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Growth of vapour bubbles in boiling polymer solutions—II. Nucleate boiling heat transfer

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Abstract—The effect of polymer additives on the growth of vapour bubbles in an infinite volume of superheated fluid was analysed in part 1 of the present paper. The second part discusses the reasons for the experimentally observed qualitatively different (with respect to a pure solvent) changes in the heat transfer coefficient in bubble boiling of polymer solutions with different macromolecule content. An explanation is suggested for the phenomenon of heat transfer enhancement in boiling of diluted solutions of high polymers with a concentration of about 0.01%.

INTRODUCTION

Until now, the problem of the effect of polymer additives on boiling heat transfer [1] has been studied only experimentally. The data accumulated reflect the diversity of physical mechanisms manifesting themselves in the processes investigated, the relative role of which, even for the same kind of polymers of the same molecular mass, can substantially change with concentration, temperature, external conditions, etc. This was convincingly confirmed by the experimentally discovered effect of heat transfer enhancement in the bubble mode of boiling for diluted solutions ($c \approx 15$–500 ppm) and the inverse effect, i.e. a decrease in boiling in heat transfer of weakly concentrated systems ($c \approx 1\%$). A qualitatively different character of the effect of highly molecular compounds on heat transfer in various concentrational ranges has not found theoretical justification as yet. The connection of macroscopic effects with the experimentally found changes in the internal characteristics of boiling (shape and size of bubbles, frequency of nucleation, etc.) is not called into question. However, the problem of the reasons for such changes needs additional investigation. In view of this, it seems worthwhile to carry out a more detailed discussion of experimental data on the basis of the results of theoretical analysis [1, 2].

EXPERIMENTAL RESULTS

The role of small additives of water-soluble polymers in boiling appears to have been investigated for the first time in ref. [3]. Using a plane heating element, the authors of that work discovered a substantial growth in a heat flux under the conditions of a fixed superheat $\Delta T = 10$–35 K on the introduction into water of PAA additives Separan NP 10 ($M = 10^6$), NP 20 ($M = 2\times10^6$) and HEC ($M \approx 7\times10^4$–10^5) with concentrations from 62.5 to 500 ppm. The dynamic viscosity of the solution did not exceed 3.57×10^{-3} Pa·s and the experiments were conducted at atmospheric pressure. Visual observations showed that in a boiling solution the separation diameter of bubbles decreases, their size uniformity increases and the tendency to coalescence becomes smaller. It is noted that additions of HEC lead to a more rapid coating of the heating surface with bubbling in the beginning of boiling, with the size of bubbles being smaller than in a PAA solution and water.

In ref. [4], on a setup similar to that described in ref. [3], a nonmonotonous change was discovered in the heat transfer coefficient α with an increase in the concentration of PIB Vistanex L80 ($M = 7.2\times10^5$) and L100 ($M = 1.4\times10^6$) in boiling cyclohexane. When $22 \leqslant c \leqslant 300$ ppm, they observed a growth in α and, for $300 \lesssim c \leqslant 5150$, its decrease. The $\alpha_{\max}$ value corresponded to insignificant changes in the viscosity of the fluid. Within the entire range of concentrations at supercritical (for a pure solvent) superheats, a film mode of boiling did not set in up to the maximum attainable value $\Delta T \approx 60$ K. The growth of polymer concentration in the region of 'delayed' bubble boiling led to a considerable decrease in heat transfer.

The results of refs. [3, 4] were confirmed in ref. [5] during investigations into bubble boiling of the aqueous solutions of the HEC Natrosol 250 HR ($M = 2\times10^5$), 250 GR ($M = 7\times10^4$) and PEO [$M = \sim(2$–$4)\times10^6$] in a tube with forced convection.

NOMENCLATURE

M	molecular mass
c	mass concentration of polymer
k_0	equilibrium mass concentration of solvent
q	heat flux
p	pressure
$\Delta\hat{T}^*$	conditional limiting superheat introduced in [1]
R	bubble radius
a_2	thermal diffusivity of liquid
t	time
Ja	Jacob number, $c_2\Delta T\rho_2(\rho_1 l)^{-1}$
c_2	specific heat of liquid
l	specific heat of evaporation
Pr	Prandtl number, ν/a_2
We	Weissenberg number, $\lambda\dot{\gamma}$.

Greek symbols

α	heat transfer coefficient
η	Newtonian viscosity of liquid
σ	surface tension coefficient
ρ	density
ν	kinematic viscosity
λ	time of relaxation
$\dot{\gamma}$	shear rate in microlayer.

Subscripts

s	solvent
p	solution
1	vapour
2	liquid
PEO	polyethylene oxide
PAA	polyacrylamide
HEC	hydroxyethylcellulose
PIB	polyisobutylene.

A decrease in the size of bubbles in a solution and decrease in their coalescence were noted. Similar observations were described in refs. [6, 7].

