

Experimental study on spatial temperature distribution characteristics of sodium droplet combustion

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Abstract: In the operation of the sodium-cooled fast reactor, the accident caused by the leakage and combustion of liquid sodium is common and frequent. In this paper, the combustion and spatial temperature distribution characteristics of sodium droplet were studied by carrying out the experiments of the oxidation and combustion under different conditions of initial temperatures (200°C-350°C) of the sodium droplets and oxygen concentrations (12%-21%). The oxidation and combustion behaviors were visualized by a set of combustion apparatus of sodium droplet and a high speed camera. The results show that the temperature of sodium droplet rises slowly in surface oxidation stage, but rapidly in pre-ignition stage and combustion stage. There is a delay time in the change of the spatial temperature compared to that of sodium droplet. However, they arrived peak at almost the same time. The peak temperature of sodium droplet sufficient combustion could roughly reach 600-1000°C. The spatial temperature distribution fits well with exponential function of descent and tend to a stable value at 20mm away from the sodium droplet.

Keyword: sodium-cooled fast reactor; liquid sodium; combustion; spatial temperature distribution

1 Introduction

The liquid sodium is used as a coolant in liquid metal fast reactors (LMFRs) for its thermal hydraulic characteristics [1]. However, when exposed to water or air, the high chemical reactivity could cause a potential sodium fire hazard [2]. At present, the accident caused by the leakage and combustion of liquid sodium is common and frequent [3]. The research on sodium fire is basis for study, design, construction and operation of sodium cooled fast reactor. What's more, the sodium droplet combustion is the basic of various forms of sodium fire research, and it is of great significance to study the combustion characteristics of sodium droplet [4-6].

At present, scientists have carried out some experimental studies on sodium fire in the world, however, there are a few studies on sodium droplet combustion. Richard studied the combustion characteristics of static sodium droplet firstly in 1968. His study shows that the sodium droplet combustion follows the D²-law that the diameter of the sodium droplet decreases linearly with time [7]. Then, in 1969, Krolkowski improved the sodium droplet combustion model [8]. In his research, reaction rate was controllable, and the result shows that the

burning rate of sodium droplet and the flame temperature decrease with the decrease of oxygen concentration on the surface of droplet. When the oxygen concentration is less than 4%, the sodium vapor cannot burn, and they emphasize that the size of the sprayed sodium droplet is the most important factor of the change in container pressure in a closed container during spray fire. In 1979, the American scientist Morewitz studied the relationship of the sodium droplet burning rate with the droplet diameter and the falling velocity by experiments [9]. Japanese scientist Yuasa studied the characteristics of sodium combustion in the air under different environmental humidity in 1985. He found that spontaneous combustion temperature of sodium droplet increases with the increasing of humidity. Sodium droplet combustion can be divided into two stages: the first stage is the surface reaction stage, which is mainly related to humidity. In this stage, the temperature increase is relatively slow; the second stage is burning stage, which is related to the oxygen concentration on the surface of the sodium droplet, in this stage, the temperature increases quickly [9]. Miyahara and Ara of Japan carried out a study on combustion of single falling droplet and recorded the combustion characteristics throughout the whole process, and measured the weight and falling speed of droplet in 1998 [10]. Another Japanese scientist

