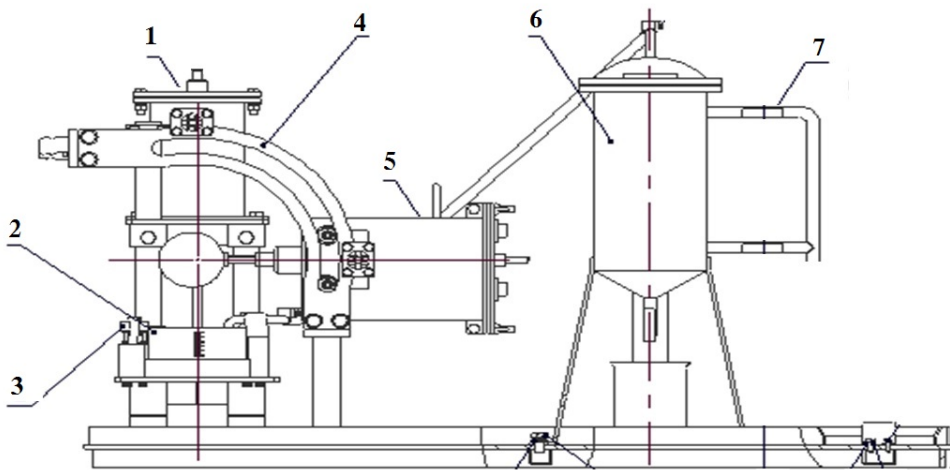
Figure 1. The apparatus diagram of SFED. 1. Lifting motor; 2. Electrolytic cell; 3. Pt cathode; 4. Guide rail of rotating motor; 5. Rotating motor; 6. Separating tank; 7. Vacuum pump tube.
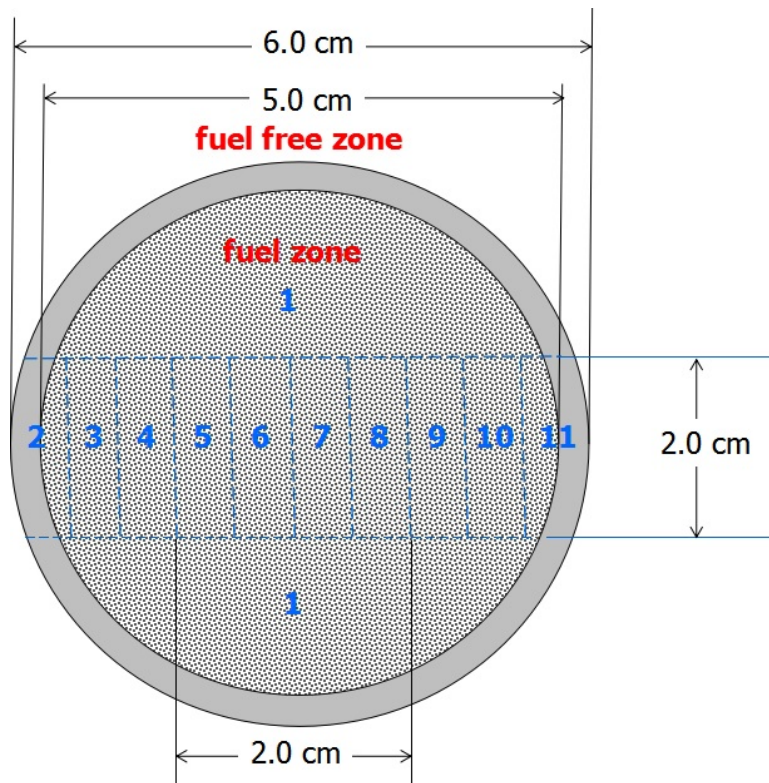


Figure 2 Samples obtained by two-step electro-deconsolidation of spherical fuel element

## 3. Results and discussion

3.1 Study of deconsolidation process

The raw graphite matrix powder is composed of natural graphite, synthetic graphite and resin binder in a certain proportion, and experiences a series of high temperature treatment to form graphite matrix [5]. The binder phenol resin is used to provide some adhesion to the mixture and helps to adhere the graphite matrix powder to the TRISO-coated fuel particles during the

overcoating process, and will be hardened into a glassy carbon during the subsequent carbonization process [6]. Compared to natural and synthetic graphite, the structure of glassy graphite is not so ordered, so it is probably easier to be electro-chemically oxidized. Also, micropore structure will be formed on the surface or in the bulk after manufacture processing due to the compression of the powder and volatilization of light components in binder. During the electrolysis, the nitric acid electrolyte gradually infiltrated into the micropores of graphite matrix, and the electrochemical reaction would take place around micropore structure as well as on matrix surface.

