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THE ROLE OF NUCLEATION IN THE INITIAL PHASES OF A RAPID DEPRESSURIZATION OF A SUBCOOLED LIQUID

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Abstract—A two-fluid model with zero slip between the phases but which allows for unequal phase temperatures, changing interfacial area and the effect of heterogeneous nucleation has been used to evaluate the initial depressurization of a subcooled or saturated light. The results compare favourably with existing water and dichlorodifluoromethane experiments and suggest that the algorithm, in association with well-tabulated data, could be used to evaluate the heterogeneous nucleation factor.

Key Words: blowdown, nucleation, subcooled liquid

1. INTRODUCTION

The depressurization of a vessel containing saturated or subcooled liquid may occur in a variety of industrial processes and often poses a potentially hazardous situation. It is a well-documented occurrence with many detailed observations available in the literature (e.g. Edwards & O'Brien 1970; Alamgir & Lienhard 1981; Winters 1979). Attempts to model the fluid mechanics have met with varying success but it is clear from the work of Ferch (1979) and Hancox *et al.* (1975), among others, that a homogeneous equilibrium model is incapable of predicting the earliest phase of a rapid depressurization. The minimum requirement is for a two-fluid model which allows for thermal non-equilibrium. Edwards & O'Brien (1970), Wolfert (1976) and Ferch (1979) have demonstrated that such models usually have to build in some empiricism in order to obtain a match with experimental data. In none of the studies is the effect of nucleation taken into account.

The problem of establishing averaged conservation equations for two-phase flows has received a considerable amount of attention (i.e. Vernier & Delhaye 1968; Ishii 1975; Bouré 1979). The major difficulty in modelling the equations arises through the complex geometry of the interfaces and their subsequent motions, the differing flow patterns depending on the geometrical boundaries of the flow field and the time scale of the flow. It is of course, the averaging in time and space of such complex interactions that gives rise to a loss of information and the introduction of source terms (Delhaye 1974) in the averaged equations which then have to be modelled.

The source terms provide the link between the two phases at the interfacial boundaries and largely determine the success of predicting the behaviour of the motion of the two-phase fluid. For flows involving significant phase changes it is important to correctly represent the heat transfer from the liquid to the vapour and the corresponding mass transfer due to evaporation, flashing or diffusion. When slip between the phases becomes significant correct modelling of the interfacial forces of drag, virtual mass, lift etc. arising from the transient non-uniform pressure distribution along the interfacial boundaries becomes important (Drew 1983; Banerjee & Hancox 1978). As indicated by Kocamustafaogullari & Ishii (1983), the source terms also involve the interfacial area concentration which is not only dependent on the phase change across existing interfacial boundaries but also on the rate at which new interfaces are created by nucleation or destroyed by coalescence.

The present study is primarily concerned with modelling the earliest phenomena involved with depressurization. As such slip between the phases is likely to be small, the effects of thermal non-equilibrium are significant. The role of nucleation and its effect on the interfacial area, and hence heat and mass transfer, is assessed and compared with available experimental data.

2. AVERAGED CONSERVATION EQUATIONS

Averaged equations governing the motion of a one-dimensional two-phase flow, with no relative velocity are:

$$\frac{\partial}{\partial t}\epsilon_k \rho_k + \frac{1}{A}\frac{\partial}{\partial z}A\epsilon_k \rho_k u = \dot{m}_{ik} + \dot{m}_{NU}, \qquad [1]$$

$$\frac{\partial}{\partial t}\epsilon_k \rho_k u + \frac{1}{A}\frac{\partial}{\partial z}A\epsilon_k \rho_k u^2 + \epsilon_k\frac{\partial p}{\partial z} = \dot{m}_{ik}u - \tau_{ik} - \tau_{wk} - \epsilon_k \rho_k g\frac{\partial Z}{\partial z} + \dot{m}_{NU}u \qquad [2]$$

and

$$\frac{\partial}{\partial t}\epsilon_k \rho_k\left(h_k + \frac{u^2}{2}\right) + \frac{1}{A}\frac{\partial}{\partial z}A\rho_k\epsilon_k u\left(h_k + \frac{u^2}{2}\right) - \epsilon_k\frac{\partial p}{\partial t} = \dot{q}_{ik} + \dot{q}_{wk} + \tau_{ik}u + \dot{m}_{ik}\left(h_k + \frac{u^2}{2}\right)_i - \epsilon_k \rho_k u g\frac{\partial Z}{\partial z} + \mathrm{BCE}. \qquad [3]$$