The conclusions made in the above-mentioned works agree with the data of works [8–11]. In ref. [8] an improvement in heat transfer was observed in boiling the aqueous solutions of PEO WSR-301 ($M = 2 \times 10^6$) and PAA Separan AP-30 ($15 \leqslant c < 150$ ppm) on a conical heater with the apex angle 143°. For PAA with $c > 60$ ppm the value of α began to decrease. The separation size of bubbles decreased with an increase in c, the generation frequency grew, and the tendency towards the coalescence of bubbles deteriorated. In refs. [9–11] the boiling of the solutions of PEO with $c = 0.002$–1.28% was investigated in the region of atmospheric and subatmospheric pressure for subheatings from 0 to 80 K. It is shown that the dependence of the heat transfer coefficient on the concentration of the polymer c in saturated boiling is nonmonotonous, since with a growth in c first α increases, attaining maximum at $c \approx 0.04\%$, whereas at $c = 1.28\%$ the value of α turns out to be smaller than in water ($\alpha < \alpha_s$). With a decrease in pressure, the effect of polymer additives deteriorates, and for solutions with the greatest concentrations of PEO the α value increases, approaching α_s from below. The critical densities of heat flux in PEO solutions appeared to be smaller than the corresponding values for water.

Finally, in ref. [12] a decrease in α with the growth of c was discovered for bubble boiling of aqueous solutions of technical PAA with concentration from 0.2 to 1% on a vertical tube of diameter 4.5 mm. In a subheated liquid the critical heat flux decreased with an increase in the concentration of PAA. This agrees with the observations made in ref. [9].

In view of the data given, work [13] attracts special attention. The result obtained in that work for diluted solutions was opposite to that in refs. [3–5]. When additives of PAA, PEO and HEC were introduced into water to concentrations corresponding to the increase in the viscosity of the liquid up to $\eta_p \leqslant 1.32 \times 10^{-3}$ Pa·s, a deterioration in heat transfer was observed. The boiling curve in terms of the variables q and ΔT displaced almost congruently to the region of large values of ΔT. Using Rohsenow's correlational formula for the heat transfer coefficient, the authors of ref. [13] showed that the observed decrease in α on the addition of polymer to water can be both qualitatively and satisfactorily quantitatively associated with the growth in viscosity. Note that experiments in ref. [13] were conducted on a thin platinum wire with diameter 0.3 mm.

Going over to the analysis of the aforementioned experimental results, it is first of all necessary to isolate physical factors, the manifestation of which is possible in boiling solutions of high-molecular compounds. Changes in capillary forces on phase interfaces in the presence of polymers, the adsorption of macromolecules on a heating surface, increase in the number of weak points [14] in a solution that promote the growth of the number of nuclei, thermodynamic features of the polymer–solvent system, the effect of macromolecules on diffusional mass transfer in evaporation of a solvent, the hydrodynamics of convective flows in a boiling layer, the motion of bubbles, and the manifestation of the rheological properties of a solution will also be due to these factors.

The role of capillary effects was indicated as one of the reasons for heat transfer enhancement, since many of the polymers (in particular, HEC and PEO) are capable of decreasing the surface tension coefficient. As a result, the work of the formation of a nucleus decreases [15]:

$$W_{cr} = 16/3\pi\sigma^3\Phi(\theta)[(\mathrm{d}p/\mathrm{d}T)\Delta T(1-\rho_1/\rho_2)]^2$$

$$\Phi(\theta) = 1/4(2+3\cos\theta-\cos^3\theta) \qquad (1)$$

where θ is wetting angle, and the critical size of the bubble decreases,

$$R_{cr} = 2\sigma[(\mathrm{d}p/\mathrm{d}T)\Delta T(1-\rho_1/\rho_2)]^{-1}.$$

However, it should be noted that the integral effect of the growth of heat transfer in ultimately diluted solutions cannot be explained by capillary phenomena alone, since the main change in σ occurs in the region of low polymer concentrations ($c \lesssim 50$ ppm [13]), and with a further growth in c the value of σ does not change, whereas α continues to grow. Moreover, PAA, for example, does not relate to surfactants at all. It should also be pointed out that, in the presence of a polymer, not only does the value of σ change, but also that of the wetting angle θ in formula (1). As noted in ref. [16], this factor may lead to the manifestation of the opposite tendency.

Adsorption of macromolecules onto a heating surface favours the formation of new centres. This, together with an increase in the number of weak points in the boundary layer of a boiling liquid, explains the general growth in the number of bubbles. Together with a decrease in σ for polymers–surfactants, this very factor is also responsible for a certain decrease in the superheat needed for the start of boiling [3, 9] of diluted solutions.

