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Particle Size Measurement of Reaction Product Aerosol of Sodium-Oxygen

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A multi-level scenario simulation system as a safety infrastructure technology is required for design optimization, safety margin adjustment, and innovative technology developments of sodium-cooled fast reactor (SFR). In order to properly implement the Verification and Validation (V & V) of the simulation system, it is indispensable to ensure experimental database of sodium chemistry as specific SFR safety issue. In this study, measurement results of aerosols generated by sodium-oxygen reaction for sodium fire event were reported with the aim of clarifying the radiation heat transport phenomena in the reaction field. The sodium-oxygen counter-flow diffusion flame was formed one-dimensionally above the sodium pool by the reaction between sodium vapour and oxygen. Argon (Ar) including 2% oxygen were introduced to a liquid sodium pool (temperature: 820K) under the reduced pressure condition (0.05MPa). Ar guard flows were employed to stabilize the reaction. The reaction continued more than 600 seconds without any changes in terms of flame shape and position. Aerosol size was measured as a function of Z (the distance from the sodium pool surface) and r (the distance from the center of the sodium pool). Laser Induced Incandescence (LII) and the Mie scattering method using the different wavelength laser beams (405nm, 450nm, 520nm, 532nm, 638nm and 650nm) were employed to ensure the measurement accuracy. Aerosol sizes from several hundred nm to 1 μm were measured in this reaction field and the aerosol size increased toward the sodium pool. This stemmed largely from aerosol growth and polymerization because the flow rate decreased near the sodium pool. It was also confirmed that the size of aerosol measured by LII was in good agreement with the measurement using the Mie scattering method under the same conditions. The refractive index of the aerosol was also evaluated to be 1.42-0.5i.

1. Introduction

Liquid sodium is used as coolant in a sodium-cooled fast reactor (SFR) to transfer heat from its reactor core to a steam generator (SG) because of its excellent heat transport capability. On the other hand, sodium has strong chemical reactivity with oxygen and water vapor. The sodium-water and sodium-oxygen reactions become one of design basis accidents [1]. A multi-level scenario simulation system as a safety infrastructure technology is required for design optimization, safety margin adjustment, and innovative technology developments of SFR. In order to properly implement the

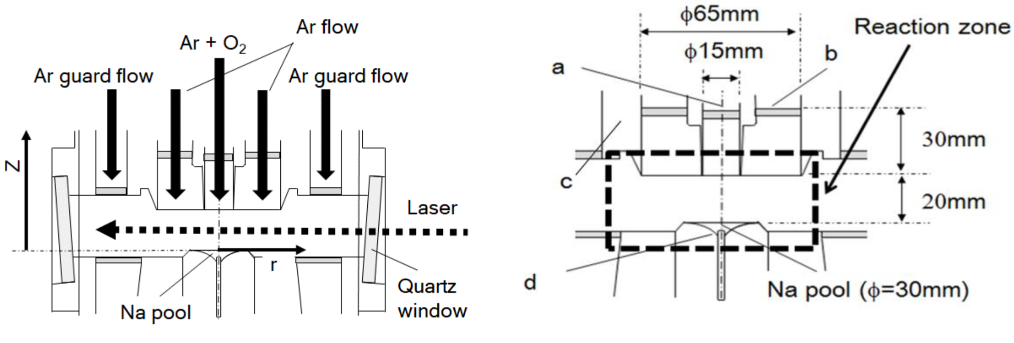
Verification and Validation (V & V) of the simulation system, it is indispensable to ensure experimental database of sodium chemistry as specific SFR safety issue [2]. One of the technical difficulties in developing the evaluation technique of this process has been to clarify the phenomena on sodium-water and sodium-oxygen chemical reactions. In particular, it is important to consider the surface and gas-phase reactions because water vapor and oxygen react with gas and liquid sodium at the boundary surface of reaction fields. The surface reaction mechanism has been investigated theoretically and experimentally by Kikuchi et. al. to reveal the sodium-sodium hydroxide reaction as secondary surface reaction of the sodium-water reaction phenomena [3, 4]. On the other hand, the gas-phase and its relevant reactions were studied mainly as a single-step reaction [5-12]. Deguchi et. al. have developed an elementary reaction set of sodium-water and sodium-oxygen gas-phase reactions which consist of 17 H_2-O_2 and 8 Na-H_2O reactions including 14 species [13]. The theoretical results using it were compared to the measurement by laser diagnostics [14, 15] using the sodium-water counter-flow reaction vessel [13, 14, 15]. In these studies, it was revealed that the major reaction of sodium-water reaction in gas phase was $Na+H_2O \rightarrow NaOH+H$ and OH was produced by $H_2O+H \rightarrow H_2+OH$. The quantitative analysis, however, was not enough to evaluate the products such as aerosols produced by sodium-oxygen reactions in gas phase.

