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THE REACTION BETWEEN WATER DROPLETS AND MOLTEN SODIUM

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Abstract—The behaviour of measured quantities (0.02 ml) of cold water (295 K) when injected into hot sodium (480–730 K) has been followed using high speed (5000 frames s^{-1}) ciné film. From the results, a model of the reaction system based on transient and steady state film boiling processes is constructed, and equations are established which enable the temperatures, rates of reaction and life times of water droplets in hot sodium to be estimated. The application of the results to the sodium filled heat exchanger circuits of nuclear reactors is discussed.

NOMENCLATURE

a,	radius of droplets dispersed in sodium [m];
$c_{1,2}$,	specific heat of media 1,2 (Fig. 3) [J kg^{-1} K^{-1}];
d,	inside diameter of syringe [m];
h,	height of droplet above hot surface (Appendix 3) [m];
k,	thermal conductivity of water–hydrogen mixture [W m^{-1} K^{-1}];
k_W, k_H,	thermal conductivities of water vapour and hydrogen respectively [W m^{-1} K^{-1}];
k_0,	thermal conductivity of water vapour at 373 K [W m^{-1} K^{-1}];
r,	cavity radius on symmetrical model [m];
r_0,	radius of contact between droplet and hot surface [m];
u,	characteristic velocity [equation (7)] [m s^{-1}];
$\bar{u}$,	average vapour velocity out from gap h [m s^{-1}];
x,	distance from interface [m];
D,	mutual diffusion coefficient of water vapour and hydrogen (Table A1) [m^2 s^{-1}];
G,	ideal gas constant [8317 J kg mol^{-1} K^{-1}];
L,	latent heat of vaporisation of water [$L_0 + 2.5(373 - T_S)$] kJ kg^{-1}. ($L_0 = 2256.67$ kJ kg^{-1});
P_0,	one atmosphere (1.01325×10^5 N m^{-2});
P_W,	vapour pressure at droplet surface (normalised to 1 atm);
R, T,	dimensionless quantities r/a, ut/a respectively;
T_∞,	temperature of bulk sodium [K];
T_B,	373 K;
T_S,	droplet temperature [K];
V_0,	(0.02×10^{-6} m^3) volume of injected water;
V_T,	total volume of vapour generated [m^3];
W,	evaporation rate [kg mol s^{-1}].

Greek symbols

α_{Wt},	thermal diffusivity of cold water [1.44×10^{-7} m^2 s^{-1}];
ρ_v,	density defined by equation (A3.4) (Appendix 3) [kg m^{-3}];
ρ_W,	density of cold water [1000 kg m^{-3}];
$\rho_{1,2}$,	density of media 1,2 (Fig. 3);
σ,	surface tension of cold water [0.075 N m^{-1}];
μ,	average viscosity of the water–hydrogen mixture (Table A1) [N s m^{-2}];
ν,	kinematic viscosity of cold water (295 K) [1.010×10^{-6} m^2 s^{-1}].

1. INTRODUCTION

WATER leaks into the sodium circuit of a secondary heat exchanger in a fast reactor can result in severe corrosion to adjacent plant surfaces, leading to extensive damage. Although details of the process remain to be settled, corrosion is known to be most intense at the sodium-water reaction interface [1]. It is important to know how much of the plant has been put at risk, and therefore how far round the sodium circuit water can be carried before being consumed.

Substantial gaps in the present understanding of the basic parameters which control the reaction rate of water in sodium, however, hamper a precise evaluation of this problem. To gain a better insight into these factors, a series of experiments was carried out to measure the behaviour of known volumes of cold water when injected into hot sodium. The experimental results and their interpretation in terms of heat fluxes and vapour films are described in Sections 2 and 3. It is proposed that steady film boiling develops for the water–sodium interaction, and the consequences of this are examined in Sections 4 and 5. In particular, a simple model is constructed which allows estimates to be made of the lifetimes of water droplets immersed in hot sodium.

