ABSTRACT

Graphite is used as a structural material, neutron moderator and reflector in some designs of nuclear reactors. The core of a prismatic or pebble bed type High Temperature Reactor (HTR) is made up of graphite components. In addition to HTRs, it is also used as a structural material in the Molten Salt Breeder Reactor (MSBR). Air ingress during normal and off-normal conditions is a serious safety concern in High Temperature Nuclear Reactors. Oxidation of graphite, leading to increased porosity, affects its mechanical strength and may lead to core collapse, resulting in a severe accident. During such a scenario, generation of graphite aerosol particles could be the main hazard. Once generated, these particles, often in fine and ultrafine size ranges, may carry radioactivity to far distances for long times. These particles, owing to their higher surface to volume ratio possess an additional inhalation hazard. Ultrafine particles have the potential to enter into respiratory tract and cause damage to body organs. Emitted graphite particles also have the potential to influence radiative balance and thereby, modify atmospheric and oceanic circulations, hydrodynamic cycle and regional precipitation pattern. Therefore, an understanding about the combustion behaviour of graphite, including generation of aerosol under different conditions is very important for human safety and safe reactor operation. Hence, in the present work, the objective was to study the aerosol emission behaviour of graphite during an accident scenario caused by air ingress in a high temperature nuclear reactor by simulating graphite burning in a closed combustion environment. To study the effect of coating in inhibiting oxidation behaviour, Alumina (Al_2O_3) coated graphite samples were also burnt at various conditions.

The aerosol emission behaviour of a material was studied in a high temperature facility that was specifically designed to attain a maximum furnace temperature of 1350 °C with a maximum air flow rate of 25 $Lmin^{-1}$ (at atmospheric temperature and pressure).

Combustible materials like biomass, wood, agricultural residue as well as commercially used materials like graphite can be burnt and characterized using this facility. A cooling and dilution mechanism was incorporated to lower the temperature of the aerosol to the desired value (around 40 °C) for aerosol size measurement using spectrometers. Isokinetic sampling was ensured using a specially designed probe. In addition, for accurate measurement of aerosol, transport losses due to diffusion, thermophoresis and gravitational settling were calculated.

In the present research, firstly, nuclear grade graphite was burnt at constant temperature (in the range 500 - 900 °C) in this facility. Apart from measuring number and mass size distributions of generated particles at different temperatures and flow rates, emission of CO and CO_2 was also monitored. Besides studying the oxidation behaviour at different temperature, the effect of heating rate was also analysed. Heating of the sample was carried out at air flow rates of 10, 15, 20 and 25 $Lmin^{-1}$ at four different heating rates of 2, 4, 6 and 8 °C/min. The concentration of CO_2, CO and particles was evaluated at these test conditions. The uncoated graphite reacted easily with oxygen and rapidly burned away as the temperature exceeded 500 °C. To protect graphite from oxidation at high temperatures, an alumina (Al_2O_3) coating of about 100 microns thickness was deposited on bare graphite. The effect of Al_2O_3 coating on the generation of aerosol particles from coated graphite surface was also investigated. Concentrations of gases (CO and CO_2) and particles were measured at the chamber exit for the test matrix.

In case of graphite burning at constant temperature, the burning of graphite surface was observed as the temperature exceeded 500 °C. Particle concentration and CO gas concentration was found to be maximum at 700 °C. However, with graphite heating at different heating rates, the highest concentration of particulate matter and CO gas was achieved in the lower temperature range (600 - 800 °C). For the case of coated samples,

graphite surface was protected against oxidation till 700 °C. At higher temperatures (800 and 900 °C), surface degradation starts increasing the diffusion of oxygen and oxidation rate. the maximum particles and CO generation occurred at 900 °C. The phenomena of evaporation-condensation and transport of unburnt graphite particles could be linked to the generation of particles in nucleation and accumulation mode size ranges, respectively. This study found that the transition of graphite oxidation reaction framework from progressive to shrinking core model takes place around 700 °C.