The subscript k (= L or G) denotes the phase (liquid or vapour). The flow quantities ϵ_k, ρ_k and h_k are the volume void fraction, density and enthalpy of phase k, respectively; u and p are the axial velocity and pressure of the mixture; $\dot{m}$ and $\dot{q}$ are the rates of mass and heat transfer per unit volume; and τ is the shear force per unit volume. The subscripts w, i and NU denote transfer from the wall, the interface and nucleation, respectively. BCE is the energy per unit volume transferred to the vapour when the liquid flashes (Fisher 1948; Frenkel 1955). A is the cross-sectional area, Z is the elevation of the gravitational acceleration and t and z are the time and space independent variables, respectively.

Given the thermal and caloric equations of state, [1]–[3] may be transformed into their characteristic form and solved numerically. The boundary conditions for the present problem require that the pressure at the open end of the vessel be equal to the surrounding atmospheric pressure or to be choked. At the closed end of the vessel the velocity is zero.

2.1. Interfacial heat transfer

The missing information, in the averaged equations of the equal velocity but unequal temperature (EVUT) approximation, is associated with the shear forces, the interfacial heat and mass transfers. The EVUT approximation is the simplest way of providing a description of the sudden depressurization of an initially subcooled or saturated liquid contained in a vessel. The usual assumptions (Winters 1979) in the modelling of the interfacial terms are that the two-phase mixture consists of a uniform dispersion of bubbles which are spherical and independent from one another. In addition, the slip between the phases is taken as zero. As shown by Ishii *et al.* (1982), the mass transfer is $\dot{m}_i = \dot{q}_i/h_{LG}$, where h_{LG} is the latent heat, and the heat transfer is $\dot{q}_i = \alpha_i F_D$, where α_i is the interfacial area concentration and F_D is a driving force.

Most investigators assume that

$$\dot{q}_i = \frac{A_i}{V}h_i\,\Delta T, \qquad [4]$$

where A_i is the averaged interfacial area, h_i is the heat coefficient, V is the volume of the two-phase mixture and ΔT is the difference between the interfacial temperature T_i and the phase temperature T_k. For a mixture consisting of spherical bubbles:

$$A_i = N_b 4\pi r_b^2, \qquad [5]$$

where N_b is the number density of the bubbles and r_b is the radius at the bubble. By definition the void fraction ϵ is given as

$$\epsilon = N_b \tfrac{4}{3}\pi r_b^3 \qquad [6]$$

and hence,

$$\dot{q}_i = 3\frac{\epsilon}{r_b}h_i\,\Delta T. \qquad [7]$$

The heat transfer coefficient is a function of the Nusselt number Nu, the thermal conductivity k_L and the bubble radius r_b. For a stationary bubble,

$$h_i = \frac{\mathrm{Nu}}{2r_b} k_L. \tag{8}$$

Nu is usually obtained through experimental correlations and for the present study the following value for a single sphere (Bird 1960) is used:

$$\mathrm{Nu} = 2 + 0.6 \frac{(2r_b U_s \rho_L)^{1/2}}{\mu_L} \frac{(C_p \mu_L)^{1/3}}{k_L},$$

where U_s is the relative velocity between the phases and μ_L and C_p are the dynamic viscosity and specific heat (constant pressure) for the liquid phase.

For zero slip,

$$\dot{q}_i = \frac{3}{2r_b^2} \epsilon \mathrm{Nu} k_L \Delta T. \tag{9}$$

It is also assumed that $T_G = T_i$. Similar forms of [9] have been used previously by Ferch (1979), among others, and invariably involve an undetermined constant which is chosen to provide a close fit to the available data.