2. EXPERIMENTS AND RESULTS

The apparatus is shown in Fig. 1. After filling with dry argon, sodium (65 ml) in store A was melted and filtered through C into the quartz reaction vessel E. Water (0.02 ml at 295 K) was injected into the sodium as a stream of fine droplets from syringe H. Vessel E was fitted with optical windows, and subsequent events were recorded on high speed (5000 frames s^{-1}) ciné film.

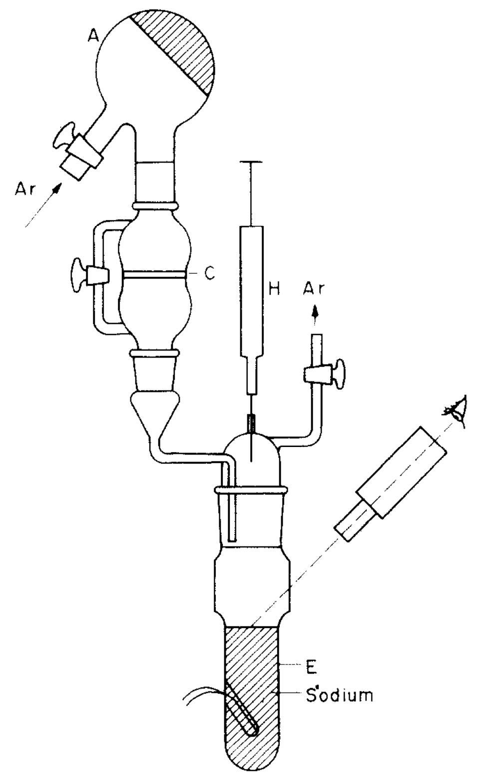

FIG. 1. Apparatus used to study the behaviour of water injected into liquid sodium.

At temperatures between 480 and 650 K, the sodium surface rose twice, firstly on completion of the water injection (0.1 s), and then as a mushroom of sodium (0.15–0.20 s) which was thrown to a height 2–4 cm (Fig. 2a). In all cases timings are given from the moment of initial water impact on to the sodium surface.

A few large water drops with a volume approximately equal to the original (0.02 mm^2) were ejected from the sodium as the mushroom collapsed (Fig. 2b). Events in the range 650–730 K were essentially similar, except that there was no initial rise in the sodium surface. The mushroom was formed after only 0.1 s.

Strikingly different results were obtained from a contrasting series of measurements in which cold water was injected into chemically inert silicone oil at temperatures between 420 and 720 K. The surface of the oil rose steadily with the generation of a mass of fine vapour bubbles, until, after 0.1–0.2 s, the reaction vessel was completely filled (Fig. 2c). The rate of surface rise increased with oil temperature, and there was no evidence of droplet coalescence.

3. INTERPRETATION OF THE EXPERIMENTAL RESULTS

Firstly, it is necessary to determine whether or not transient boiling effects in the initial stages could account for the observations. The time t over which transient energy transfer occurs is found by equating the thermal boundary-layer thickness $2(\alpha_{Wt}t)^{1/2}$ to the droplet radius a.

$$t = a^2/4\alpha_{Wt}. \tag{1}$$

If, however, there is circulation in the drop due to relative motion of the ambient vapour, the time taken for the hydrodynamic boundary layer to reach the centre is

$$t_H = a^2/4\nu \tag{2}$$

and the ratio $t/t_H = \nu/\alpha_{Wt} = Pr$, the Prandtl number. For water this number decreases with temperature ($Pr = 6.8$ at 295 K, $Pr = 1.8$ at 373 K).

Assuming the column of water (diameter $d = 0.15$ mm) breaks up by Rayleigh instability [2], water enters the sodium as a stream of droplets with mean radius $a = 0.95d = 0.142$ mm. Thus t, t_H = 0.035, 0.005 s respectively at 295 K, and 0.030, 0.017 s respectively at 373 K. Equilibrium conditions in the droplet should be established by about 0.04 s.

