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Thermal shock fracture of hot silicon carbide immersed in water

Youho Lee [*, 1], Thomas J. McKrell, Mujid S. Kazimi [2]

Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA, 02139, United States

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ABSTRACT

High purity CVD-SiC, considered as a nuclear grade cladding material, exhibits thermal shock tolerance ~1260 °C in room temperature water and beyond it (>1260 °C) in saturated water. Being thinner than the tested specimen thickness (1.5 mm × 2.0 mm), the actual cladding (0.57 mm) is anticipated to exhibit enhanced thermal shock tolerance. This implies that thermal shock alone may not shatter the SiC cladding in reflood. Level of fuel rod internal pressure will be a decisive factor in predicting cladding fracture during reflood. Decreasing water subcooling significantly reduces thermal shock fracture danger of ceramic materials. Thermal shock experiments showed strength retention for both pressureless sintered-SiC and CVD SiC, as well as Al_2O_3 samples quenched from temperatures up to 1260 °C in saturated water. Solid–liquid contacts during nucleate and transition boiling, and boiling incipience upon water bath entering are a highly probable origin of thermal shock fracture in water quenching.

1. Introduction

Silicon Carbide (SiC) is being considered as a potential replacement for the current zirconium-based cladding of LWR fuel to make the fuel more accident-tolerant [1–32]. SiC shows excellent high temperature and irradiation tolerance [11,12,20–27] as well as 2–3 orders of magnitude slower oxidation rate compared to zirconium-based alloy at 1200 °C [9,14,18]. In addition, SiC absorbs fewer neutrons than Zirconium-based cladding [4,5]. Those identified characteristics make SiC a promising cladding candidate for the development of an accident tolerant fuel for LWRs under beyond design basis accidents (DBAs) without a need for large changes in the current core design.

Yet, SiC is a brittle ceramic material that lacks ductility. This implies that the safety evaluation of SiC clad LWR fuel should be considered in a drastically different framework from the current practice for the metallic and ductile Zirconium-based cladding. Retention of cladding ductility is a key safety metric for Zirconium-based clad fuel, given that not enough load-bearing capability is expected by elastic deformation of embrittled phase alone. The SiC fracture, initiated by a local critical flaw, is inherently statistical whereas the Zirconium-based alloy failure occurs in a deterministic manner, thanks to the plasticity-mediated dispersed fracture. Recognizing such a fundamental difference in strength-failure between SiC and Zirconium-based alloy, increasing attention is being made to structural failure analysis of SiC cladding [7,17] at steady-state operation as a starting point.

Under accident conditions, a fuel cladding may experience significant stresses beyond the level of usual steady-state operation due to thermal shock in the reflooding phase of a loss of coolant. In general, thermal shock fracture often sets the maximum service temperature for high temperature ceramic applications below the melting point. Indeed, thermal shock fracture is also a limiting failure mode for the Zirconium-based alloy cladding in accidents involving reflood quenching [33]; brittle fracture caused by oxygen-affected brittle phase and oxide layer (ZrO_2) in the cladding serves as a basis for the current emergency core cooling system (ECCS) criteria for loss of coolant accidents (LOCA). That is, the current 17% equivalent cladding reacted (ECR) limit and 1204 °C of peak cladding temperature are considered to ensure retention of cladding ductility and strength even after thermal shock [34]. In case of SiC, which inherently lacks ductility, occurrence of brittle fracture under reflood quenching should be carefully addressed. The fuel, ECCS, and reactor core designs should ensure that the

* Corresponding author. Korea Advanced Institute of Science and Technology (KAIST), Department of Nuclear and Quantum Engineering, Daejeon, 305–701, South Korea.

E-mail address: euo@kaist.ac.kr (Y. Lee).

[1] The first author's current affiliation is Korea Advanced Institute of Science and Technology (KAIST), Department of Nuclear and Quantum Engineering, Daejeon, 305–701, South Korea.

[2] Deceased, 30th of June 2015. Professor Mujid S. Kazimi passed away while this article was being prepared with his inputs. This work was conducted under his supervision.

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reflood quenching in loss of coolant accidents do not fracture the cladding. It should be noted that in GENIII + reactors, the core is expected to remain covered with water after a LOCA, and thus the fuel does not reach very high temperatures, and no quenching need to be addressed.

In this early stage of the concept development and feasibility study, basic phenomenological aspects of silicon carbide fracture behavior subject to water quenching should be explored. The goals of this study are to (1) experimentally explore thermal shock tolerance of SiC in different subcoolings of water bath relevant to probable reflooding scenarios (saturated and room temperature) and (2) investigate a heat transfer origin (responsible heat transfer modes) of thermal shock fracture.