2.2. *Number density production*

Ishii *et al.* (1982) and Kocamustafaogullari & Ishii (1983) postulated that the number of bubbles in a given control volume can be conserved. Thus, making use of the transport theorem, it follows that

$$\frac{\mathrm{d}n_b}{\mathrm{d}t} = \int_V \frac{\partial N_b}{\partial t} \mathrm{d}v + \int_S N_b \nabla \cdot \mathbf{u} \, \mathrm{d}s,$$

where N_b is the number density of the bubbles, n_b is the total number of bubbles and **u** is the velocity field. It therefore follows that for an infinitesimal control volume

$$\frac{\partial N_b}{\partial t} + \mathrm{div}\, N_b \mathbf{u} = \text{total change of } n_b.$$

The change in N_b arises from homogeneous nucleation in the bulk of the liquid (H_{HOM}), heterogeneous nucleation on the surfaces (H_{HET}) and the rate at which bubbles coalesce (H_{coal}). A term involving bubble growth due to the disintegration of larger bubbles could also be included but it is thought to be negligible for the size of the bubbles likely to be encountered in the initial phases of an expansion. The number density for one-dimensional flow is then given by

$$\frac{\partial N_b}{\partial t} + u \frac{\partial N_b}{\partial z} + N_b \frac{\partial u}{\partial z} = H_{\mathrm{HOM}} + H_{\mathrm{HET}} - H_{\mathrm{coal}}. \tag{10}$$

Utilizing [1] and [3] together with the state equation $\rho_G = f(p, h_g)$, allows [10] to be written as

$$\frac{\mathrm{D}N_b}{\mathrm{D}t} - \frac{N_b}{\rho_G C_G^2} \frac{\mathrm{D}p}{\mathrm{D}t} - \frac{N_b}{\epsilon} \frac{\mathrm{D}\epsilon}{\mathrm{D}t} = H_{\mathrm{HOM}} + H_{\mathrm{HET}} - H_{\mathrm{coal}} - \frac{N_b}{\rho_G \epsilon} (\dot{m}_{iG} + \dot{m}_{NU}) + \frac{N_b}{\rho_G^2 \epsilon} \mathrm{BCE} \left[\frac{\partial \rho_G}{\partial h_G} \right]_p, \tag{11}$$

which is in a form compatible with a solution by the method of characteristics. C_G is the speed of sound in the vapour phase and D/Dt is the directional derivative following the fluid particle path.

Equation [11] requires that the nucleation rate be given. The theory of nucleation (Kagan 1960; Skripov 1974; Doring 1937; Fisher 1948; Volmer & Weber 1926) states that the rate of bubble formation is proportional to $\exp(-\mathrm{ME}/K_B T_L)$, where ME is the minimum molecular energy needed for the critical cluster to survive and is equal to $(4/3)\pi r_*^2 \sigma$. According to nucleation theory this energy is directed towards (a) the formation of voids of critical size and (b) the evaporation from the interface and filling of the void with vapour molecules. K_B, T_L and σ are the Boltzmann constant, the liquid temperature and the surface tension, respectively. r_* is the radius of the critical cluster, which is defined as the smallest bubble which can survive in the metastable liquid during the irreversible process of phase transition. The proportionality factor depends on the rate per unit area at which a bubble surface gains or loses molecules. Assuming that the critical bubble is in mechanical and chemical equilibrium with the surrounding liquid, the critical size is given by

$$r_{*} = \frac{2\sigma}{\Delta P}, \qquad [12]$$

where ΔP is the pressure difference between the saturated vapour and the superheated liquid. The rate of critical bubble formation is then given by

$$H_{\mathrm{HOM}} = N\left(\frac{2\sigma N}{\pi \cdot \mathrm{MW} \cdot B}\right)^{1/2} \exp\left(-\frac{16\pi\sigma^3}{3K_{\mathrm{B}} \cdot T_{\mathrm{L}} \cdot \Delta P^2}\right), \qquad [13]$$

where N is the number of molecules per unit volume, MW is the molecular weight and B is a coefficient in the equation of the work needed for bubble formation which takes into account chemical or mechanical equilibrium. In this study $B = 2/3$ and N_{A} is the Avogadro number. Using [12] and [13] it can be shown that homogeneous nucleation is only possible when the degree of superheat is in excess of a few hundred degrees. In most experimental studies the temperature difference is less than the superheat for homogeneous nucleation. This indicates that bubble generation does not occur in the bulk of the liquid but mainly on the walls of the container or on impurities in the liquid. Therefore, H_{HOM} can be considered negligibly small. At the beginning of a depressurization H_{coal} might also be considered small.