The purpose of this study aims to clarify the sodium-oxygen gas phase reaction path and reaction products. Measurement of aerosols generated by sodium-oxygen reaction for sodium fire event are reported with the aim of clarifying the radiation heat transport phenomena in the reaction field. Aerosol size was measured using sodium-oxygen counter-flow reactions. Laser Induced Incandescence (LII) [16] and the Mie scattering method using the different wavelength laser beams (405nm, 450nm, 520nm, 532nm, 638nm and 650nm) were employed to ensure the measurement accuracy.

2. Experimental Setup

Fig. 1(a) - (b) show an overview of the experimental apparatus. The reaction chamber had four windows (diameter: 50mm) for the measurement of aerosols generated by sodium-oxygen reaction using laser diagnostics [16]. Sodium-oxygen counter-flow reactions were applied in this study. 2 and 4 % oxygen concentrations in Ar were introduced from the pipe (diameter:15mm) to a liquid sodium pool (diameter:30mm, temperature: 820 K) under the reduced pressure condition (0.05 MPa). Ar and Ar guard flows (diameters of 65mm and 130mm, respectively) were employed to stabilize the sodium-oxygen reactions. Experimental conditions are summarized in Table 1. The sodium-oxygen counter-flow diffusion flame was formed one-dimensionally above the sodium pool by the reaction between evaporated sodium and oxygen. The reaction continued more than 600 seconds without any changes in terms of flame shape and position. Aerosols generated by sodium-oxygen reaction were measured as a function of Z (the vertical distance from the sodium pool surface) and r (the horizontal distance from the center of the sodium pool).

Fig. 2 shows the experimental apparatus of aerosol measurements. The sodium-oxygen counter-flow diffusion flame was visualized by a CCD camera using the Mie scattering signals from a 532 nm diode laser. Aerosol characteristics were measured by using LII and the Mie scattering method. LII is a method detecting aerosol number density and size using emission from the heated aerosol by the laser radiation. The experimental apparatus comprises an Nd:YAG laser (Spectra physics, Model PS100, laser power : 100mJ/p, pulse width : 5ns), a spectrometer (JASCO, CT-10S) and an ICCD camera (Andor, iStar DH334T-18U-03). Laser wavelengths of 532nm (second harmonic) was used to generate LII signals. 415 nm and 653 nm LII signals were detected two-dimensionally to evaluate the aerosol size using the ICCD camera. Aerosol size and its refractive index were also evaluated using the Mie scattering signals from diode lasers with the wavelengths of 405nm, 450nm, 520nm, 532nm, 638nm, 650nm as listed in Table 2. Each laser beam was combined by beam combiners to have the same laser path as shown in Fig. 3 and was adjusted to be the same laser power at 100 mW. The Mie scattering signals were also detected by the ICCD camera to evaluate the aerosol size and its refractive index. The experimental Mie scattering signal intensity was corrected by the efficiency of experimental apparatus such as the ICCD camera. The experimental accuracy was confirmed by measuring the Mie scattering signals from the latex particles (Magsphere, Inc, particle diameter : 1μm, refractive index : 1.59) in water and compared to the Mie scattering theory.



