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# Characterization of graphite dust produced by pneumatic lift

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## HIGHLIGHTS

- Generation of graphite dust by pneumatic lift.
- Determination of morphology and particle size distribution of graphite dust.
- The size of graphite dust in this study is compared to AVR and THTR-300 results.
- Graphite dust originates from both filler and binder of the matrix graphite.

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## ABSTRACT

Graphite dust is an important safety concern of high-temperature gas-cooled reactor (HTR). The graphite dust could adsorb fission products, and the radioactive dust is transported by the coolant gas and deposited on the surface of the primary loop. The simulation of coagulation, aggregation, deposition, and resuspension behavior of graphite dust requires parameters such as particle size distribution and particle shape, but currently very limited data on graphite dust is available. The only data we have are from AVR and THTR-300, however, the AVR result is likely to be prejudiced by the oil ingress. In pebble-bed HTR, graphite dust is generally produced by mechanical abrasion, in particular, by the abrasion of graphite pebbles in the lifting pipe of the fuel handling system. Here we demonstrate the generation and characterization of graphite dust that were produced by pneumatic lift. This graphite dust could substitute the real dust in HTR for characterization. The dust, exhibiting a lamellar morphology, showed a number-weighted average particle size of 2.38 μm and a volume-weighted average size of 14.62 μm. These two sizes were larger than the AVR and THTR results. The discrepancy is possibly due to the irradiation effect and prejudice caused by the oil ingress accident. It is also confirmed by the Raman spectrum that both the filler particle and binder contribute to the dust generation.



## 1. Introduction

The pebble-bed high-temperature gas-cooled reactor (pebble-bed HTR), a Generation IV reactor concept, is featured by the inherent safety. The meltdown of the core is physically impossible, due to the proper design of the core structure and the utilization of all-ceramic fuel elements. Since the reactor has a large negative temperature coefficient, and decay heat can be safely removed without the need for helium coolant circulation, the maximum core temperature is limited at a tolerable level in a loss of forced cooling accident scenario.

The pebble-bed HTR employs spherical fuel element, which is composed of matrix graphite and uniformly dispersed tristructural-isotropic (TRISO)-coated fuel particles. During normal operation, graphite dust is generated due to the contact between spherical fuel elements and other components. This dust acts like radioactivity carrier by adsorption of the fission products, and is transported by coolant, and deposited on the surface of the primary loop under steady-state operation conditions (Gottaut and Kruger, 1990). In the event of a breach of the primary system, a certain amount of the contaminated dust can be resuspended and released. Several modeling works have been performed to analyze the deposition and resuspension of graphite dust in HTR-10 (Peng et al., 2016, 2013a,b, 2014). These computational predictions rely on the accurate description of the dust properties such as particle

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size distribution and particle shape. So far, no more information on the graphite dust of HTR is available except AVR and THTR data and, moreover, the AVR data was regarded to be prejudiced, since the oil ingress accident probably changed the dust nature (Kissane et al., 2012). On the other hand, the examination of real graphite dust is of difficulty because of its radioactivity.

There are some researchers who investigated the characteristics of graphite dust particles by wear experiment (Troy et al., 2012, 2015; Luo et al., 2004). However, in a real pebble-bed HTR, graphite dust results from the abrasion during fuel element recirculation (IAEA, 1997), particularly in the lifting line of the fuel handling system (Verfondern et al., 2012). Accordingly, an experimental platform was designed to simulate the graphite dust production in HTR. In our previous paper (Shen et al., 2015), we investigated the abrasion of graphite pebble in the lifting pipe and successfully showed the mechanism of graphite dust production. Meanwhile, graphite dust sample was collected from the inner surface of the pipe. The process of generating the dust sample was similar to the formation of the real graphite dust in the HTR, making it a good substitute for real graphite dust.

## 2. Experiment

### 2.1. Graphite pebble

The graphite pebbles used in this study were fabricated by the Institute of Nuclear and New Energy Technology of Tsinghua University with the same process that was used for spherical fuel element in HTR-10, except that the graphite pebbles did not contain TRISO-coated fuel particles. The graphite matrix material was manufactured using a powder mixture composed of 64 wt.% natural flake graphite, 16 wt.% artificial graphite, and 20 wt.% phenol resin binder (Tang et al., 2008; Zhao et al., 2006). The highest heat treatment temperature did not exceed 2000 °C in order to avoid damage to the SiC layer in case TRISO particles are present.

