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Experimental Determination of the Spontaneous Nucleation Temperature of Sodium[1]

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The superheat limit of sodium was determined using a submillisecond laser-pulse heating technique. Specimens of 1 g of liquid sodium were encapsulated and heated with a laser pulse through a thin tungsten window. A pyrometer measured the increase in surface temperature. Rates of the temperature rise were between 2000 and 7000 $K \cdot ms^{-1}$. Incipient boiling of the sodium resulted in a break in the slope of surface temperature. Using a numerical solution of the heat equation, the measurements gave a value of 2128 K for the spontaneous nucleation temperature with a standard deviation of ± 61 K. Systematic errors contribute ± 28 K to the total inaccuracy.

KEY WORDS: high temperatures; homogeneous nucleation; laser heating; pulse heating; spontaneous nucleation; superheat limit; sodium; transient techniques.

1. INTRODUCTION

The spontaneous nucleation temperature, T_{SN}, of sodium is of interest for the safety analysis of large sodium-cooled nuclear reactors. This parameter appears to play an important role in estimating the mode of thermal interaction between molten core materials and liquid coolant [1, 2]. As a first estimate, Zimmer [3] obtained a value of 2060 ± 160 K from a study of alumina-dropled injection into sodium. Thus, a more straightforward determination of T_{SN} seems desirable.

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Spontaneous nucleation refers to homogeneous or gas-free heterogeneous nucleation because of density fluctuations rather than nucleation from preferred sites [4]. In appropriate experiments T_{SN} corresponds to the homogeneous nucleation temperature of the liquid if the surfaces being in contact with sodium are completely wetted. It can be determined as the maximum superheat temperature accessible by delayed boiling. Following the results of a study on explosive boiling of water and organic liquids [5], maximum superheating of sodium should occur at heating rates of some 1000 $\text{K} \cdot \text{ms}^{-1}$. Such rates are easy to achieve with pulsed laser heating in a suitable experimental procedure.

2. METHOD AND APPARATUS

The present experimental procedure consists of rapid laser-pulse heating of an encapsulated sodium sample while measuring the power input and the surface temperature rise. Figure 1 schematically shows the measurement apparatus. A cylindric capsule (14 mm in diameter), shown at the lower right side of Fig. 1, encloses 1 g of pure sodium. On the inner side of the capsule, the tantalum front plate has a thin window of 5-mm aperture that consists of a smooth 50-μm tungsten foil.

Within an inert gas-filled chamber an induction coil (400 MHz) preheats the capsule to a constant temperature of 1000 K. In a series of experiments

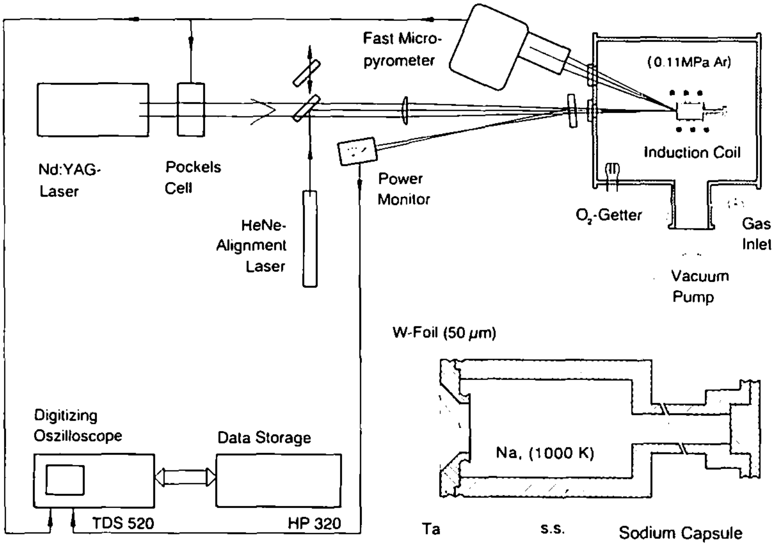

Fig. 1. Schematic diagram of the apparatus.

a 1-ms Nd:YAG-laser pulse rapidly heats a distinct area of the tungsten foil up to 3000 K. The heating rates vary between 10^3 and 10^4 K · ms^{-1}. At comparable rates the heated foil transfers heat to the adjacent sodium layer that becomes superheated. When the temperature of the liquid reaches its superheat limit, incipient boiling interrupts the continuous heat transfer, resulting in a break in the surface temperature slope. Considering heat conduction, the measured surface break temperature yields the superheat-limit temperature, i.e., the spontaneous nucleation temperature of the sodium.