(a) Schematic of reaction chamber (b) Detailed diagram of reaction zone

Fig. 1 Experimental apparatus of sodium-oxygen counter-flow diffusion flame

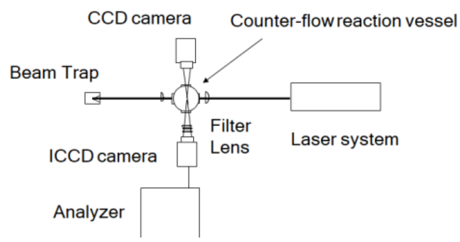


Fig. 2 Experimental apparatus of aerosol measurements

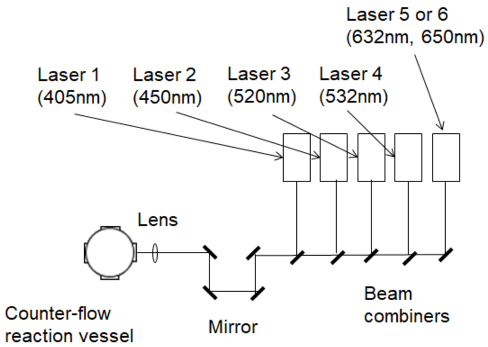
Table 1 Experimental conditions

Item	Temperature	Position (Fig.1(b))	Flow rate
Ar + O_2 [1)]	873 K	a	4.33 x 10^{-6} m^3/s
Ar flow	873 K	b	2.16 x 10^{-5} m^3/s
Ar guard flow	340 K	c	1.33 x 10^{-4} m^3/s
Na pool	820 K	d	-

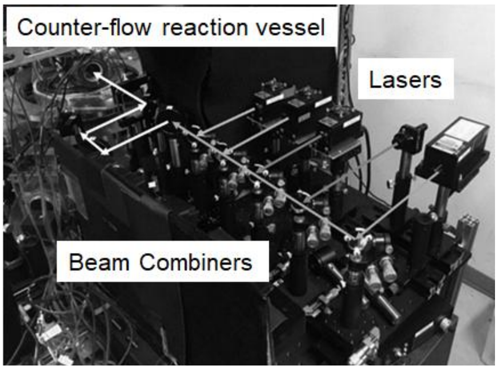
[1)] Concentration of O_2 : 2 and 4 %

Table 2 List of diode lasers

NO.	Wavelength	Maximum laser power
1	405nm	250mW
2	450nm	500mW
3	520nm	700mW
4	532nm	150mW
5	638nm	250mW
6	650nm	250mW



(a) Laser configuration



(b) Picture of laser configuration

Fig. 3 Optical setup of Mie scattering experiment

3. Results and Discussion

The visualization results of sodium-oxygen reaction fields are shown in Fig. 4. In the case of the reaction between sodium and oxygen, there is no luminescence from the reaction [15] and the Mie scattering signals from the reaction products were observed to identify the reaction zone. In case of 2% oxygen concentration, the strong Mie scattering signals were observed at the reaction front between sodium and oxygen at Z=7-8mm.

Fig. 5 shows the time dependent intensity of LII signals. The LII signal intensity has a relation with the cube of aerosol diameter and it decreases with time because of the decrease of the aerosol temperature [15]. The decrease rate of the LII signal depends on the aerosol size can be measured by measuring the decay of the LII signal intensity. 415 nm and 653 nm LII signals were detected two-dimensionally to evaluate the aerosol size with the correction of aerosol temperature effects. The decrease rates of LII signal intensity at different r positions were almost the same. Fig. 6 shows the particle size distributions measured by LII. Aerosols from several hundred nm to 1 μm were measured in this sodium-oxygen reaction field and the aerosol size increased toward the sodium pool. This stemmed largely from aerosol growth and polymerization. These phenomena were eminent toward the sodium pool because the flow rate decreased near the sodium pool [15]. The reaction front was moved toward the sodium pool with the increase of the oxygen concentration and the aerosol growth rate became bigger with the increase of oxygen concentration as shown in Fig. 6(a) and (b). The number density of aerosol increased with the increase of oxygen concentration and it accelerated the aerosol growth and polymerization. The dependence of aerosol size on r was not eminent and it was consistent with Fig. 5.

Fig. 7 shows the comparison of measured and calculated Mie scattering intensity at different wavelengths and aerosol size. The theoretical Mie scattering intensity between 400-800nm was calculated by the Mie scattering theory with the parameters of the aerosol size and its refractive index. The experimental Mie scattering signal intensity was compared to the theoretical Mie scattering intensity using the least-square technique to determine the aerosol size and refractive index. The aerosol size and refractive index (Z=6mm, r=10mm) generated by sodium-oxygen reaction were determined to be 1.02-1.04 μm and 1.42-0.5i, respectively. The measurement results showed good agreement with the measurement using LII under the same conditions.

The aerosol characteristics such as size and refractive index play an important role in the radiation heat transport phenomena in the reaction fields and the measured results were employed for the evaluation of the SFR simulation system.