Heat transfer from the hot sodium should sustain transient boiling if the temperature of the interface is maintained above the boiling point of water throughout this initial time. At times less than 0.04 s when all boundary layers are smaller than the radius of the drop, the interfacial temperature between the water (medium 1) and sodium (medium 2) may be calculated from the one dimensional model shown in Fig. 3.

$$\left.\begin{aligned} T_1 &= A_1 + B_1 \operatorname{erf} x/2(\alpha_1 t)^{1/2} \\ T_2 &= A_2 + B_2 \operatorname{erf} |x| 2(\alpha_2 t)^{1/2} \end{aligned}\right\} \tag{3}$$

where A_1, A_2, B_1, B_2 are determined from the initial values and continuity of heat flux at $x = 0$. If T_∞ is the bulk sodium temperature, and T_0 (295 K) that of the cold water, the interface temperature at $x = 0$ ($t \neq 0$) is

$$\text{where}\quad \left.\begin{aligned} T_1 &= T_2 = (\gamma T_\infty + T_0)/(1+\gamma) \\ \gamma &= (k_2 c_2 \rho_2 / k_1 c_1 \rho_1)^{1/2}. \end{aligned}\right\}. \tag{4}$$

For the water–sodium interaction, $\gamma = 6.014$ and interface temperatures corresponding to bulk sodium temperatures of 480, 594 and 726 K are 454, 551 and 665 K respectively. In all cases this temperature is

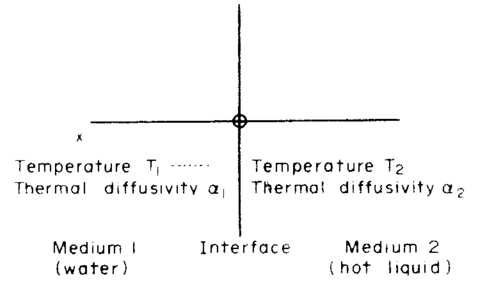

FIG. 3. Model of heat transfer processes at interfaces.