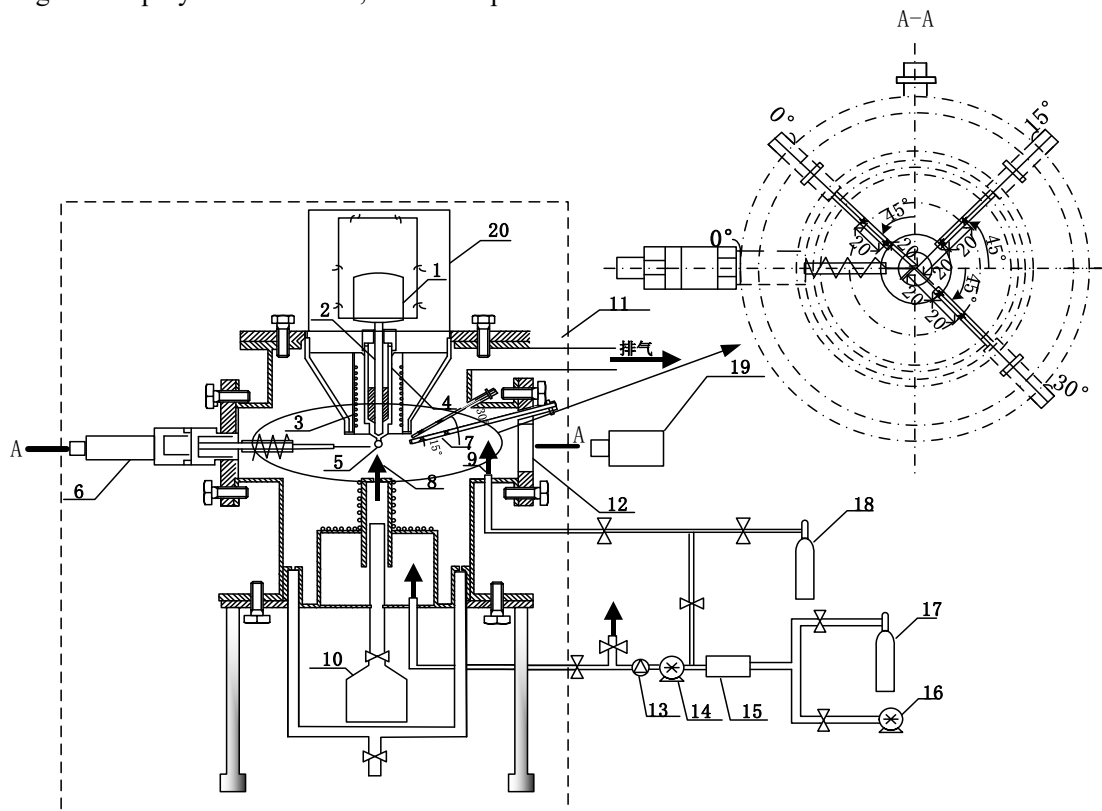
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Makino found that the obvious factors affecting the ignition temperature of sodium droplet are the initial temperature, the diameter of sodium droplet, oxygen concentration and the relative speed by studying the combustion characteristics of free-fall sodium droplet in 2005 [11]. Nishimura analyzed the effect of columnar oxides in combustion and study the oxidative combustion and re-combustion of sodium droplet in 2011 [12]. These studies have made large amount of works. However, the existing research are not systematic and lack detailed experimental data. What's more, there are few experiments to study the spatial temperature distribution characteristics of the sodium droplet combustion. In the process of calculating the spray sodium fire, the lumped

parameter method is wildly used without considering the interaction between sodium droplets. The study of the spatial temperature distribution of the sodium droplet could be helpful to simulate the sodium columnar fire and spray fire more accurately.

In this paper, the experiments were carried out under the condition of different initial temperature (200°C-350°C) and different oxygen concentration (12-21%) to study the spatial temperature distribution characteristics of sodium droplet combustion. These studies could provide technical support for the safety evaluation and preventive measures on different kinds of sodium fire accidents.



1-motor; 2-piston; 3-heater; 4-crucible; 5-sodium droplet; 6-thermocouple; 7-spatial thermocouples; 8-mixed gas; 9-argon; 10-the sodium tank; 11-glove box; 12-observation hole; 13-oxygen concentration controller; 14-gas pump; 15-gas mixer; 16-air pump; 17-nitrogen; 18-argon; 19-speed camera; 20-organic glass.

Fig.1 Experimental apparatus.

2 Experiments

2.1 Experimental Apparatus

The experimental apparatus of sodium droplet combustion consisted mainly of a droplet combustion device, a glove box, equipment for photographing the combustion process and a gas supply system as shown in Fig.1.

The combustion vessel consisted of two dependent chamber components. A crucible with a nozzle to form the sodium droplet, an electric wire around the crucible to preheat the sodium and a piston for extruding liquid sodium. A motor was mounted on the upper flange to move the piston up and down. Under the nozzle, a tube was set for the gas supply. The viewing was positioned at the height of the

nozzle for the observation of the liquid sodium droplet. In order to measure the sodium droplet temperature, a thermocouple (outside sheath diameter: 0.5mm) that could move in horizontal direction was installed under the nozzle. Nine thermocouples were set at the angle of 0°, 15° and 30° and in the distance of 10mm, 20mm and 40mm to measure the spatial temperature distribution. And all the thermocouples were connected to the data collector (HIOKI-LR8402 E.E). The precision was $\pm 0.8^{\circ}\text{C}$, and the minimum data acquisition interval was 10ms. The measurement accuracy of the thermocouple was $\pm 0.75^{\circ}\text{C}$ between 0°C to 800°C.