As shown by Blander & Katz (1975), the heterogeneous nucleation rate can be written as

$$H_{\mathrm{HET}} = N^{2/3}\frac{(1-\mathbf{m})}{2}\left(\frac{2 \cdot \sigma N_{\mathrm{A}}}{\pi \cdot \mathrm{MW} \cdot B \cdot \phi}\right)^{1/2} \exp\left(\frac{-\mathrm{ME} \cdot \phi}{K_{\mathrm{B}} T_{\mathrm{L}}}\right). \qquad [14]$$

It may be noted that the surface tension is generally a function of the temperature and of the form $\sigma = \sigma_{\circ}(1 - T/T_{\mathrm{c}})^n$, where the subscript $_{\circ}$ is a reference condition; ϕ is a heterogeneous factor, <1, which physically implies that the molecular clusters formed on a rough surface or suspended particles need less energy to survive. Bubbles formed in this manner assume a shape bounded by a plane and a portion of spherical surface. Applying Young's equation at the edges of the bubble yields $\mathbf{m} = (\sigma_{\mathrm{SL}} - \sigma_{\mathrm{SG}})/\sigma_{\mathrm{LG}}$, where σ_{SL}, σ_{SG} and σ_{LG} are the surface tensions between liquid and solid, vapour and solid, and vapour and liquid, respectively. It is readily shown that $\mathbf{m}$ is equal to $-\cos\theta$, where θ is the contact angle at the bubble surface. Blander & Katz (1975) show that ϕ and $\mathbf{m}$ are linked by

$$\phi = \frac{(2 - 3\mathbf{m} + \mathbf{m}^3)}{4}. \qquad [15]$$

Equation [14] in conjunction with [11] allows the number density of the bubbles to be evaluated at all points in the flow field. From [6] and [7] the interfacial heat transfer can be written in terms of the number density and the void fraction as

$$\dot{q}_{\mathrm{i}} = 3.9\, N_{\mathrm{b}}^{2/3}\epsilon^{1/3}\mathrm{Nu}k_{\mathrm{L}}\,\Delta T. \qquad [16]$$

As one can observe there are no free parameters attached to the above equation (unlike [9], where r_{b} has to be specified *a priori*). N_{b}, ϵ and ΔT are readily evaluated by the equations of motion. Thus, the restriction of specifying a constant value of the bubble size or the number density used in many models is now removed.

3. DISCUSSION

Although it has been recognized that thermal non-equilibrium has to be allowed for, most previous studies (Edwards & O'Brien 1970; Hancox *et al.* 1975; Banerjee & Hancox 1978; Winters 1979; Ferch 1979) have only obtained agreement with experimental observations by assigning an arbitrary constant value to r_{b} or N_{b} in [9]. Edwards & O'Brien (1970) introduced a constant number density N_{b} linked to the interfacial heat transfer and showed that a value of $N_{\mathrm{b}} = 10^9$ bubbles per unit volume yielded a solution close to experimental observations. Winters (1979) and Wolfert (1976) used a similar formulation for the heat transfer and by judicious choice of the constants secured agreement with experiments.

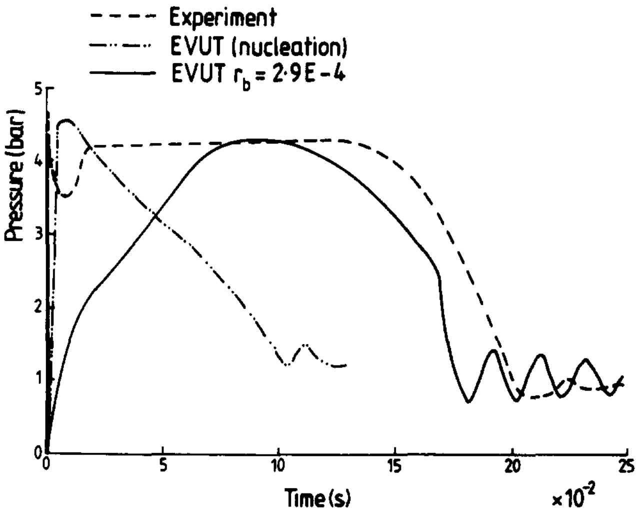

Figure 1. Long-term pressure history [comparison with the Freon 12 experiments of Winters (1979)].