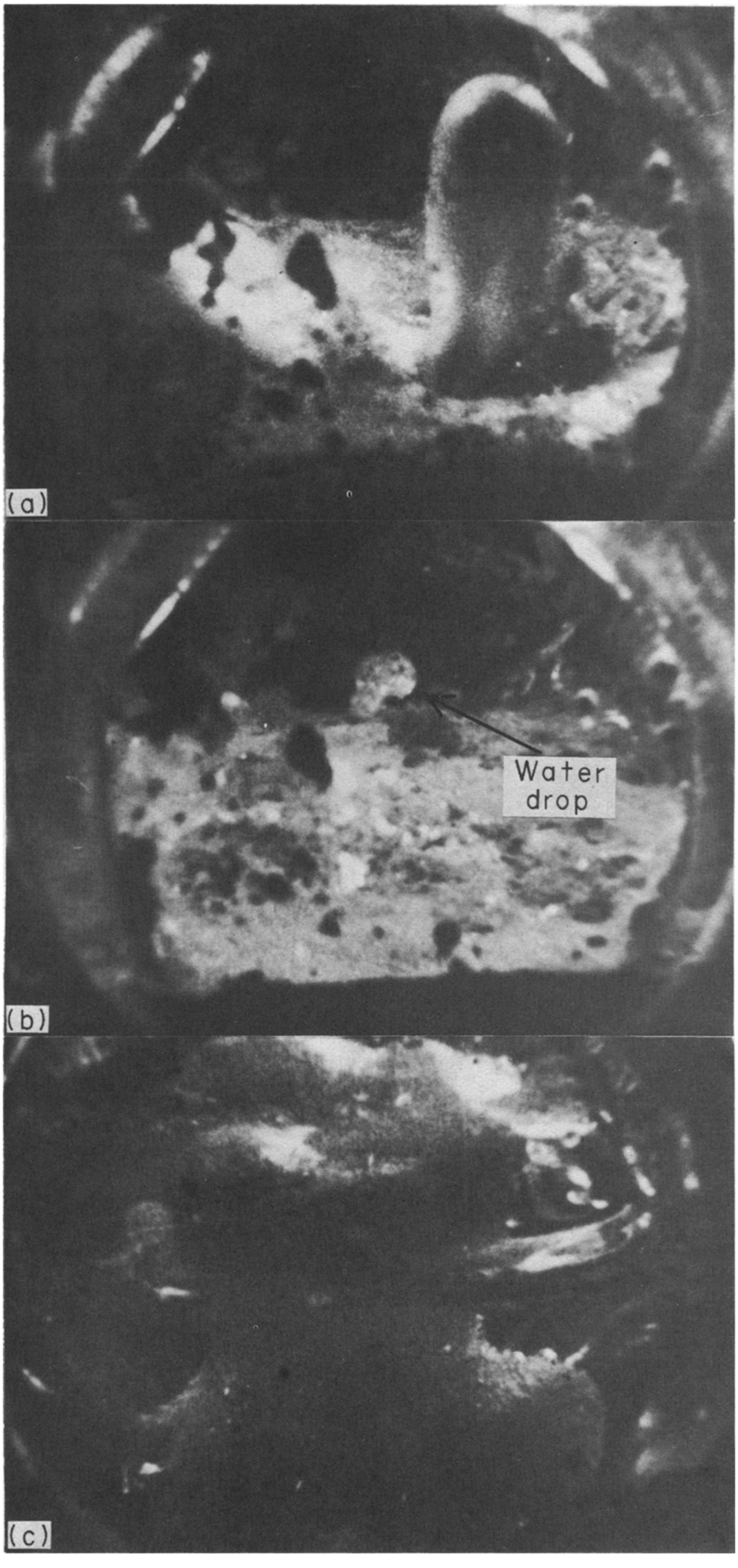

FIG. 2. Experimental observations: (a) Sodium mushroom on point of collapse (sodium temperature 594 K). (b) Water droplet ejected from the sodium following the collapse of the sodium mushroom. (c) Interaction of water droplets with silicone oil at 572 K.

much higher than the boiling point of water, so that transient boiling of the droplets should be maintained.

The initial surface rise (around 0.1 s) is believed due to transient film boiling. Studies by Board *et al.* [3] suggest that for surface temperatures below ~ 620 K (at 1 atm), the vapour film due to transient boiling is unstable, and periodically collapses permitting large heat fluxes into the drop. However, this vapour film tends to stabilise with increasing surface temperature, and the average heat flux is reduced. As observed, the initial surface rise should become less marked.

Support for this model comes from the behaviour of the water–silicone oil system. Due to the low thermal conductivity of the oil (0.142 $W m^{-1} K^{-1}$ compared with 76 $W m^{-1} K^{-1}$ for sodium) large temperature differences are required to maintain the transient heat fluxes. Corresponding to bulk oil temperatures of 423, 507, 717 K, the interface temperatures from equation (4) are 325, 345 and 395 K respectively. With the exception of 717 K (the boiling point of the oil) the interface temperatures lie well below the boiling point of water, so there is no initial vapour generation. In this situation direct contact occurs between the two liquids, and there is no vapour film to moderate the heat flux into the drop [3]. Once sufficient heat has entered the drop, boiling occurs with vigorous, rapid vapour growth.

4. STEADY STATE BOILING IN THE WATER–SODIUM SYSTEM

The initial presence of a vapour film round each droplet opens the way for a steady state situation in which film boiling is dominant. The continuing generation of vapour by this process probably accounts for the formation (0.15–0.20 s) of the sodium mushroom. As confirmed by the subsequent ejection of large drops (Fig. 2b), the high surface tension of sodium (0.17 $N m^{-1}$) should encourage droplet coalescence during this stage. Figure 4 illustrates a possible sequence of events.

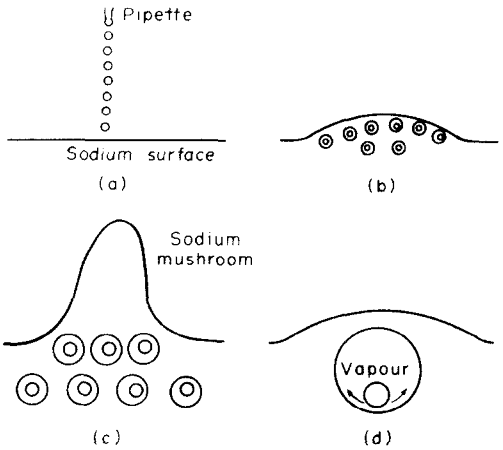

FIG. 4. Experimental system. (a) Droplets impinge on the sodium surface. (b) Initial transient boiling of the droplets. The vapour causes a small surface rise. (c) Steady film boiling before coalescence. Growth of the sodium mushroom. (d) The Leidenfrost effect. After coalescence a single large drop rests at the foot of a vapour cavity.