A fast spectral micropyrometer measures the increase in surface temperature in the center of the heated zone utilizing the known spectral emissivity data of tungsten [6]. To keep the tungsten surface clean and unoxidized the chamber is filled with pure argon (impurities, <1 ppm). In addition, a glowing zirconium getter serves to remove residual oxygen. The spectral micropyrometer is equipped with a silicon detector (EG&G, Type YAG 444) and an interference bandpass-filter (Oriel). Its center wavelength is 940 nm at a bandwidth of 43 nm. The temperature resolution of the instrument is ± 2 K at 2500 K, while the rise time is limited to 2 μs. A sight telescope allows an accurate adjustment of the measurement spot (0.35 mm in diameter) controlled by a HeNe-alignment laser. The calibration of the pyrometer is repeatedly checked with a tungsten strip lamp (GEC, Type 10/G) having been calibrated by the PTB (Physikalisch Technische Bundesanstalt, Berlin). The total accuracy of the pyrometer, including the calibration error, is ± 8 K at a radiance temperature of 2200 K. However, the determination of the real surface temperature additionally is subjected to the uncertainties that result from erroneous values of the spectral emissivity. An error of ± 2%, e.g., in the spectral emissivity additionally, leads to an uncertainty of ± 10 K at the same radiance temperature.

The Nd:YAG laser (LASAG, Model LAK 400/LV 480) is multimode operated, yielding a uniform spatial power distribution (0.8 mm in diameter) in the focal plane on the tungsten surface. A Pockels cell (Gsänger, Model DPZ8) suppresses the initial spiking of the laser pulse and sets the pulse length. A silicon detector (EG&G, Type YAG 444) monitors the time resolution of the laser pulse power. To avoid an early destruction of the tungsten window the maximum surface temperature is limited to 3000 K. Exceeding that temperature the pyrometer signal switches the Pockels cell via a comparator. The pyrometer signal and the reading of the power monitor are digitally stored with 11 bit resolution for evaluation (Tektronix, Model TDS 520).

3. MEASUREMENTS AND RESULTS

3.1. Specimens

Four sodium capsules were prepared for use in measurements. Each of them was filled with pure sodium, leaving a small cavity to allow thermal expansion compensation during heating. The sodium was carefully refined (KfK/IMF-III, Karlsruhe) up to residual impurities of oxygen <5 ppm and other metals <1 ppm. To accomplish a tighly welded junction between the tungsten foil (Goodfellow Corp.; purity, 99.95) and the steel jacket (AISI, Type 316L), an intermediate ring of tantalum was introduced. Welding was done with laser and electron-beam heating, respectively. The filled capsules were degassed and sealed finally by electron beam welding.

3.2. Procedure

Before applying laser pulse heating to a preheated sodium capsule in an actual measurement, the capsule was subjected to thermal cycling around