After deconsolidation, the fuel elements were disintegrated to mixture of graphite matrix powders and coated particles. Actually, the graphite matrix powders usually aggregate into larger bulks than coated particles (Figure 3, left), which makes it difficult to separate clean coated particle samples for further examination. In this work, ultrasonication with high energy is used to break the large graphite matrix bulks into small pieces, which facilitates the fractional filtration with only coated particle samples obtained (Figure 3, right).
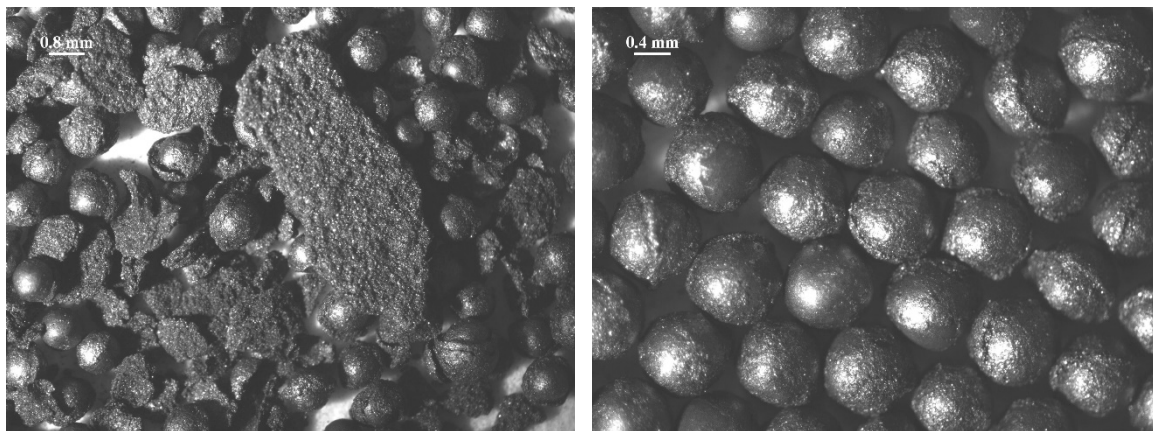


Figure 3. Left: the graphite matrix and coated particle samples after deconsolidation at 7 A; Right: the coated particles after fractional filtration from graphite matrix.