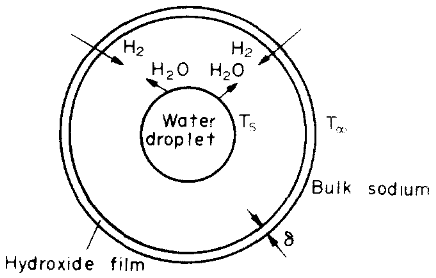

FIG. 5. Film boiling with chemical reaction.

A model of film boiling is shown in Fig. 5, in which a spherically symmetrical mixture of water vapour and hydrogen surrounds a droplet at temperature T_S. The following assumptions are made.

(i) Heat and mass transfer take place under steady state conditions.

(ii) The pressure inside the vapour cavity is approximately 1 atm. This is confirmed from the Rayleigh equation, provided the velocity of the cavity wall is less than ~ 1 $m s^{-1}$.

(iii) The temperature of the surface of contact is equal to the bulk sodium temperature. The high thermal conductivity of sodium ensures that the heat fluxes for steady film boiling can be supported for small temperature differences.

(iv) The concentration of water vapour is zero at the cavity wall. In fact the hydroxide layer on the cavity wall is found to be so thin (< 1 μm) that water molecules and sodium will react instantaneously.

(v) The effect of convection from the drop is negligible. Assuming that the heat reaches the drop purely by conduction, the Nusselt number for a drop in an infinite gas obeys the Langmuir equation

$$Nu = 2ah/k = 2$$

where h is the coefficient of heat transfer, $2a$ the diameter of the sphere, and k the thermal conductivity of the vapour. However, when there is also convection present, the Nusselt number is [4]

$$Nu = \frac{2L}{C_p \cdot \Delta T} \cdot \ln\left(1 + \frac{C_p \cdot \Delta T}{L}\right)$$

where ΔT is the temperature difference between the gas and the liquid surface. The greatest value of $C_p \Delta T/L$ is 0.292 (for a bulk sodium temperature of 726 K), and the corresponding value of Nu is 1.76. Thus convection is expected to remain small for all sodium temperatures considered here.

An equation of cavity growth can be determined once the droplet temperature T_S and the corresponding vapour pressure P_W are known. In steady state conditions the diffusion rate of water vapour in hydrogen is equal to the evaporation rate from the droplet, and for spherical symmetry (Fig. 5)

$$P_W = k(T_\infty - T_S) \cdot G T_S / 18 L D P_0. \qquad (5)$$

P_W and T_S are also related by the vapour pressure curve for water, and hence may be found.

Defining a characteristic velocity

$$u = k_0 G T_B^2 / P_0 L_0 a = 0.8375\,\mathrm{m\,s^{-1}} \tag{6}$$

from Appendix 2, in dimensionless quantities $R = r/a$, $T = ut/a$, the cavity growth equation is

$$\frac{\mathrm{d}R}{\mathrm{d}T} = \frac{[kGT_S(T_\infty - T_S)/(12L\,\mathrm{au}\,P_0)]\cdot\dfrac{R}{R-1}}{(T_S/T_\infty)\cdot(3R^2 - R - \frac{1}{2}) + (1 - \frac{1}{2}P_W)\cdot(R + \frac{1}{2})}. \tag{7}$$

This model can be tested by comparing the estimated cavity growth with observation. The dimensionless radius R for the vapour cavity surrounding an individual droplet is found during the period of film boiling, i.e. up to $t = 0.1$ s. The total volume of vapour produced is

$$V_T = V_0 \cdot (R^3 - 1). \tag{8}$$

Thus V_T varies from $6 \times 10^{-6}\,\mathrm{m}^3$ (at 480 K) to $26 \times 10^{-6}\,\mathrm{m}^3$ (at 726 K).