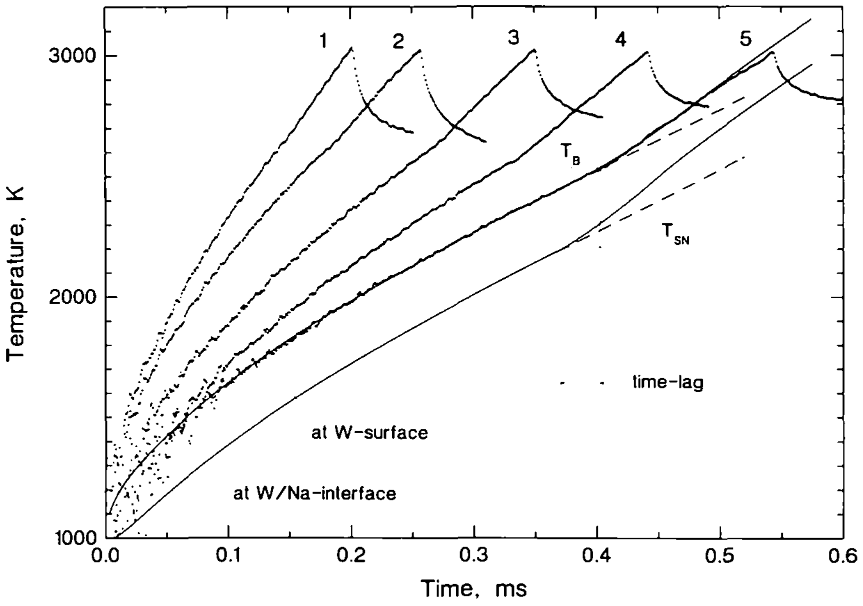

Fig. 2. Typical temperature rises measured in five experiments at the tungsten-foil surface of the sodium capsule belonging to different laser power densities: (1) Na1-08, (2) Na3-04, (3) Na1-15, (4) Na1-20, and (5) Na1-21 (cf. Table II). As an example of evaluation, No. 5 shows the corresponding temperature curves calculated for the tungsten-foil surface and the tungsten sodium interface, respectively (solid lines).

1000 K. This procedure reduces already existing centers of vapor formation, e.g., residual gas bubbles, in the preheated sample. Each capsule should work in a series of measurements. However, to avoid destruction of the foil by advancing recrystallization, the position of the laser heated zone on the tungsten window must be changed after a few experiments. Two capsules have already failed during preheating. They had to be rejected because a leak had occurred in the welding seam between tantalum and steel. This seam proved to be most critical.

Figure 2 shows typical temperature rises measured at the center of the laser heated zone during five experiments. The curves represent different laser power densities in decreasing order from left to right. In all the experiments the laser heating had ceased at 3000 K. The scatter of the digitized signals reveals the sensitivity of the pyrometer, allowing reliable temperature measurements above about 1500 K. The break appearing in the slope of each temperature curve indicates incipient boiling of the sodium due to spontaneous nucleation at the tungsten/sodium interface. If this interface is insulated, no heat can dissipate and the temperature within the tungsten foil will rise more rapidly. The break is clearly visible, but it appears less pronounced at higher power densities. This fact finally leads to an upper limit of the rate of the temperature rise of some 7000 $K \cdot ms^{-1}$ up to which an evaluation seems practicable.

3.3. Evaluation

Sodium boiling arises after a heating time of 0.1 to 0.4 ms. After this time the distance that the heat wave has penetrated the sample is small compared to the transverse dimension of 0.8 mm of the uniformly heated surface area. At least in the center of the heated area within the pyrometer measurement spot the heat flow can be considered linear. To a good approximation, therefore, the spontaneous nucleation temperature T_{SN} can be calculated by solving the one-dimensional heat transfer equation. In the numerical solution the temperature dependence of the thermal data is considered using time steps of 1 ns and mesh cells 0.33 μm in width. The calculation begins with an estimate of T_{SN}. If the temperature of the tungsten/sodium interface reaches this value, the heat transfer to sodium diminishes. With a certain time lag the change in the heat flow induces a break in the surface temperature slope. By trial and error the proper value of T_{SN} is found when the calculated break temperature T_B matches the measured one.

The evaluation of T_{SN} from the measured surface temperature curves is based on the thermal data of sodium and tungsten found in the

Table I. Coefficients of Eq. (1) for the Temperature Dependency of Material Data, $Y(T)$