3.2 Electric current effect on particle size of deconsolidated graphite matrix

If look further into the current intensity effect on the particle size of graphite matrix after deconsolidation, it could be found that the particle size decreased as the current increased, and increased again if the electric current increased to 13 A (Figure 4). These results indicate that, when the electric current are relatively small, larger deconsolidated particles are more likely to be formed caused by more graphite oxidization inside the bulk. In this case, the permeation rate is faster than electrolyzation rate, and the latter is a rate control step. Inversely, when the electric current becomes large enough, fine particles are more likely to be formed caused by more graphite oxidation on the interface. In this case, the permeation rate is slower than electrolyzation rate, and the former is a control step. Therefore, the rate control step turns from electrolyzation rate to permeation rate at the electric current of around 10 A. According to this figure, the electric current from 7 A to 10 A is recommended, because the large difference in particle sizes of graphite fragments and TRISO-particles facilitates further separation of deconsolidated graphite matrix and coated particles.
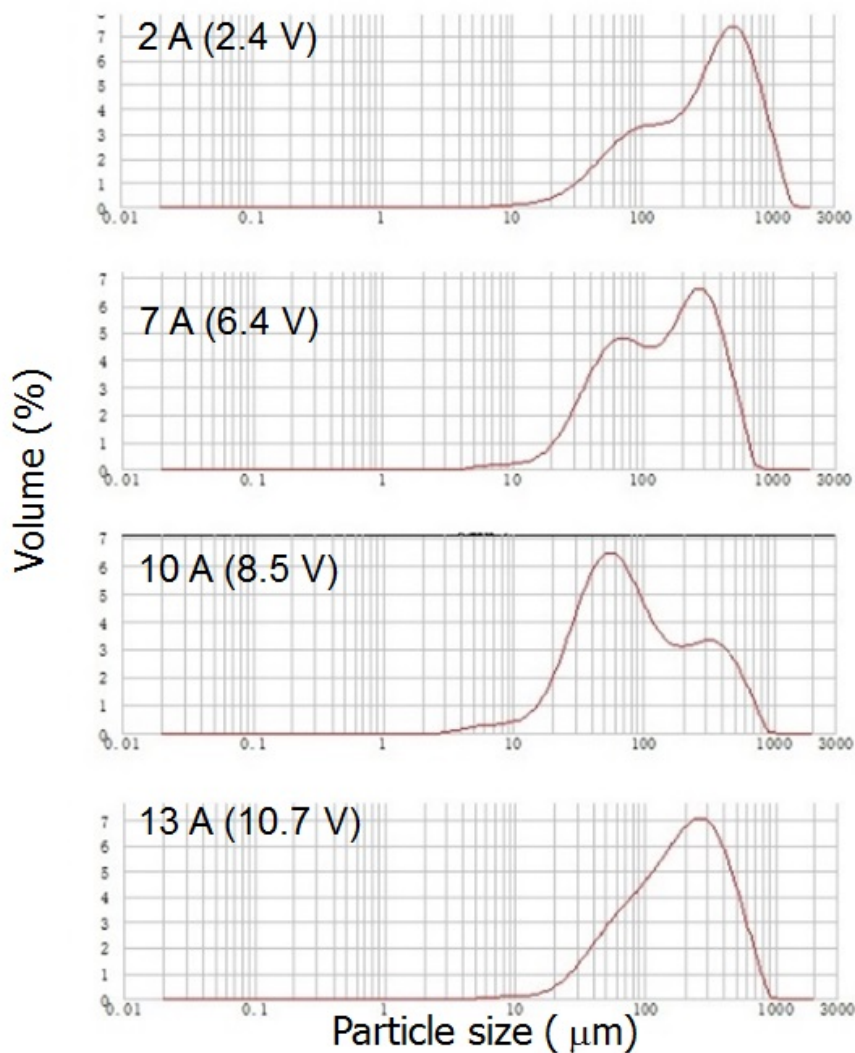
Figure 4. The electric current effect on particle size of deconsolidated graphite matrix. From top to bottom: 2 A (2.4 V), 7A (6.4 V), 10 A (8.5 V) and 13 A (10.7 V).

3.4 The components of graphite matrix before and after deconsolidation

In our previous work, elemental analysis, XPS and XRD were used to examine the components changing of graphite matrix after deconsolidation [7]. It is evidenced that more abundant graphite oxide compounds were formed on the interface after deconsolidation. In this work, Raman spectra were used to characterize the graphitization changing regulation when the fuel element samples were deconsolidated at different electric currents. The results were shown in Figure 5. The peak at 1580 cm$^{-1}$ ascribes to the graphite lattice structure, which is called G-band; and the one at 1360 cm$^{-1}$ ascribes to the graphite lattice structure with disturbance, which is called D-band. The ratio of 1360 cm$^{-1}$ to 1580 cm$^{-1}$ could be used to characterize the graphitization, with larger ratio value indicating lower graphitization [8]. According to the results from Table 1, it could be concluded that the graphitization of deconsolidated graphite matrix becomes lower as the electric current intensified.
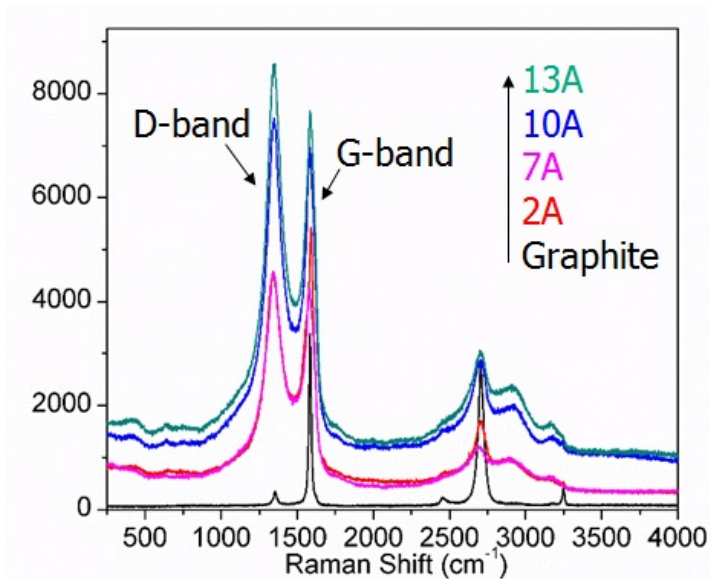
Figure 5 Raman spectra of deconsolidated graphite matrix at different electric currents.