In practice, the film boiling stage is concluded by the collapse of a sodium mushroom of radius ~ 1 cm and height varying from 2–4 cm, i.e. of volume $(6–12) \times 10^{-6}\,\mathrm{m}^3$. These are seen to lie in the same range as the estimated volumes V_T.

5. THE LIFETIME OF WATER DROPLETS IMMERSED IN LIQUID SODIUM

The model developed so far assumes that a droplet evaporates uniformly from the centre of a spherical vapour cavity. Although this is reasonable for short periods of film boiling (0.1 s), relatively long times are required for total evaporation to occur. The droplet is expected to settle near the base of the cavity, and is supported against gravity by the rapid efflux of vapour (Fig. 6).

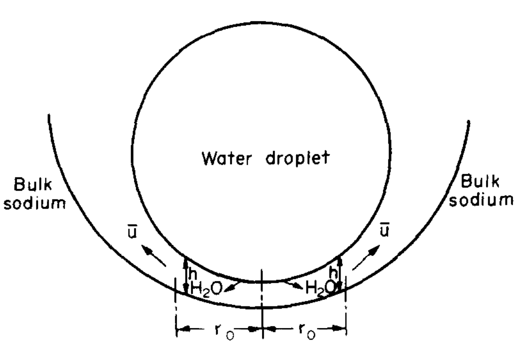

FIG. 6. The Leidenfrost effect in detail.

This phenomenon, the Leidenfrost effect, is described by Gottfried and Bell [5], although the theory requires some modification to take account of chemical reaction. Details of the process are given in Appendix 3, and the time taken for a droplet of radius a to disappear is

$$t = \tfrac{16}{5} \cdot [L\rho_W / k(T_\infty - T_S)]^{3/4} \cdot (9\mu / 8\rho_v g)^{1/4} \cdot a^{5/4}. \tag{9}$$

Using the values given in Appendix 1, corresponding to bulk sodium temperatures of 480, 594 and 726 K the time taken for a droplet of radius 1.42×10^{-4} m to disappear is 1.3, 0.9 and 0.7 s respectively.

Finally, in the later stages of the reaction when $r > a$, the ratio of evaporation rates for the symmetrical, and the Leidenfrost model is found to vary from 1.2 (at 480 K) to 1.7 (at 726 K). Thus the evaporation rate is not critical on the position of the drop within the cavity, and, in many cases, the simple, spherically symmetrical model will be adequate.

6. DISCUSSION AND CONCLUSIONS

The experimental/analytical work described in Sections 2 and 3 shows that for water entry into molten sodium, the temperature of the interface remains well above the boiling point of water at all times. The heat flux into the droplets is reduced by a vapour film, and steady film boiling is allowed to develop. By contrast, when water is added to hot oil, there is no vapour film to moderate the heat flux, and nucleate boiling with vigorous vapour growth occurs instead. In Sections 4–5, two different models of film boiling are described for a droplet in molten sodium, and differences between them are seen to be relatively unimportant.

The actual lifetime of such a droplet depends on several factors, including its size and the bulk sodium temperature, but for the fine droplets discussed in Section 5, it is of the order of a second. If a droplet of this size was injected from a leak into sodium flowing at, say $3\,\mathrm{m\,s^{-1}}$ at a temperature of 726 K, a possible situation in a secondary heat exchanger in a fast reactor, then the droplet would be transported approximately 3 m before being consumed. Thus plantwork up to this distance downstream from the original site of the leak should be regarded at risk.