2. Experiments

An experimental facility, which was originally designed to run pool boiling experiments [35] was modified to bring specimens up to 1500 °C and drive them into a pool of water, as illustrated in Fig. 1. Specimens were suspended in air inside a quartz tube located at the center of a furnace. A K-type thermocouple wire connected to an air-pressure driven alumina rod was used to hold the specimen. By employing bottom-flooding, this experiment has similar experimental designs/conditions to what was used to establish the current Zr cladding safety criteria given in 10 CFR 50.46 [33,34]. A B-type thermocouple reads the temperature adjacent to the outer surface of the quartz tube, in which the specimen is located. The temperature reading was recorded by a data acquisition system (DAS). Temperature calibrations were made with a SiC specimen between this B-type thermocouple reading and the temperature obtained by a thermocouple attached to the sample's surface. Comparing these two temperatures, an empirical relation between the furnace temperature and the true sample surface temperature was established and used to report SiC specimen temperatures. Specimens were suspended inside the furnace until they reached a constant temperature. Then, specimens were quickly inserted into either a room temperature water pool (~22 °C) or a saturated water (~100 °C) pool by the air-pressure driven rod. A high speed video camera was used to record the quenching of the specimens.

The SiC materials used in this experimental study are monolithic tubular pressureless-sintered SiC and bar-shape CVD SiC. The presureless-sintered SiC specimens were directly obtained from Saint-Gobain, Inc.; their density was 3.05 g/cc, and their trade name is SE type Hexoloy. Each specimen had dimensions of 14.0 mm OD, 1.56 mm thickness, and 13.0 mm height. High purity CVD SiCs samples were obtained from Bomas (Somerville, MA). Their density is 3.21 g/cc and each is in a bar shape of the standard ASTM C1161 size: 25 mm × 2 mm × 1.5 mm. Samples were ultrasonically cleaned with detergent added water, deionized (DI) water, acetone, and methanol prior to furnace exposures. Experiments with alumina were also conducted. The alumina specimens were obtained from McMaster-Carr and the name of the product is 'Very High Temperature Nonporous High-Alumina Ceramic' whose density is 3.875 g/cc. Two different dimensions were tested for the alumina specimens: 30 mm (l) × 6.35 mm (w) × 6.35 mm (t), and 30 mm (l) × 3.175 mm (w) × 3.175 (t). Appropriate extrapolations of temperature dependent material properties were made by referring to relevant studies when necessary.

Tubular presureless-sintered SiC specimens in the temperature range from 350 °C to 1265 °C were quenched into either room temperature or saturated water. CVD SiC specimens in a temperature range from 500 °C to 1260 °C were quenched into either room temperature or saturated water. For each temperature condition in SiC experiments, at least two tests (in fact 3–4 on average) were

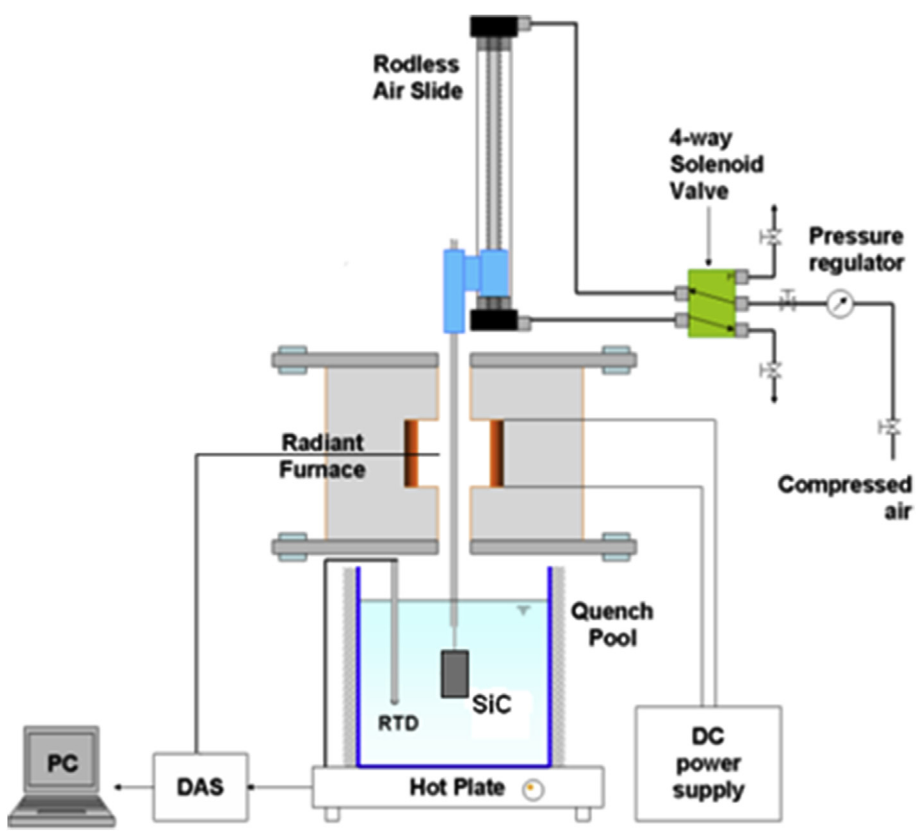

Fig. 1. SiC Quenching experiment facility (DAS: Data Acquisition System, and RTD: Resistance Temperature Detector).