

On the oxidation of gaseous iodine at elevated temperatures

T. Kärkelä¹, J. Holm², A. Auvinen¹, C. Ekberg² and J.K. Jokiniemi^{1,3}

¹VTT Technical Research Centre of Finland, Emission Control, Espoo, FI-02044 VTT, Finland

²Chalmers University of Technology, Nuclear Chemistry, Göteborg, SE-41296, Sweden

³University of Eastern Finland, Environmental Science, Kuopio, FI-70211, Finland

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Presenting author email: Teemu.Karkela@vtt.fi

The aim in this study was to examine the oxidation of gaseous inorganic and organic iodine when exposed to gaseous ozone and UV (c-type) radiation. The experiments were conducted with EXSI-CONT (EXperimental Studies on Iodine chemistry – CONTainment) facility. Gaseous iodine was exposed to UV radiation with a radiation wavelength maximum at 185 nm, which is able to generate ozone (O₃) from air efficiently. Additional ozone was produced with an ozone generator. The oxygen to nitrogen ratio and the humidity of the carrier gas was varied in the tests as well. The experiments were conducted at 50 °C, 90 °C and 120 °C. The residence time of the gas flow inside the facility was varied from 1.75 s to 7 s.

The formation and growth of the iodine containing aerosol particles was characterised online with Scanning Mobility Particle Sizer (SMPS), Electrical Low Pressure Impactor (ELPI), Condensation Particle Counter (CPC) and Tapered Element Oscillating Microbalance (TEOM) devices. During the tests the speciation of gaseous reaction products was measured using Fourier Transform Infra-Red analyser (FTIR). After the tests, the concentration of gaseous and particulate iodine fractions was quantified with Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The size, morphology and elemental composition of particles was also analysed using Scanning Electron Microscopy coupled with Energy-Dispersive X-ray spectroscopy (SEM-EDX).

The formation of reaction products was very fast when gaseous iodine was exposed to UV radiation or ozone. The diameter of the formed iodine oxide particles, measured in the gas phase, ranged from dozens to a few hundred of nanometres. The colour of iodine oxide particles formed at 100 °C, and then collected on a plane filter, was observed to be light/yellow. The particles were most likely a mixture of I₂O₄ (diiodine tetroxide) and I₂O₅ (diiodine pentoxide). The formation of I₂O₅ is expected to be the dominant reaction at dry conditions when temperature exceeds 100°C. When the samples were exposed to air, they reacted with the humidity and their colour changed to purple/dark brown. This reaction was very fast, because the diameter of the particles was very small. The uptake of humidity decomposed the IO_x particles to iodic acid (HIO₃) and elemental iodine (I₂) (Dählie, 1964b; Chase, 1996). The diameter and mass concentration of particles increased when the residence time, relative humidity, radiation intensity, ozone concentration or the temperature of the

facility were increased. A model on the oxidation of elemental iodine was derived.

The formation of gaseous reaction products was studied as well. The most important detected species, resulting from the decomposition of gaseous CH₃I, were methanol (CH₃OH) and formaldehyde (CH₂O). When the concentration of ozone was very high, significant formation of secondary gaseous reaction products was observed, e.g. formic acid (CH₂O₂) and methyl formate (HCOOCH₃).

When the coolant of fuel in a nuclear power plant (NPP) is lost, the melting of overheated fuel rods will take place. As a consequence, the vaporized fission products (FPs) are transported through the primary circuit of nuclear reactor and released through a break in the circuit into the gas phase of containment building. There FPs, such as gaseous iodine, are exposed to air radiolysis products as well as to the radiation - such as alpha, beta, gamma and X-ray - itself. This may lead to oxidation of gaseous iodine and subsequent formation of iodine containing aerosol particles. As radiotoxic iodine constitutes one of the major hazards of the possible source term (e.g. Fukushima Daiichi nuclear accident), it is of interest to understand the behavior of iodine in the conditions of severe accident.

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