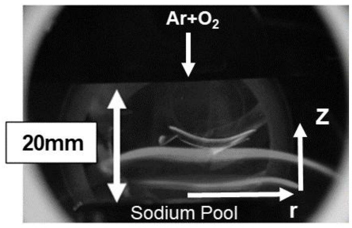


Fig. 4 Visualization of sodium-oxygen reaction (O_2 : 2%)

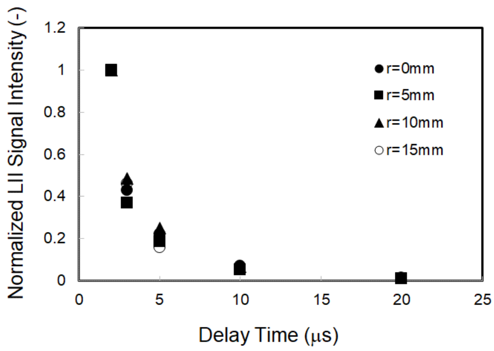
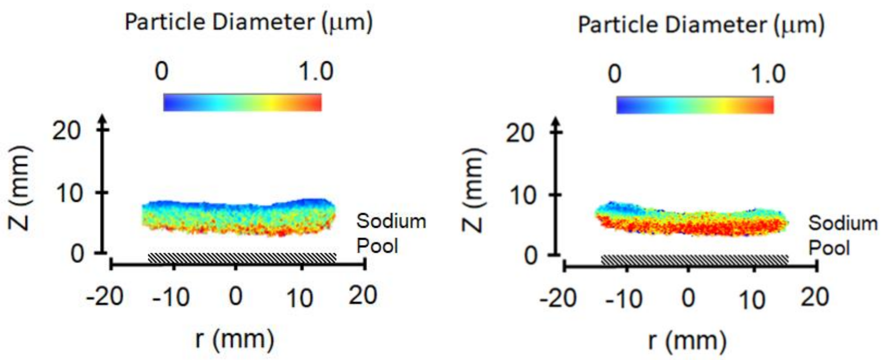
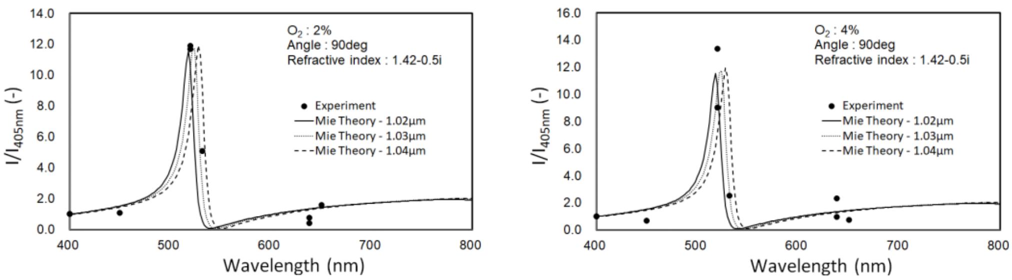


Fig. 5 Time dependence of LII signal intensities (O_2 : 2%, Z=7mm, 405nm)



(a) O_2 : 2% (b) O_2 : 4%

Fig. 6 Particle size distributions Measured by LII.



(a) O_2 : 2% (b) O_2 : 4%

Fig. 7 Comparison of measured and calculated Mie scattering intensity at different wavelengths and size (Z=6mm, r=10mm).

4. Conclusions

LII and Mie scattering methods measurement techniques were applied to the sodium-oxygen reaction fields to clarify the aerosol characteristics generated by sodium-oxygen reaction. The aerosol characteristics play an important role in the radiation heat transport phenomena and the following results were obtained.

1) LII was successfully applied to measure the aerosol size. Aerosols from several hundred nm to 1 μm were measured in the sodium-oxygen reaction field. The aerosol diameter increased toward the sodium pool because of the aerosol growth and polymerization.

2) The experimental Mie scattering signal intensity was compared to the theoretical Mie scattering intensity to determine the aerosol size and refractive index. The aerosol size and refractive index (Z=6mm, r=10mm) generated by sodium-oxygen reaction were determined to be 1.02-1.04 μm and 1.42-0.5i, respectively. The measurement results showed good agreement with the measurement using LII under the same conditions.

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