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APPENDIX 1

Numerical values of component properties used in the calculations

T_∞(K)	T_S(K)	k_W (W m^{-1} K^{-1})	k_H (W m^{-1} K^{-1})	α_S (m^2 s^{-1})	D (m^2 s^{-1})	μ (N s m^{-2})
480	327.5	0.030	0.249	7.07×10^{-5}	1.54×10^{-4}	0.138×10^{-4}
594	335.7	0.034	0.272	7.37×10^{-5}	1.90×10^{-4}	0.152×10^{-4}
726	341.4	0.040	0.298	7.77×10^{-5}	2.34×10^{-4}	0.170×10^{-4}

APPENDIX 2

Derivation of n_W, n_H *and* k *for the water vapour–hydrogen mixture*

1. n_W (*Number of water moles in the cavity*). The concentration of water vapour at the hydroxide surface (r) is zero, so, assuming steady-state diffusion, at distance r' ($a < r' < r$)

$$C(r') = \left(\frac{aC_W}{r-a}\right)\left(\frac{r}{r'} - 1\right) \text{kg mol m}^{-3} \qquad \text{(A2.1)}$$

where $C_W = P_0 P_W / G T_S$ is the concentration of water vapour at $r = a$. Thus

$$n_W = 4\pi \int_a^r r'^2 C(r') \, \mathrm{d}r'$$
$$= \tfrac{2}{3}\pi a P_0 \frac{P_W}{G T_S} (r-a)(r+2a). \qquad \text{(A2.2)}$$

2. n_H (*Number of hydrogen moles in the cavity*). Let $C_1 = P_0(1-P_W)/GT_S$, and $C_2 = P_0/GT_\infty$ be the concentrations of hydrogen at (1) the droplet surface and (2) the cavity wall respectively. The concentration at a distance r' is

$$C(r') = \frac{rC_2 - aC_1}{r-a} + \frac{ar}{r-a}(C_1 - C_2)\frac{1}{r'}$$

and similarly to equation (A2.2)

$$n_H = \frac{4\pi}{3}(r-a)\frac{P_0}{GT_S}$$
$$\times \left(\frac{T_S}{T_\infty}\left[r^2 + \tfrac{1}{2}ar\right] + \left[1 - P_W\right]\left[a^2 + \tfrac{1}{2}ar\right]\right). \qquad \text{(A2.3)}$$

3. k. An approximate expression for the conductivity of the water vapour–hydrogen mixture is

$$k = (n_W k_W + n_H k_H)/(n_W + n_H) \qquad \text{(A2.4)}$$

so

$$k = k_H - f(k_H - k_W)$$

where

$$f = P_W \Bigg/ \left[1 + \frac{T_S(r^2 + \frac{1}{2}ar)}{T_\infty(a^2 + \frac{1}{2}ar)}\right]. \qquad \text{(A2.5)}$$

The cavity growth equation

If $W = 4\pi kra(T_\infty - T_S)/18L(r-a)$ (kg mol m^{-2} s^{-1}) is the evaporation rate from the droplet, the rate of absorption of water vapour in the cavity wall is $W - \mathrm{d}n_W/\mathrm{d}t$. The increase in hydrogen content of the cavity will be

$$\frac{\mathrm{d}n_H}{\mathrm{d}t} = \frac{1}{2}\left(W - \frac{\mathrm{d}n_W}{\mathrm{d}t}\right).$$

The cavity growth equation (7) is obtained by substitution from the above.

APPENDIX 3

The Leidenfrost effect and chemical reaction

The theory set out by Gottfried and Bell [5] requires modification when chemical reaction is present. Let $\bar{u}$ (m s^{-1}) be the average velocity of a gas out from the gap h (Fig. 6). The average concentration of hydrogen in the vapour is (kg mol m^{-3})

$$C_H = \frac{P_0}{2}\left(\frac{1}{GT_S} + \frac{1}{GT_\infty} - \frac{P_W}{GT_S}\right). \qquad \text{(A3.1)}$$

If the water evaporation rate from the base of the drop is W (kg m^{-2} s^{-1}), the flux of hydrogen from the sodium surface is $\frac{1}{2} \cdot (W/18)$ kg mol m^{-2} s^{-1}. The continuity equation for the flow field beneath the drop becomes

$$C_H \cdot \frac{\partial}{\partial r_0}(\bar{u} r_0) = \frac{W}{36} \cdot \frac{r_0}{h}. \qquad \text{(A3.2)}$$