A similar calculation based on [9] with $r_b = 2.9\ 10^{-4}$ m is shown in figures 1 and 2 and compared with the experimental data of Winters (1979). Although a reasonable fit to the long-term behaviour of the pressure transient is observed (figure 1), this is not true of the minimum pressure and the rate of pressure recovery (figure 2). The effect of changing the initial bubble size is shown in figure 2, where a decrease in bubble radius tends to increase the rate of pressure recovery and the maximum pressure. This variation with r_b is slightly different to that observed by Ferch (1979) and is due to the different exit boundary condition he imposed, which had the effect of increasing the mass hold up in the vessel.

To improve the model for heat transfer it is essential that the effective bubble radius is allowed to vary in time and space. From section 2 this implies that nucleation has to be allowed for. To illustrate the role of nucleation the EVUT models are used to simulate the experiments of Winters (1979), which used a 0.64 m cylindrical pressure vessel containing Freon 12. The advantage of using a small vessel is that all nucleation and heat transfer effects can be assumed to be independent of the slip between the phases.

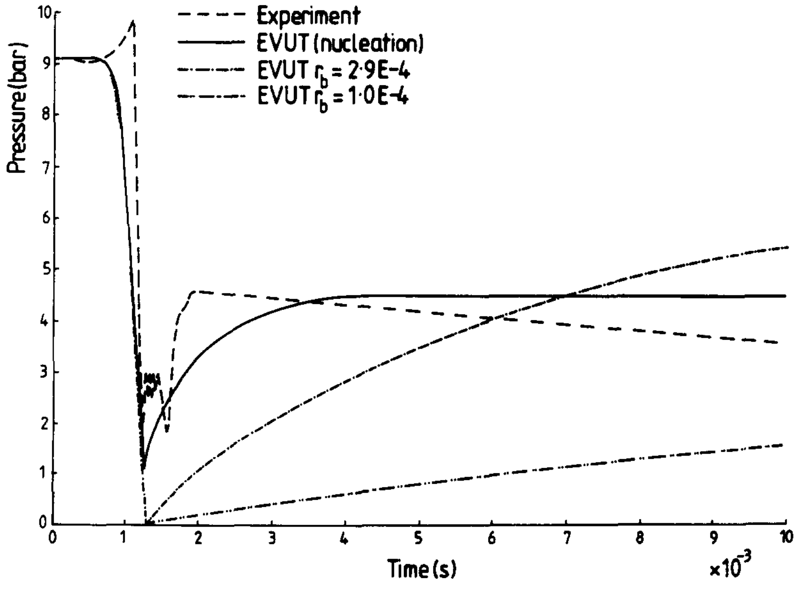

Figure 2. Initial depressurization.

The nucleation model presumes N_b is initially zero and it is therefore free of the problems associated with previous models. However, a new parameter ϕ is introduced which is a function of the contact angle θ between the tangent to the interface and the wall at the vessel or any impurities in the bulk of the liquid.

Because of the inherent difficulties in prescribing a value of ϕ, the nucleation model relies on careful selection of ϕ to agree with the experiment; ϕ is chosen so that the minimum pressure is predicted. From figure 2 it will then be noted that once this value is fixed the initial rate of depressurization and the rate of recovery agrees quite well with experiment. However, in the longer term (figure 1), the model seriously under-predicts the pressure, which suggests nucleation is no longer the main sustaining mechanism of the pressure inside the vessel.

