Particuology xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Particuology

journal homepage: www.elsevier.com/locate/partic

A novel coated-particle design and fluidized-bed chemical vapor deposition preparation method for fuel-element identification in a nuclear reactor

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ARTICLE INFO

Article history:
Received 1 May 2015
Received in revised form 23 June 2015
Accepted 17 May 2016
Available online xxx

Keywords:
Particle coating
Particle design
Cobalt
Fluidized-bed chemical vapor deposition
Nuclear fuel

ABSTRACT

Particle coating is an important method that can be used to expand particle-technology applications. Coated-particle design and preparation for nuclear fuel-element trajectory tracing were focused on in this paper. Particles that contain elemental cobalt were selected because of the characteristic gamma ray spectra of ^{60}Co. A novel particle-structure design was proposed by coating particles that contain elemental cobalt with a high-density silicon-carbide (SiC) layer. During the coating process with the high-density SiC layer, cobalt metal was formed and diffused towards the coating, so an inner SiC–Co_xSi layer was designed and obtained by fluidized-bed chemical vapor deposition coupled with in-situ chemical reaction. The coating layers were studied by X-ray diffractometry, scanning electron microscopy, and energy dispersive X-ray spectroscopy techniques. The chemical composition was also determined by inductively coupled plasma optical emission spectrometry. The novel particle design can reduce the formation of metallic cobalt and prevent cobalt diffusion in the coating process, which can maintain safety in a nuclear reactor for an extended period. The experimental results also validated that coated particles maintain their structural integrity at extremely high temperatures (∼1950 °C), which meets the requirements of next-generation nuclear reactors.

Introduction

Particle-coating layers provide one or more of the following advantageous attributes: particle surface modification to adapt to a new environment (Lin, Wang, & Jin, 2002), rate control of active-ingredient release (Y. Liu, Wang, Qin, & Jin, 2008; Turton, 2008), particle protection from their surroundings (air, moisture, and light) (Werner, Jones, Paterson, Archer, & Pearce, 2007), or they can provide a means for identification (Cole et al., 2014). Particle-coating design is an important way to obtain different physical and/or chemical properties in industrial practical use. Particle-coating technology is used extensively in the nuclear research field (Garnaud, Han, Jacquet, Ndombo, & Limaiem, 2015; Hidayatullah, Hartanto, & Kim, 2015), especially in high-temperature gas-cooled nuclear reactors (Liu, Liu, Li, Shao, & Liu, 2014). A specially designed particle, which is termed the tristructural-isotropic (TRISO)-type coated particle, is the first security assurance to block the release of fission products from a nuclear fuel particle to the primary coolant circuit (Nickel, Nabielek, Pott, & Mehner, 2002). A TRISO-type coated fuel particle consists of a microspherical nuclear fuel kernel that is surrounded by four coated layers, namely, a porous buffer pyrolysis carbon layer (buffer PyC), an inner dense pyrolysis carbon layer (IPyC), a high-density silicon-carbide (SiC) layer, and an outer dense pyrolysis carbon layer (OPyC). These coating layers with different functions have been prepared by fluidized-bed chemical vapor deposition (FB-CVD) (Liu, Liu, Liu, & Shao, 2015).

In the development of next-generation nuclear power plants, inspection methods are required to detect and distinguish different fuel elements in the reactor core because of inhomogeneities in burnup. Rod-like fuel elements are installed securely in traditional nuclear pressurized-water reactor cores and fuel-element identification can be achieved using simple markers. However, in other new reactor types such as the pebble-bed (Liu, Du, Han, Zou, & Pan, 2015) or fluidized-bed (Rots, Mudde, Van Den Akker, Van Der Hagen, & Van Dam, 1996) nuclear reactors, spherical fuel elements are mixed to some extent, and these cannot be distinguished

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http://dx.doi.org/10.1016/j.partic.2016.05.009

easily by direct discrimination using simple markers. Therefore, an urgent demand exists to develop new trajectory tracers to distinguish different fuel elements in nuclear reactors. Compared with conventional inspection methods, particles that contain elemental cobalt are a good choice for particle trajectory tracers in nuclear power plants because of the high-level radioactive environment.

In a nuclear reactor, ^{59}Co in the particles will be transformed to ^{60}Co by neutron bombardment. The characteristic ^{60}Co gamma-ray spectrum is used to detect the fuel element. Because gamma-ray spectrometers are usually used in the transfer of spent fuel elements (Yan, Zhang, Zhang, Zhang, & Xiao, 2014), this method can be used directly and does not need any modification for use in the nuclear-reactor core. However, cobalt can diffuse into the reactor core and even be volatilized at high temperature if it does not have an appropriate coating layer, thus coating layers should be designed to block the release of cobalt at high temperatures.

Because particle trajectory tracers will be integrated into fuel elements, the main design principle of coating particles that contain elemental cobalt is that the coating-layer structure should be kept nearly the same as the TRISO-coated fuel particles that have been used in the nuclear reactor. The SiC layer is the most important layer in these TRISO-coated particles because this high-density ceramic layer is the primary layer that blocks most radioactive fission products at elevated temperatures (Petti, Buongiorno, Maki, Hobbins, & Miller, 2003; Snead et al., 2007). This work is focused on producing particles that contain elemental cobalt with a dense SiC-coated layer. Conventional FB-CVD was evaluated and TRISO-type coated particles that contain elemental cobalt were prepared. However, this method could not meet requirements and required modification. Based on detailed analysis, a novel coated-particle design and a FB-CVD method that is coupled with an in-situ chemical reaction were proposed. This newly designed coated particle was prepared, characterized and validated experimentally and by using different methods of analysis.

