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Deposition of SiC/Si coatings in a microwave plasma-assisted spouted bed reactor

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Abstract: Silicon carbide (SiC) layers were deposited onto alumina particles in a microwave plasma-assisted spouted bed reactor using methyltrichlorosilane (MTS) and hydrogen mixtures, in argon, as precursor gas feed. The operating parameters studied were enthalpy, gas composition, and pressure. Microwaves were guided from a generator, operating at 2.45 GHz, along a rectangular waveguide intersecting a quartz tube, acting as the reaction zone. A graphite nozzle at the bottom of the tube facilitated the spouting action. Growth rates varied from 50 to 140 μm/h. Overall results indicate that the optimal region for SiC deposition requires relatively high enthalpy (~5 MJ/kg) and pressure (>-60 kPa) conditions, with hydrogen-to-MTS ratios ~5:1. The quality (i.e. crystallinity, particle size, Si/C ratios) of the layers improve at these conditions, at the cost of decreased deposition rates. Characterisation was done by XRD, FTIR, XPS, SEM, TEM and EDX, which assisted in developing colour and morphological charts to indicate the changes as a function of changing operating parameters. A microwave plasma spouted bed reactor is demonstrated to be a viable alternative technique for SiC layer deposition onto microspheres.

Keywords: silicon carbide, microwave plasma, spouted bed, methyltrichlorosilane, particle coating

1. Introduction

The high-temperature mechanical performance of silicon carbide (SiC) is an important factor contributing towards its use as a fission product barrier in tri-isotropic (TRISO) fuel, and its consideration as a host material for high level radioactive waste immobilisation [1]. As a nuclear ceramic, SiC exhibits excellent mechanical properties and dimensional stability under irradiation and is a proposed replacement material for zirconium alloy-based fuel cladding in light water reactors [2]. Next generation high-temperature reactor (HTR) designs currently project operating temperatures up to 1000 ºC which emphasises the importance of a thorough understanding of the SiC layer properties and deposition mechanisms [3]. The current standard method for TRISO fuel fabrication involves chemical vapour deposition (CVD) in a fluidized bed furnace [4]. This technique, although stable and reliable, demands a high-energy input to provide the necessary temperatures needed for the decomposition of methyltrichlorosilane (MTS) to form SiC. The synthesis reaction is shown in Eq. (1), and is thoroughly reported in the literature [5-7]

$$\mathrm{CH_3SiCl_3(l) \rightarrow SiC(s) + 3HCl(g)} \quad (1)$$

Various plasma-enhanced chemical vapour deposition (PECVD) techniques have been investigated for the synthesis and deposition of SiC [5, 8, 9]. Among these techniques, microwave plasma-enhanced CVD (MW-PECVD) has produced promising results [10-14], and provides economic advantages such as decreased energy requirements and reduced equipment footprints [15]. In combination with this plasma technique, fluidised -and spouted bed reactors have also produced promising results for deposited coatings on particles [16, 17] due to the added advantage of high mass- and heat-transfer rates, and high bulk temperatures [18].

In this study, a microwave plasma-assisted spouted bed reactor was used for the *in situ* synthesis and deposition of SiC layers onto alumina spheres in order to investigate MW-PECVD as a viable alternative deposition process for high quality