During a fast depressurization the initiation of boiling, for a given set of initial conditions, does not take place at any particular pressure but can occur over a whole range of pressures. The vapour production, however, depends on the mechanics of nucleation (homogeneous or heterogeneous) and on the degree of superheat of the system. Using [12] and assuming that the liquid is incompressible and that the depressurization rate is fast, the temperature of the liquid remains almost constant as does the saturation temperature of the vapour and the surface tension. The reduction of the liquid pressure will give a whole range of critical sizes, i.e. critical bubble areas (see figure 3) corresponding to a given superheat. For homogeneous nucleation figure 3 shows that the surfaces of the critical bubbles for low superheats are quite big and since the critical work is proportional to the bubbles' area, this means that large amounts of energy have to be directed towards the point of flashing by the temperature and pressure fluctuations, which in turn mean almost zero probability of bubble production. There is a theoretical limit the metastable liquid can reach before it nucleates and this is the border between the metastable liquid region and the unstable two-phase region; the border is known as the spinodal line. In reality, homogeneous nucleation starts before that point if there are no artificial nucleation sites to provoke heterogeneous nucleation. For heterogeneous nucleation figure 3 shows that the critical bubble is smaller. Assuming that a critical bubble is a small segment of a sphere, with radius r, and θ is the angle between the tangent to the interface and the wall, the surface area of the segment is

$$S = 4\pi r^2 \left(\frac{2 + 3\cos\theta - \cos^3\theta}{4} \right).$$

With the aid of [15] this shows that $S = 4\pi r^2 \phi$.

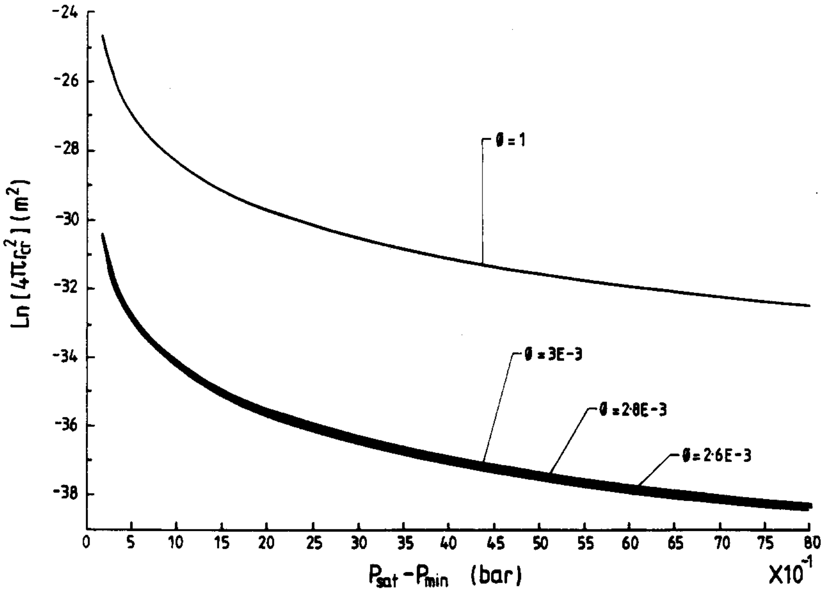

Figure 3. Dependence of the liquid superheat on the critical surface area of the vapour bubble in equilibrium.

From the definition of ϕ is it apparent that the "heterogeneous factor" gives an indication of how well the critical cluster is protected by the wall. For example, if the wall is plane and $\theta = 90°$, that will mean that only half of the bubble is exposed to the flow and for $\theta = 90°$ ϕ can be calculated to be 0.5 (50%). When 100% of the bubble rests on the wall $\phi = 1$ and θ can be calculated to be 0°. In the present study ϕ has an averaged value, i.e. an averaged value for θ which corresponds to the majority at the nucleation sites which when they nucleate the vapour production will be enough to release the liquid's superheat. From the above it is apparent that the presence of the wall or any impurity in the liquid means less interface area (between the critical bubble and the liquid), which means less critical work and therefore a higher probability for nucleation.