$Y(T)$	A_0	$A_1 \times 10^2$	$A_2 \times 10^5$	$A_3 \times 10^9$	$A_4 \times 10^{12}$	Range of validity
Cp_{Na} ($J \cdot kg^{-1} \cdot K^{-1}$)	1712.9	−124.43	113.81	−453.32	106.61	$400 < T < 2300$ K
ρ_{Na} ($kg \cdot m^{-3}$)	968.53	−1.9942	−31.527	179.76	−35.570	$400 < T < 2300$ K
K_{Na} ($W \cdot m^{-1} \cdot K^{-1}$)	109.60	−6.5694	1.5556	−2.2377	0.	$400 < T < 2400$ K
Cp_W ($J \cdot kg^{-1} \cdot K^{-1}$)	133.37	−1.2795	5.2913	−28.967	5.3806	$300 < T < 3200$ K
ρ_W ($kg \cdot m^{-3}$)	19447.	−32.273	0.	0.	0.	$300 < T < 3200$ K
K_W ($W \cdot m^{-1} \cdot K^{-1}$)	188.37	−11.268	5.9750	−16.157	1.7244	$600 < T < 3200$ K

literature. These are the specific heats Cp_{Na} [7, 8] and Cp_W [9], the densities ρ_{Na} [7, 8] and ρ_W [10], and the thermal conductivities K_{Na} [7, 8] and K_W [11]. The polynomial fit, Eq. (1), describes their dependence on temperature. In Table I the respective coefficients are given.

$$Y(T) = A_0 + A_1 T + A_2 T^2 + A_3 T^3 + A_4 T^4 \qquad (1)$$

To calculate both the absorption of the laser light at 1064 nm and the emission of the thermal radiation at 950 nm, emissivity values of tungsten were

Table II. Experimental Results on the Spontaneous Nucleation Temperature of Sodium

Expt No.	Laser power density (10^9 W · m^{-2})	Break-temp. at W surface T_B (K)	Spontaneous nucl. temp T_{SN} (K)	Rate of temp. rise at T_{SN} (K · ms^{-1})	Time lag between T_B and T_{SN} (ms)
Na1-22	2.02	2508	2184	2620	0.028
Na1-21	2.04	2538	2207	2650	0.029
Na1-24	2.30	2420	2065	3110	0.022
Na1-23	2.40	2450	2079	3260	0.023
Na1-20	2.41	2571	2189	3300	0.025
Na1-19	2.54	2549	2153	3450	0.025
Na1-15	2.98	2674	2216	3710	0.024
Na1-16	2.96	2695	2231	3990	0.025
Na1-30	3.42	2559	2055	4870	0.020
Na1-29	3.47	2615	2098	4910	0.022
Na3-06	3.51	2634	2106	5000	0.022
Na1-31	3.68	2627	2096	5240	0.019
Na3-04	3.76	2677	2109	5400	0.023
Na1-13	3.86	2772	2190	5540	0.023
Na1-14	4.00	2637	2053	5860	0.020
Na3-10	4.60	2739	2073	6790	0.020
Na1-08	4.70	2745	2060	6930	0.021
Na2-08	4.80	2832	2131	7030	0.022

used for $1600 < T < 2800$ K as given by De Vos [6]. The values were extrapolated from 1000 to 3200 K for the laser and the pyrometer wavelength as a function of the temperature and the radiance temperature, respectively.

Curve 5 in Fig. 2 shows an example of evaluation. The calculated surface temperature curve fits the curve of the experimental data of the measured temperature rise. To produce the best fit, the laser power density has been adapted below the break temperature in the range up to some 2000 K. The resulting temperature curve at the tungsten/sodium interface is nearly parallel to the surface curve. It reveals the temperature increase above T_{SN} and the time lag between T_{SN} and T_B. To evaluate T_B, the second derivative of the surface temperature curve has been considered which passes a maximum at T_B. The broken lines show the projection of the curves hypothetically expected to appear if no boiling occurs.