The examination gas of arbitrary oxygen fraction could be supplied by mixing nitrogen with the stand gas (oxygen: 21%, argon: 99%). The under chamber was the combustion chamber. The combustion vessel was located in a glove box that could control the oxygen and moisture fractions. The video images were recorded by the high-speed video camera (100 fps).

2.2 Experimental procedure

When the oxygen concentration in the glove box was lower than a predetermined value, the sodium was put into the crucible, the crucible was placed in a heating sleeve and the organic glass was closed. At the tip of the nozzle, a liquid sodium droplet was formed through the slow depression of the piston by the motor. The horizontal diameter of sodium droplet was around 5mm. The tip of thermocouple was position at the edge of sodium droplet after the temperature of sodium was stabilized. Argon gas valve was turned off and the valve of the mixed gas was opened so that the pre-proportioned certain concentration gas could be led into the combustion chamber. The flow rate was set to 650ml/min at the bypass line outlet. At the same time, video images were recorded by high speed camera and the data were collected by the data collector.

2.3. Experimental Conditions

The experiments are mainly to study the effect of the oxygen concentration, the initial temperature of sodium droplet on the ignition, combustion and the spatial temperature distribution characteristics. The

initial temperature of sodium droplet was varied from 200°C to 350°C. In consideration of the coolant temperature range of LMFR, the oxygen concentration was 12%, 16% and 21%, as shown in Table 1. At 0s, the examination gas was led to the combustion chamber.

Table 1 Experimental condition (environment atmosphere temperature 180°C).

Initial sodium temperature [°C]	Oxygen concentration [vol%]		
	12%	16%	21%
200	Case1a	--	--
250	Case2a	Case2b	Case2c
300	--	--	--
350	Case4a	--	--

3 Experimental results and analysis

3.1 spatial temperature distribution with the combustion process

It can be observed from Fig.2 that the combustion process can be divided into 3 stages: surface oxidation stage, pre-ignition stage and combustion stage. Surface oxidation stage: as shown in Fig.2, after the gas was supplied, the droplet surface was covered with a thick white oxide layer from 0s to 2s, and in this stage, the temperature rising of sodium was slight. Surface oxidation stage lasted about 5s, then it was wrinkled on the surface rapidly, and sodium droplet gradually came into the pre-ignition stage. Pre-ignition stage: It could be observed from Fig.2 that sodium droplet began to wrinkle and the shape change enormously after the oxide layer was formed on the surface. We could also find that there was column oxide on the surface of the droplet. The temperature of sodium rose obviously in this stage even though the flame couldn't be observed. Combustion stage: At 10s, the flame was a starting point in this stage, and we could find from Fig.2 that the combustion reactor was the gas phase around the liquid droplet. At 19s, the temperature of sodium droplet rose to the peak (630°C) and released the yellow-white flame. The whole process of the combustion stage lasted about 20s and it could be found that the burning product was yellow oxide at 60s.