Table 1 The Raman intensity at 1360 cm$^{-1}$ and 1580 cm$^{-1}$ with the ratio of these two values at different electric currents

|  | $I_{1360}$ | $I_{1580}$ | $I_{1360/I1580}$ |
|---|---|---|---|
| Graphite matrix | 7665 | 63978 | 0.12 |
| 2A (2.4 V) | 857606 | 635687 | 1.35 |
| 7A (6.4 V) | 806913 | 525540 | 1.54 |
| 10A (8.5 V) | 1497070 | 593611 | 2.52 |
| 13A (10.7 V) | 969744 | 322673 | 3.01 |

3.5 The characterization of coated particles after deconsolidation

During the deconsolidation, it is critical to maintain intact structure of obtained TRISO-particles for further analysis. According to EDS characterizations (Figure 6) of coated-particle surface after deconsolidation, Si was detected besides C and O if the samples were treated by large electric current like 10 A and 13 A, indicating that the outer PyC laryer was broken and SiC layer was exposed. Therefore, larger electric current might increases the breakage risk of PyC layer. Combining the above results for graphite fragments and TRISO-particles, the current of 7 A (6.2 V) is preferred in this study.
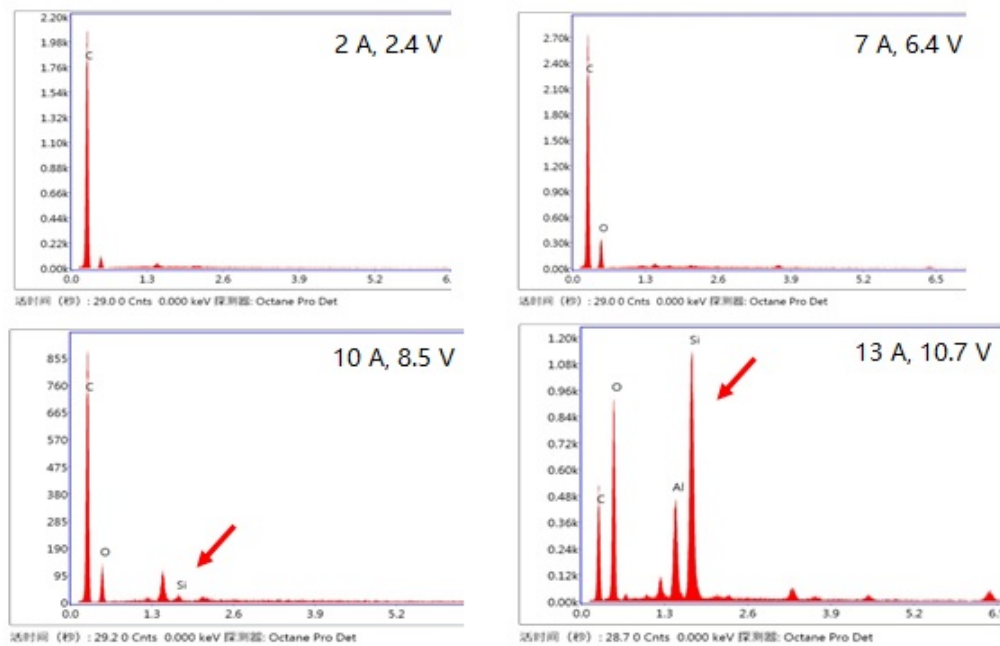
Figure 6. EDS characterizations (Figure 6) of coated-particle surface after deconsolidation at different electric currents

## 4. Conclusions

In this work, an automated spherical fuel element deconsolidation (SFED) system prototype was developed to separate the particles from the graphite matrix by disintegrating the matrix into fine graphite powder, and the effect of different electric current intensities on the deconsolidation of spherical fuel element was studied. The position of coated particles in spherical fuel element can be recorded, which aids to provide more information to coated particle breakage mechanism. According to the structure and surface analysis results, the particle size and component of deconsolidated graphite matrix are related to the current intensity, and the total process can be controlled by electric current intensity without operation-induced damage of the coated particles.

## Acknowledgment

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