3.4. Results

Table II lists the results of experiments that have been carried out with different laser power densities. It contains the spontaneous nucleation temperature appearing from different rates of the temperature rise at the

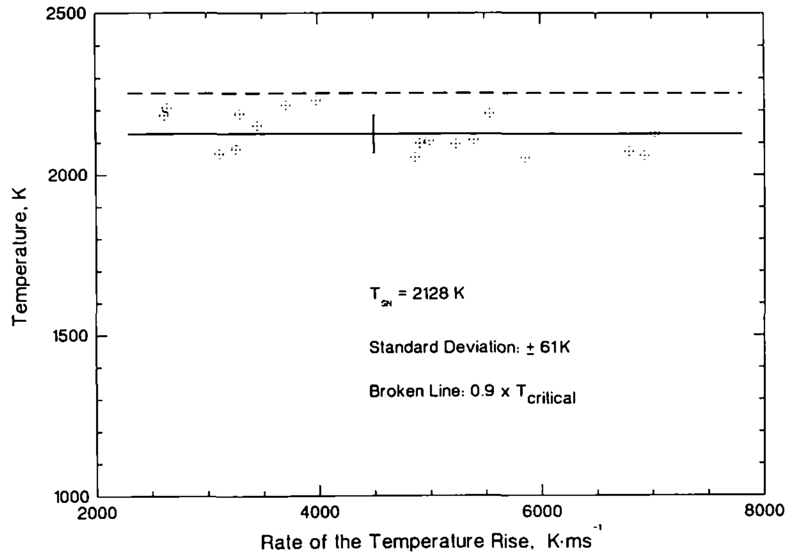

Fig. 3. Spontaneous nucleation temperature of sodium, T_{SN}, evaluated at different rates of the temperature rise. The dashed line corresponds to the thermodynamic limit of superheat [13].

Table III. Propagation of the Single Errors of the Thermal Data in Evaluation of T_{SN}

Thermal data	Cp_{Na}	Cp_W	ρ_{Na}	ρ_W	K_{Na}	K_W	ε_W
Systematic errors of data (%)	±10	±5	±5	±2	±20	±10	±2
Errors transmitted to T_{SN} (%)	±0.4	±0.3	±0.2	±0.2	±0.7	±0.8	±0.4

tungsten/sodium interface. Beside the measured break temperature, the table shows the laser power density being incident in this stage near the break point. The time lag between T_{SN} and T_B corresponds to the characteristic time τ that a thermal disturbance needs to travel through the tungsten foil of thickness d. Taking $\tau = Cp\,\rho d^2/(4\,K)$ and inserting the thermal data of tungsten for the experimentally covered temperature range yields a mean value of 0.023 ± 0.003 ms. This is close to the experimental values in Table II. The ambient pressure in the capsules was <0.1 MPa. Figure 3 reveals the resultant values of T_{SN} to be independent of the rate of the temperature rise within the experimentally covered range of 2000 to 7000 $\text{K} \cdot \text{ms}^{-1}$. The evaluation yields a mean value of

$$T_{SN} = 2128 \pm 61 \text{ K}$$

The standard deviation of ±61 K which was obtained from 18 measurements does not include the systematic errors originating in the thermal data. Deduced from the critical temperature of sodium, 2503 K [12], the broken line in Fig. 3 approaches the thermodynamic limit of superheat [13].

3.5. Systematic Errors

Systematic errors of the thermal data are estimated to $Cp_{Na} \pm 10\%$, $Cp_W \pm 5\%$, $\rho_{Na} \pm 5\%$, $\rho_W \pm 2\%$, $K_{Na} \pm 20\%$, and $K_W \pm 10\%$. In addition, deduced from a series of test measurements, the uncertainty in the emissivity of the tungsten foil, ε_W, amounts to ±2%. Table III shows the propagation of the single errors in the evaluation of T_{SN}. These errors are independent of each other. Together with the instrument error of ±0.4%, they contribute to the total error of T_{SN} by the quadratic mean of their values transmitted, i.e., with ±1.3%. The total error caused by the uncertainties in the thermal data is thus ±28 K. This value may be reduced by eventual improvements of the database.

4. CONCLUSION

Beyond the thermodynamic limit of superheat the liquid phase can no longer exist. The practically attainable superheating of sodium is lower. It is limited by the appearance of spontaneous nuclei at the hot tungsten wall. For temperatures above 1000 K it can be assumed that tungsten is completely wetted by sodium [14]. Thus, the spontaneous nucleation threshold of 2128 ± 61 K found in the experiments comes very close to the homogeneous nucleation temperature of the liquid.

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