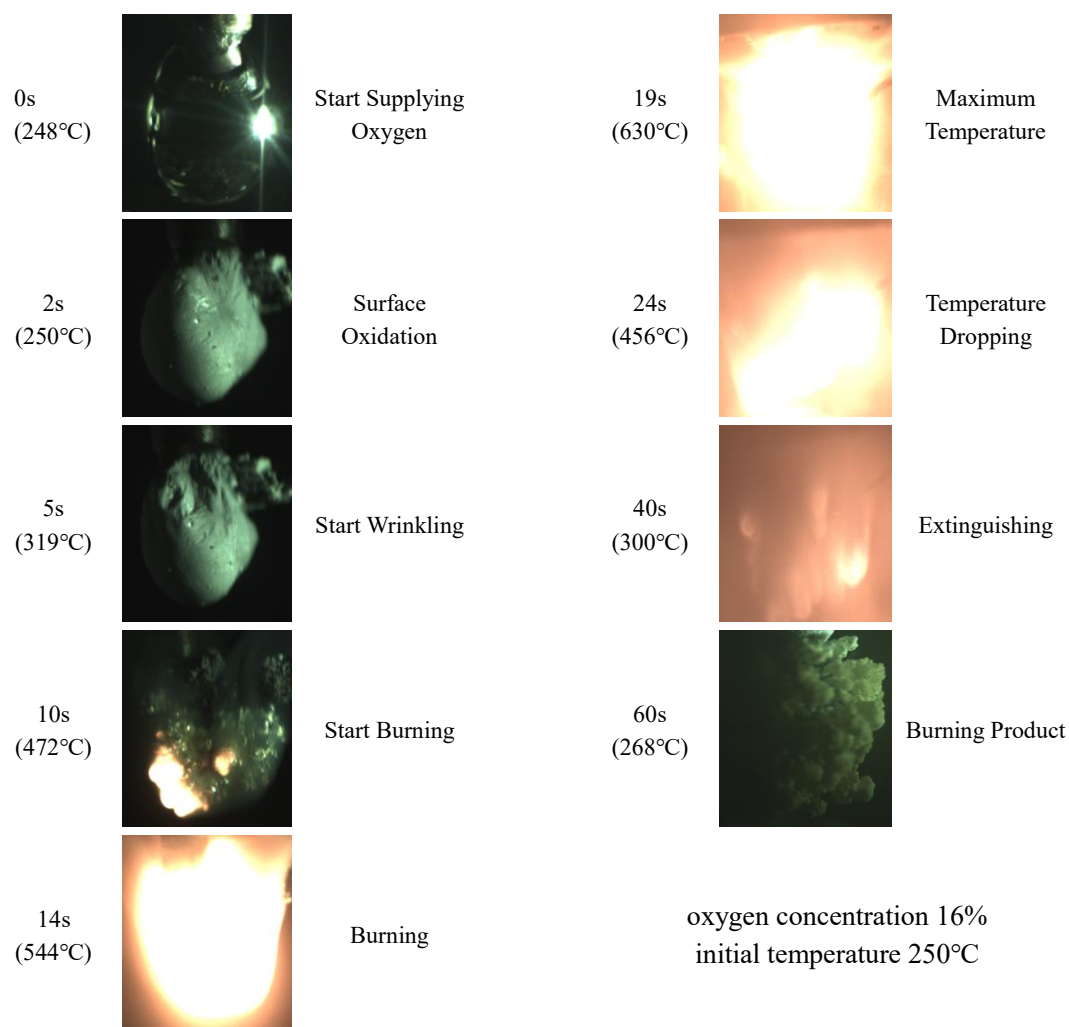


Fig.2 Sodium droplet combustion.

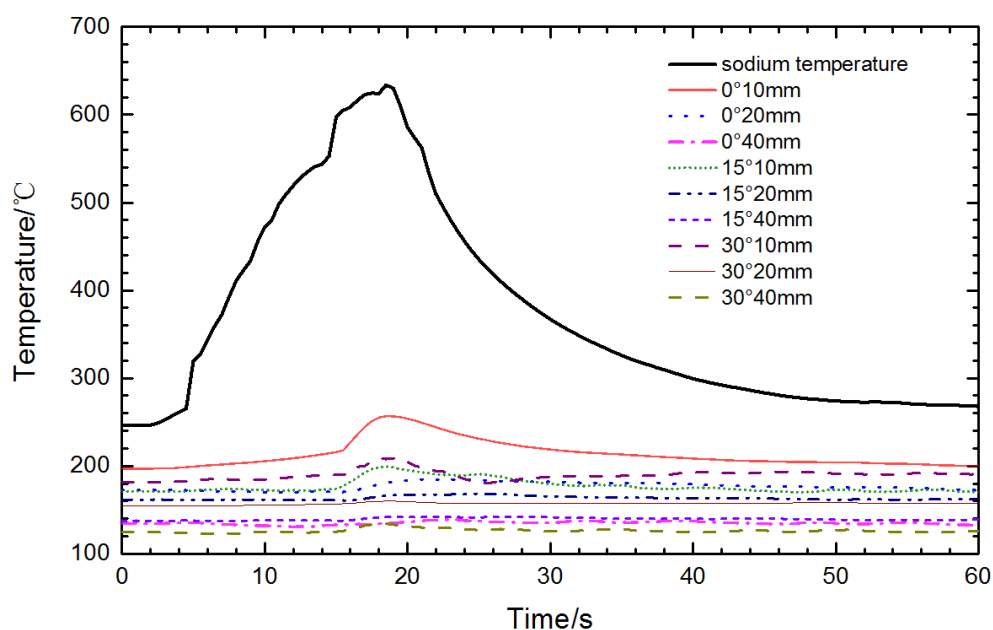


Fig.3 spatial temperature distribution (initial temperature 250 °C, oxygen concentration 16%).