On replacing equation (2) in the original analysis [5] by equation (A3.2), the final result is

$$m_0 g = 3\pi W \mu r_0^4 / 2\rho_v h^3 \qquad \text{(A3.3)}$$

where m_0 is the droplet mass, μ the viscosity of the vapour, r_0 the radius of contact, and

$$\rho_v = 18P_0\left(\frac{1}{GT_S} + \frac{1}{GT_\infty} - \frac{P_W}{GT_S}\right). \qquad \text{(A3.4)}$$

Provided the droplet diameter is less than $2(\sigma/\rho_W g)^{1/2}$, i.e. 5.4 mm, surface tension forces dominate gravity and the drop remains spherical. In this case the radius of contact (r_0) is approximately equal to the radius of the droplet. Thus the droplet mass is $m_0 = \frac{4}{3}\pi\rho_W r_0^3$, and the mass loss from evaporation is $W = k(T_\infty - T_S)/Lh$, so

$$h = K r_0^{1/4}$$

where

$$K = \left[9k(T_\infty - T_S)\mu/(8L\rho_v \rho_W g)\right]^{1/4}. \qquad \text{(A3.5)}$$

The rate of mass loss from the droplet is

$$4\pi r_0^2 \rho_W \cdot \frac{\mathrm{d}r_0}{\mathrm{d}t} = -\pi r_0^2 W \qquad \text{(A3.6)}$$

so the droplet radius as a function of time is determined from

$$r_0^{1/4} \frac{\mathrm{d}r_0}{\mathrm{d}t} = -k \cdot (T_\infty - T_S)/4L\rho_W. \qquad \text{(A3.7)}$$

Thus the time taken for the droplet of initial radius a to disappear is

$$t = \tfrac{16}{5} \cdot L\rho_W / k(T_\infty - T_S)^{3/4} \cdot (9\mu/8\rho_v g)^{1/4} \cdot a^{5/4}. \qquad \text{(A3.8)}$$

LA REACTION ENTRE EAU ET SODIUM

Résumé—En utilisant un film cinéma à haute vitesse (5000 vues par s), on suit le comportement des quantités mesurées (0,02 cm^3) d'eau froide (295 K) injectées dans du sodium chaud (480 K à 730 K). A partir des résultats, on établit un modèle du système en réaction basé sur les mécanismes de l'ébullition en film, transitoire et stationnaire. Des équations permettent d'estimer les températures, les vitesses de réaction, les durées de vie des gouttes d'eau dans le sodium chaud. On discute l'application des résultats aux circuits échangeurs de chaleur des réacteurs nucléaires à sodium.

DIE REAKTION VON WASSER MIT NATRIUM

Zusammenfassung—Das Verhalten bestimmter Mengen (0,02 ml) kalten Wassers (295 K) beim Einspritzen in heißes Natrium (480 K bis 730 K) wurde mit Hilfe einer Hochgeschwindigkeitskamera (5000 Bilder/s) verfolgt. Aus den Ergebnissen wurde ein Modell des Reaktionsverfahrens erstellt, das auf instationärem und stetigem Filmsieden beruht. Gleichungen wurden aufgestellt, die es ermöglichen, die Temperaturen, Reaktionsgeschwindigkeiten und Lebenszeiten von Wassertropfen in heißem Natrium zu bestimmen. Die Anwendung der Ergebnisse auf natriumgefüllte Wärmeaustauscherkreisläufe in Kernreaktoren wird diskutiert.

РЕАКЦИЯ МЕЖДУ ВОДОЙ И НАТРИЕМ

Аннотация — Поведение измеренных объемов (0,02 мл) холодной воды (295 К) после их впрыскивание в горячий натрий (от 480 К до 730 К) изучалось с помощью высокоскоростной кинокамеры (5000 кадров/сек$^{-1}$). По полученным результатам построена модель реагирующей системы, основанная на нестационарных и стационарных процессах пленочного кипения, и выведены уравнения, позволяющие определить температуру, скорость реакции и время жизни капель воды в горячем натрии. Обсуждается применение результатов к контурам натриевых теплообменников ядерных реакторов.