In terms of the critical homogeneous bubble size as a function of pressure, theory predicts negative pressures where $\partial p/\partial u = 0$; the spinodal line giving the theoretical nucleation point. The experiments, on the other hand, show a positive pressure. Theory can only account for this if the function of critical surface with the degree of superheat is multiplied by a factor ϕ which physically accounts for the role of the wall and impurity surfaces which can protect the nuclei. ϕ does not account for all effects, e.g. Kenning & Thirunaruktarasu (1970) showed that the phenomenon of nucleation is not necessarily reproduced when tests with the same surface and initial conditions are carried out, and it was suggested that the rewetting rate of a surface can affect the number of available nucleation sites. These non-quantifiable effects make ϕ difficult to predict.

Figure 2 demonstrates that the nucleation model follows more closely the experimental pressure history than the model which uses a simple heat transfer rate. This arises because in the first few milliseconds the system is in thermal non-equilibrium and it is in this period when most nucleation occurs. The simpler model always emphasizes the pressure undershoot and can be in error by up to 50%. Also, the rate of recovery of pressure is much slower. Some indication of the sensitivity of the solution to ϕ is given in figure 4. When ϕ is large (i.e. low vapour production), a lower pressure minimum can be observed together with smaller pressure recovery. When ϕ is small (high vapour production), the excess vapour produces the opposite effect.

Further comparisons of the model can be made with the experiments of Edwards & O'Brien (1970), which have often been used as benchmark data. In figure 5 (curve b) a comparison is made between experiments, with initial conditions of 70 bar and 515 K, and the EVUT nucleation model. By fitting the minimum pressure good agreement is obtained. In contrast to other reported models, this agreement has been obtained without any special manipulation of the exit boundary conditions or empirical constants to determine the interfacial heat transfer.

Also shown in figure 5 (curve a) is the effect of keeping the surface tension constant for simulating the water experiments. As can be seen, the rate of depressurization is too slow and clearly indicates

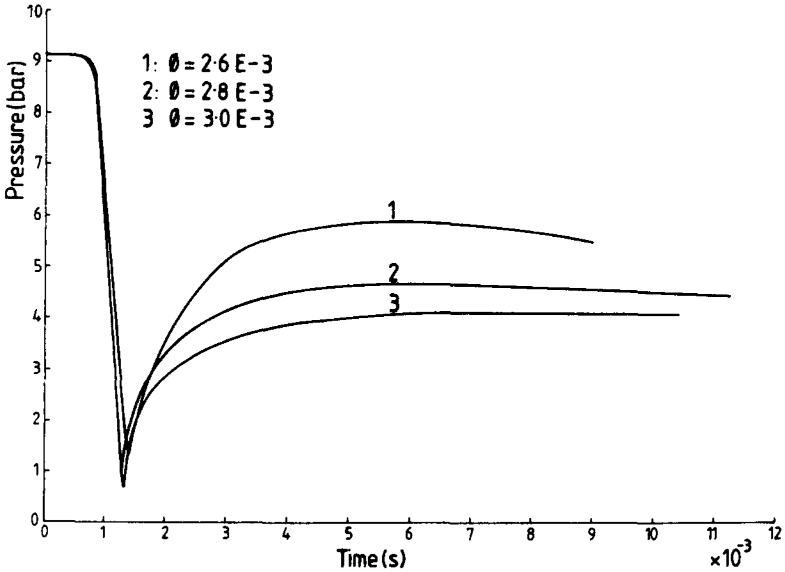

Figure 4. The effect of the heterogeneous factor ϕ on the initial pressure history.