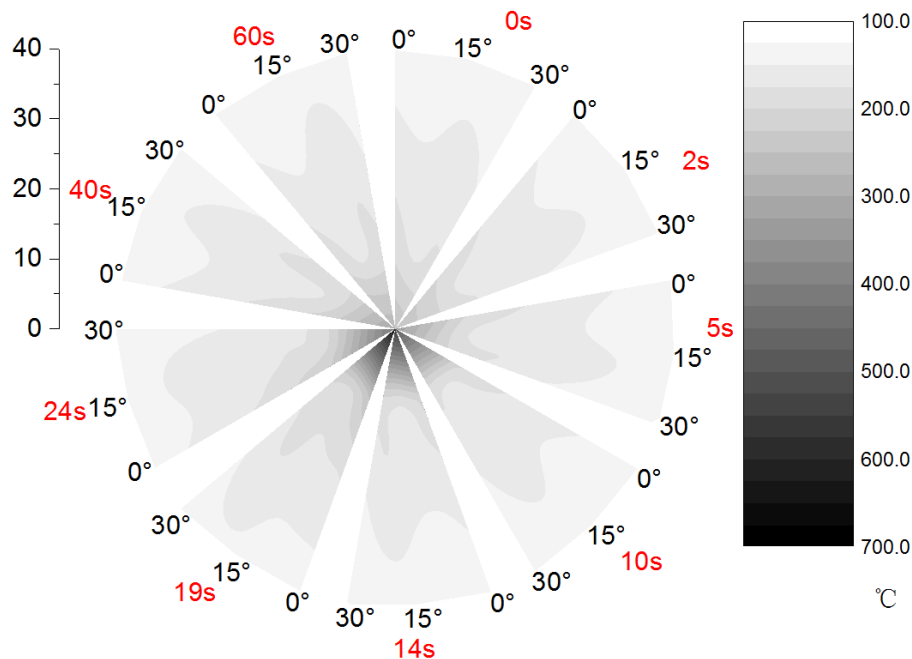


Fig.4 Spatial temperature distribution field (initial temperature 250 °C, oxygen concentration 16%).

3.2 The spatial temperature distribution in oxidation and combustion of sodium droplet

Figure 3 shows the spatial temperature distribution at nine spatial measuring points that respectively in the distance of 10mm, 20mm and 40mm from the sodium droplet and at the angle of 0°, 15°, and 30° when the initial temperature is 250°C and the oxygen concentration is 16%. The temperature rose slowly from 0s to 5s after oxygen supplied and change severely from 6s. From Fig.2 and Fig.3, it can observe and the combustion last about 20s, and the peak temperature of sodium was 630°C at 19s. The spatial temperature began to rise at 14s, later than sodium. This is because the temperature of sodium began to rise when the heat of spatial measuring point absorbs from sodium droplet surface more than that moved by air current. It can also be observed from Fig.3 that the time of spatial temperature and sodium arrived the peak almost at the same time. After 40s, the sodium temperature and the spatial temperature both change slightly and tend to a stable value, and almost all the spatial measuring temperature are less than 200°C at 60s. The spatial temperature at 40mm from droplet almost unchanged and are all less than 20°C.

Figure 5 shows the spatial temperature distribution field through linear interpolating. At different time point, we can observe the spatial temperature

distribution at different distances and angles. It can be found from Fig.3 and Fig.4 that the spatial temperature was lower when the distance was farther because the heat of spatial measuring point absorb from sodium droplet is much less when the distance is further. It can also be found that the spatial temperature in the direction of 0° was the highest among all the spatial measuring points temperature in the distance of 10mm from the droplet. This is because the combustion of droplet bottom surface is more severe as shown in Fig.2. Figure 4 shows that the spatial temperature decreased significantly between 0mm and 10mm from the droplet. However, when the distance was further than 20mm, the temperature of different measuring points had small difference.