For both HTR-10 and HTR-PM, the weight of fuel element range from 200 g to 220 g, depending on the source of raw materials. The weight of a pure graphite pebble typically differs from the fuel element by less than 10 g. The graphite pebble used in this study weighted approximately 200 g, and was considered to be an excellent substitute for the real fuel element.

### 2.2. Graphite dust generation

Graphite dust was generated by a lifting test platform, which was described in detail in our previous paper (Shen et al., 2015). The platform consisted of a lifting pipe, three gas cylinders, a gas control system and a timing system. The cold-drawn stainless steel lifting pipe was approximately 9 m in height, containing an 8 m long straight part. The upper end of the pipe was shaped into a semicircle with a radius of 0.3 m. The pipe had an inner diameter of 62 mm, and allows the graphite pebble with a diameter of 60 mm to pass through freely. Three gas cylinders were connected to the lower end of the pipe. The acceleration of the graphite pebble was controlled by the pressure regulators, and the resultant lifting velocities were in the range of 3–10 m/s. An automatic timing system was designed for measuring the lifting time of the graphite pebble accurately. The lift was performed at room temperature. According to Rowe (Rowe, 1960), the friction properties of graphite, such as the coefficient of friction, changes very slightly from room temperature to 800 °C. Since the outlet temperature of HTR-10 and HTR-PM do not exceed 750 °C, the temperature effect was not taken into account in the present study.

In our previous paper, the relationship between abrasion rate and lifting velocity was investigated by this platform in different atmospheres. The abrasion rate was evaluated by the weight loss of the graphite pebble after pneumatic lift. Because the weight loss for a single lift is too small to measure, the lift was repeated for 10 times at certain velocities so that the accumulative weight loss became measurable.

The abrasion behavior was investigated in air, nitrogen and helium at room temperature respectively. In a certain atmosphere, the abrasion rate was recorded at about 20–25 different mean lifting velocities, giving rise to total pneumatic lifts up to 600 times, and generated a number of graphite dust on the inner surface of pipeline. This dust was collected by cotton ball soaked with ethanol. Subsequently, the cotton was repeatedly washed with ethanol, and then the suspension was collected, centrifuged, and dried to obtain the graphite dust sample.

### 2.3. Characterization

The original matrix graphite and its raw materials were characterized by an optical microscope, SEM, and Raman spectrum.

The morphology of graphite dust was observed by scanning electron microscope (SEM). Raman spectrum of individual dust particle was collected with Renishaw RM2000 Raman microscope. The particle size distribution was measured using both image analysis and laser diffraction particle size analyzer. For the image analysis, the dust was dispersed in ethanol and deagglomerated in an ultrasonic bath for 5 min. One drop of the suspension was placed on a clean silicon wafer and dried. SEM images (500X) was taken at random locations, resulting in about 300 graphite dust particles, and then the particle size was measured from the SEM images manually. For the laser diffraction approach, the particle size distribution was measured by Mastersizer 2000 (Malvern).

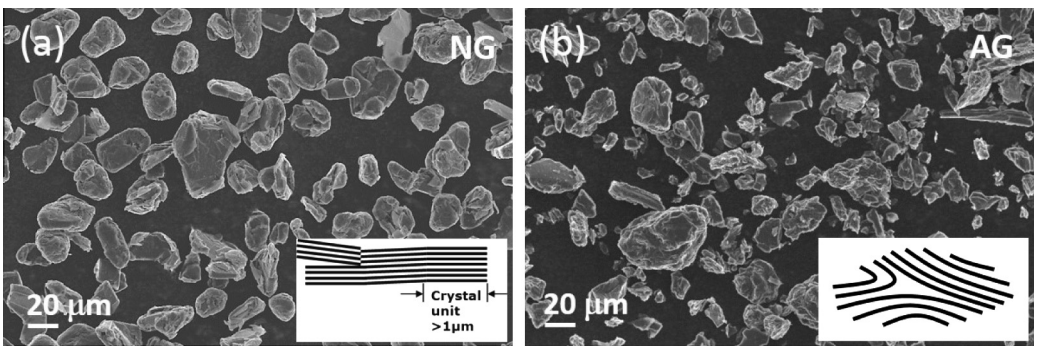


**Fig. 1.** Filler particle of matrix graphite. (a) Natural flake graphite. (b) Artificial graphite. The insets show schematics of their microstructure.