Experimental

Particle preparation by FB-CVD method

A conventional FB-CVD method conducted in a spouted-bed coating system was used to prepare the TRISO particles, as shown in Fig. 1. Kernels were fluidized in the bottom of the furnace by fluidizing gas at a high temperature. Reactive gases were injected into the coating furnace, pyrolyzed, and then coated on the kernels to obtain uniformly coated layers when appropriate parameters, such as gas velocity and temperature, were set.

All chemicals were of analytical grade and were used as received without any further purification. ZrO_2–CoO (CoO 12 wt%) kernel particles were prepared by sintering powder mixture of ZrO_2 and CoO at 1500 °C with some additive agent. Initially, coated layers were designed in the same way as the TRISO particle. A PyC buffer layer was deposited from acetylene at 1200 °C. The inner and outer high-density PyC layers were derived from a mixture of acetylene and propylene at 1400 °C. The thicknesses of the buffer, inner PyC, and outer PyC layers were controlled to be 95, 40, and 40 μm, respectively. A dense 40-μm-thick SiC layer was deposited by methyltrichlorosilane decomposition at 1600 °C for 2 h using a mixture of hydrogen and argon as fluidizing gas (H_2:Ar = 4:1, vol), and hydrogen as carrier gas. In the novel particle design, a low-temperature SiC layer was prepared at 1400 °C for 1.5 h and the other experimental parameters were the same as those of the dense SiC layer. After coating, the obtained particles were annealed at 1950 °C for 1 h in vacuum to test their structural stability.

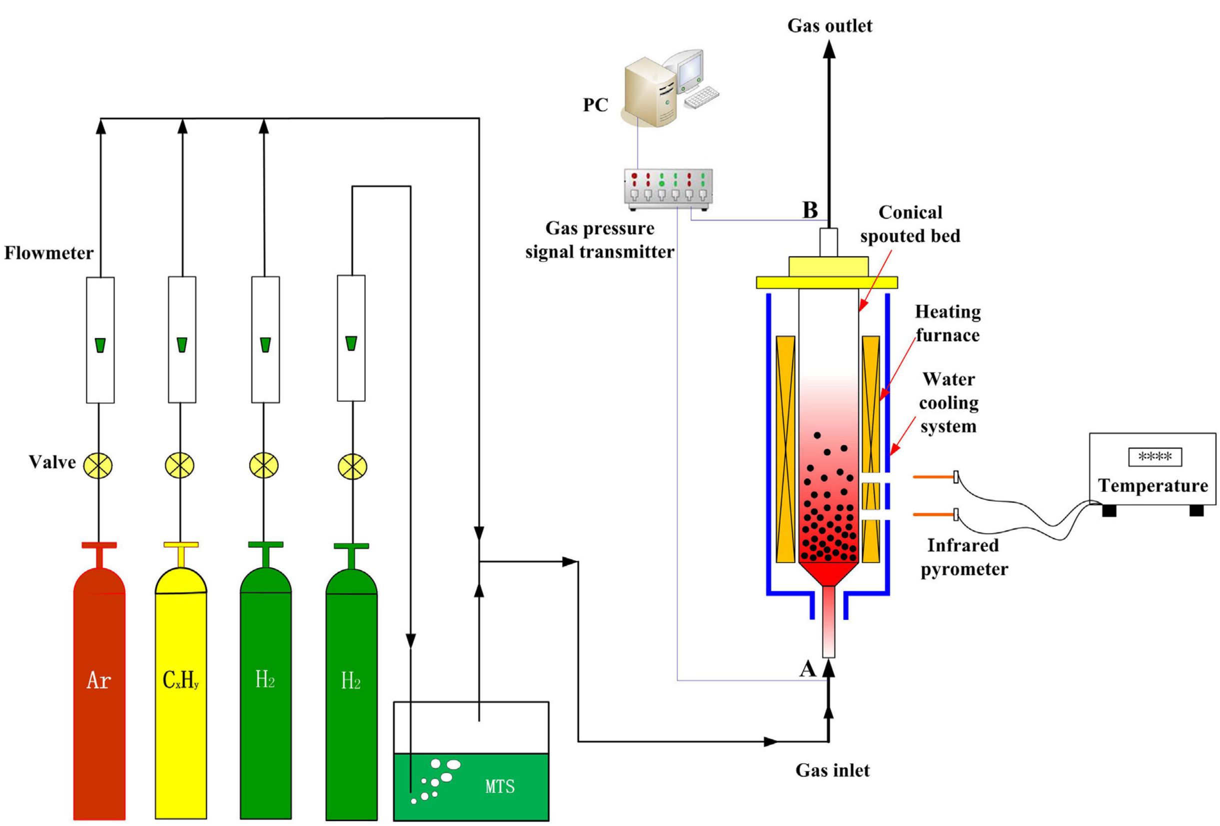

Fig. 1. Illustration of the fluidized bed chemical vapor deposition (FB-CVD) coating system.

Please cite this article in press as: Liu, R., et al. A novel coated-particle design and fluidized-bed chemical vapor deposition preparation method for fuel-element identification in a nuclear reactor. *Particuology* (2016), http://dx.doi.org/10.1016/j.partic.2016.05.009