3.3 Spatial temperature distribution under different initial temperature and oxygen concentration

Figure 5 shows the distribution of the maximum temperature and fitting curves at different distance from the droplet in the 0° direction when oxygen concentration is 12% (0 mm represent sodium droplet). As shown in Fig.5, with 21% oxygen, the peak temperature of sodium droplet reaches 390°C, 600°C and 760°C when initial temperature is 200°C, 250°C and 350°C, and the temperature in first experiment is relatively low because of insufficient

combustion. The effect of the droplet on spatial temperature decrease rapidly with increasing distance from droplet, spatial temperature is relatively high at 10mm and 20mm from droplet. But the temperature at 40mm is less than 200°C. It is found that the theoretical curve fits well with exponential function, and the fitting formula of the maximum spatial temperature is embedded below.

- (1) Oxygen concentration is 12% and ambient temperature is 180°C

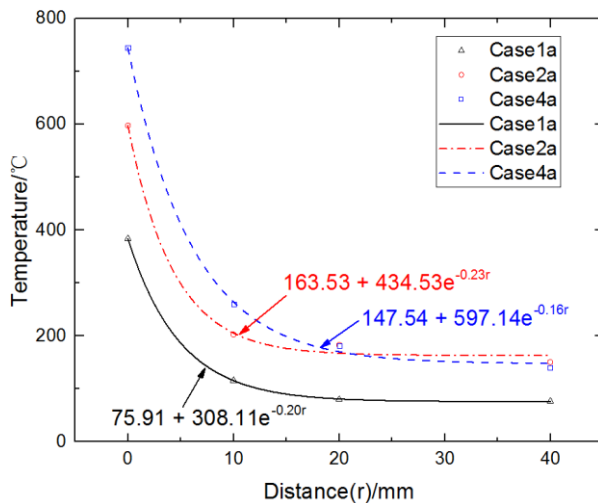


Fig.5 Spatial maximum temperature distribution at 0°
(Oxygen concentration 12%).

3.4 Spatial temperature distribution under different oxygen concentration

Figure 6 shows the distribution of the maximum temperature and fitting curves at different distance from the droplet in the 0° direction when initial sodium temperature is 250°C. As shown in Fig.6, when the initial sodium temperature was 250°C, the peak temperature of sodium droplet reached 600°C, 760°C and 1000°C respectively when oxygen concentration was 12%, 16% and 21%. The effect of droplet on spatial temperature decrease rapidly with increasing distance from droplet, spatial temperature reached 360°C at 10 mm from droplet, but the temperatures at 40 mm are very close in different situations and mostly less than 200°C. It is found that the theoretical curve fits well with exponential function, the fitting formula of the spatial maximum temperature is embedded below.

- (1) Initial sodium temperature is 250°C and ambient temperature is 180°C:

Case2b: $T(r) = 145.90 + 587.56e^{-0.16r}$
(0 mm ≤ r ≤ 40 mm, $T_0 = 250^\circ\text{C}$, $C_0 = 16\%$);

Case1a: $T(r) = 75.91 + 308.11e^{-0.20r}$
(0 mm ≤ r ≤ 40 mm, $T_0 = 200^\circ\text{C}$, $C_0 = 12\%$);
Case2a: $T(r) = 163.53 + 434.53e^{-0.23r}$
(0 mm ≤ r ≤ 40 mm, $T_0 = 250^\circ\text{C}$, $C_0 = 12\%$);
Case4a: $T(r) = 147.54 + 597.14e^{-0.16r}$
(0 mm ≤ r ≤ 40 mm, $T_0 = 350^\circ\text{C}$, $C_0 = 12\%$);
 $T(r)$: the maximum spatial temperature, °C;
 T_0 : initial sodium temperature, °C;
 C_0 : oxygen concentration.

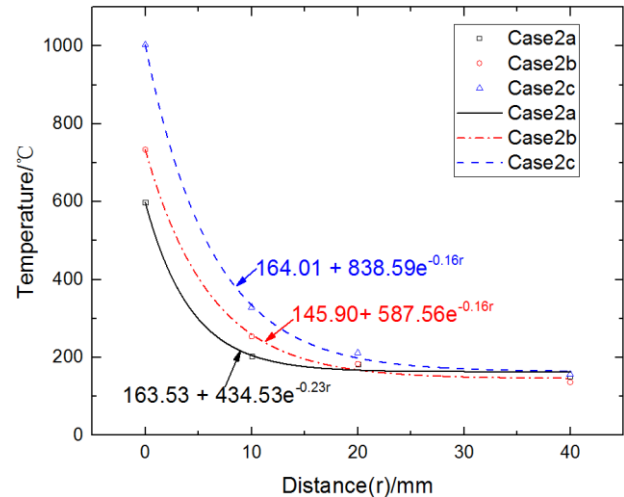


Fig.6 Spatial maximum temperature distribution at 0°
(initial temperature 250 °C)

Case2c: $T(r) = 165.92 + 831.56e^{-0.17r}$
(0 mm ≤ r ≤ 40 mm, $T_0 = 250^\circ\text{C}$, $C_0 = 21\%$);
 $T(r)$: the maximum spatial temperature, °C;
 T_0 : initial sodium temperature, °C;
 C_0 : oxygen concentration.