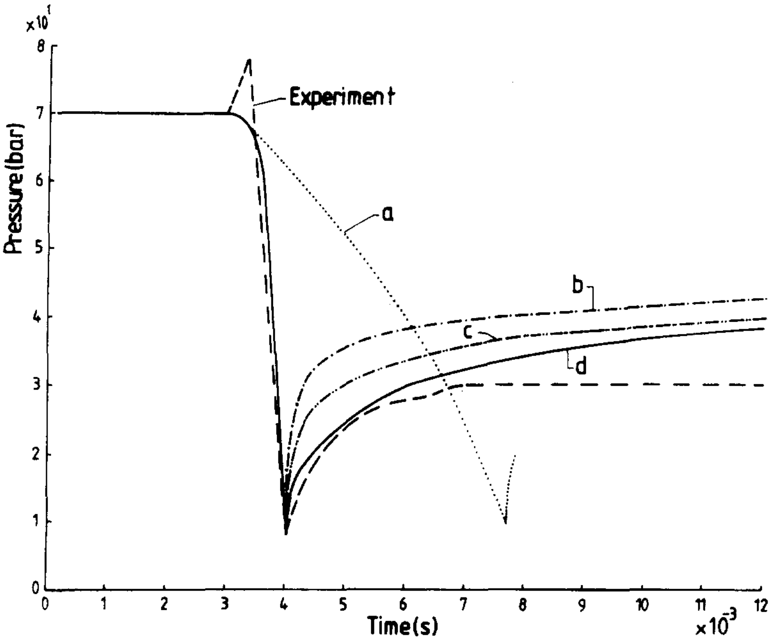

Figure 5. Comparison of the EVUT nucleation model with Edwards & O'Brien's (1970) water experiments.

that the large temperature variation arising in the water experiments has to be taken into account when evaluating the surface tension. As noted earlier, the temperature variation in the Freon case is sufficiently small for the surface tension to be considered constant. The theory predicts the metastable region but tends to over-predict the pressure maximum. This may arise because the nucleation theory provides an expression for the steady-state rate of nucleation based on the maximum free energy barrier for the given conditions of the metastable liquid surrounding the nucleus, and by assuming that these conditions are unaffected by the other surrounding nuclei. Near the pressure minimum, where the rate of nucleation is high, the growth or collapse of a newly formed bubble will strongly depend on other small bubbles and the large bubbles which are growing rapidly through heat and mass transfer through the interface. As the liquid superheat is rapidly being released into the vapour phase the conditions surrounding the nucleus change and affect the two major modes of spontaneous growth, i.e. through evaporation and condensation. At that stage, even a single molecule coming out of or going into a bubble could result in it collapsing or growing. This crowded effect will not only change the properties of the interface but the whole mechanical and chemical equilibrium between the two phases. Compression waves arising through bubble growth, slip and bubble rotation will affect the pressure difference between the phases and change it from $2\sigma/r$. Enhanced surface tension and pressure differences between the phases for the critical bubble size will increase the free energy barrier and therefore more energy will be required from the metastable liquid to nucleate. This in turn will reduce the nucleation rate.

As a simple approximation to assess the crowded effect, curve c in figure 5 is obtained by assuming $\sigma = \sigma(Ts)$ up to the minimum pressure and thereafter, when the effect of the surrounding bubbles becomes increasingly important, the surface tension is assumed constant and equal to the value at the point of minimum pressure. Curve d is similar to curve b in figure 5, but with the surface tension after the point of minimum pressure increased by 25%. This increase in σ reduces nucleation and provides better agreement in the early phases of the expansion. Further progress requires a model to explain the crowded effect on the nucleation rate.

In comparison with the Freon 12 data of Winters (1979) (figure 1) the results show better agreement. There are two factors which might be responsible for this. One is the degree of penetration of the system into the metastable region, which would change the nucleation rate and change the rate at which the metastable liquid returns to equilibrium. Using van der Waals equation it can be shown that in the water experiments the system is closer to the spinodal line than in the

Freon 12 experiments. The other relates to the number of molecules in the bulk of the liquid. Skripov (1974) showed that the number of molecules affects the frequency of the fluctuation formation of nuclei. It may be noted that water is richer in molecules than Freon 12.

The fact that the model describes quite accurately the variation of pressure up to, and beyond, the point of minimum pressure suggests that an averaged value of ϕ could be unambiguously determined from experiments similar to those reported by Alamgir *et al.* (1980).

4. CONCLUSIONS

A satisfactory description of the early phases of a rapid depressurization of a subcooled or saturated liquid requires the effect of nucleation to be accounted for and allowance to be made for the time variation of the interfacial area concentration. Whilst the reported model is free from many of the defects present in earlier studies, the agreement with experiments still depends on careful choice of one parameter. This parameter is intimately related to the effect of heterogeneous nucleation, which in the most ideal conditions is difficult to specify *a priori.*

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