4 Conclusion

In this research, the combustion behavior and the spatial temperature distribution characteristics were studied by a series of experiments. The conclusions are summarized as follows:

1. After the gas supplied, the process of sodium oxidation and combustion can be divided into three basic stages: surface oxidation stage, pre-ignition stage and combustion stage. The characteristics of oxidation and combustion were different in different stages.
2. The temperature of sodium droplet rise slowly in surface oxidation stage, but rise rapidly in pre-ignition and combustion stages. Because the temperature of the spatial measuring points did not rise until receiving enough heat transferred

from sodium droplet, there was a delay time in the change of the spatial temperature compared to that of sodium droplet. However, spatial temperature and sodium temperature arrived the peak at almost the same time, and tend to a stable value finally.

3. The peak temperature of sodium droplet during sufficient combustion was between 600-1000°C. The initial temperature and oxygen concentration would change the peak temperature of sodium. We found that the spatial temperature distribution fits well with exponential function of descent. The higher the temperature of sodium droplet, the higher the spatial temperature. The spatial temperature decrease with increasing distance from the droplet. However, the spatial temperature changes severely at 10mm from the droplet but change slowly and tend to be stable at 20mm away.

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References

- [1] XU, M.: Safety Analysis on Fast Reactor. Beijing: China Atomic Energy Press, 2011.
- [2] MOREWITZ, H.A.: Sodium Spray Fires. Nucl. Eng. Des. 1979, 55(2): 275-281.
- [3] IAEA: Liquid Metal Cooled Reactors: Experience in Design and Operation, IAEA-TECDOC-1569, IAEA, Vienna (2007).
- [4] SAITO, N., LIAO, C., and TSURUDA, T.: Ignition and Extinguishment of Sodium Fires in Air Diluted by Nitrogen. Rep. Natl. Res. Inst. Fire Disaster, 2001.
- [5] DU, H.O., WANG, R.D, and HU, W.J.: The experimental research on the sodium spray fire. Chin. J. Nucl. Sci. Eng. 2011, 31(1): 41-47 [in Chinese].
- [6] ZHANG, Z.G., PENG, K.W., and HUO, Y., *et al.*: Experimental study on combustion characteristics of sodium fire in a columnar flow. J. Nucl. Sci. Technol. 2014, 51(2): 166-174.
- [7] RICHARD, J.R., DELBOURGO, R., and LAFFITTE, P.: Spontaneous ignition and combustion of sodium droplets in various oxidizing atmospheres at atmospheric pressure. Symp. Combust. 1969, 12(1):39-48.
- [8] KROLIKOWSKI, T., LEIBOWITZ, L., and IVINS, R.: The reaction of a molten sodium spray with air in an enclosed volume part II: theoretical model. Nucl. Sci. Eng. Math. 1969, 38(2), 161–166.
- [9] H A Morewitz. Sodium Spray Fires. Nucl. Eng. Des. 1979, 55(2): 275-281
- [10] YUASA S. : Spontaneous ignition of sodium in dry and moist air streams. Symp. Combust. 1985, 20(1):1869-1876
- [11] MAKINO, A, and FUKADA, H. : Ignition and combustion of a falling single sodium droplet. Proc. Combust. Inst. 2005, 30(2), 2047-2054.
- [12] NISHIMURA Masahiro, KAMIDE Hideki, and OTAKE Shiro, *et al.* : Features of Dendritic Oxide during Sodium Combustion. Nucl. Sci. Technol. 2011, 48(12): 1420-1427
- [13] ZHANG, Z.G, SUN, S.B, and LIU, C.C. *et al.*: Experiment study on oxidation and combustion characteristics of sodium droplets. At. Energy Sci. Technol. 2015, 49(4): 667-673.