The decrease in the pressure of vapour at $c \approx 1\%$ can be neglected [17]. However, if a solution has a lower critical solution temperature (LCST) located below the wall temperature, a separation into polymer-enriched and polymer-depleted phases occurs in the boundary layer. At small concentrations of macromolecules, the first of these exists in a fine-dispersed state. This was observed for the solutions of PEO in work [9]. The role of the polymer-enriched phase reveals itself in the local increase of the saturation temperature, which can be appreciable at high polymer content after the separation, in the decrease of the intensity of convective transfer and motion of bubbles because of the increase of viscosity, and in the retardation of the growth of bubbles, as follows from the results of ref. [1]. A 'slow' crisis discovered in the solutions of PEO [9] is explained precisely by these reasons. Note that a similar phenomenon, but evidently less well expressed, was also observed in ref. [4] at a high enough concentration of polymer (a characteristic plateau on the boiling curve for the solutions of PIB in cyclohexane extending into the region of high superheats). One can agree with the fact [12] that decrease in the critical heat flux density in boiling polymer solutions with $\eta_p \gg \eta_s$ (in the cases when the usual crisis is possible at all) is caused by deterioration of the supply of liquid to the wall layer under conditions of high number densities of acting nucleation sites.

The main reason for the decrease in the heat transfer coefficient in bubble boiling of polymer solutions with $c \approx 1\%$ may be a notable increase in liquid viscosity, causing the deterioration of microconvection and increase in the resistance to rising bubbles. Thus, for example, when the Newtonian viscosity of a PEO solution in water is evaluated by the empirical formula [18]

$$\eta_p = \eta_s \exp[(250-0.9T)c],$$

then $\eta_p/\eta_s \approx 5$, at $c = 10^{-2}$, $T = 100$°C. The use of the data of refs. [19, 20] for aqueous solutions of PEO and PAA leads to values close in the order of magnitude. In the presence of LCST located below the boiling temperature, the role of this factor increases because of the saturation of the boiling layer with a polymer-enriched phase in the fine-dispersed state. Another reason for the decrease of α in the concentrational range discussed is the decrease in the rate of the growth of bubbles at the thermal stage at the superheats $\Delta T > \Delta\hat{T}^*$, as follows from the results of calculations [1].

In diluted solutions, viscosity undergoes an insignificant change and, though in ref. [4] correlation between the growth of α and increase in viscosity is noted, the reasons for this cannot be established. The well-known effect of the suppression of turbulence by small additions of polymers cannot also be the reason for the increase of α, since, conversely, the lamination of a boundary layer leads to the reduction in the intensity of convective heat transfer [21]. Nevertheless, the phenomenon of the decrease in the hydrodynamic resistance and enhancement of boiling heat transfer of diluted solutions seem to have a common nature.

EVOLUTION OF MICROBUBBLES ON A HEATING SURFACE AND BOILING HEAT TRANSFER

It seems that the effect discussed is due to the elastic properties of a solution representing the specific features of macromolecular systems in the course of the growth of vapour bubbles on a heating surface. The general character of the evolution of bubbles during boiling under conditions of atmospheric and reduced pressures, respectively, is clarified schematically in Fig. 1. In the first case, when the base of a bubble does not spread [22], but stays at the place of its initiation, a decrease in the bubble surface curvature with time due to the increase in R leads to the displacement of liquid from the zone indicated and, consequently, to the origination of the local shear in a thin layer of a polymer solution. A similar shear

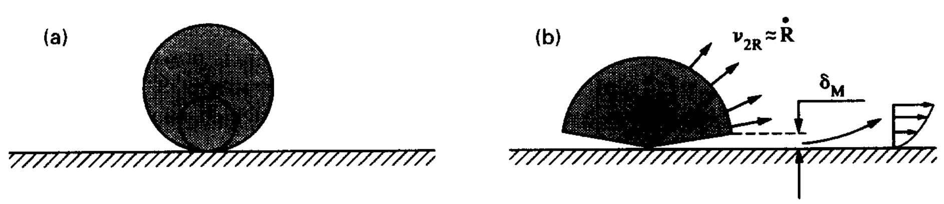

Fig. 1. Growth of vapour bubbles on a heating surface at high (a) and low (b) pressures.

flow also develops in the second case when a liquid microlayer is formed under a semi-spherical bubble. As is known, with shear of a viscoelastic medium, not only tangential but also normal stresses appear [23] (Weissenberg effect), reflecting the accumulation of elastic energy in the strained layer. The appearance of such stresses and elastic return of liquid to the vapour generation centre may turn out to be the sought-after reason for an early separation of a bubble from a heating surface, decrease in its size and growth in the frequency of the generation of bubbles observed experimentally. This ultimately leads to the enhancement of heat transfer.

In view of the above arguments, simple estimations can be made by the Cooper–Lloyd model [24] of the formation of a microlayer under a semi-spherical bubble. According to ref. [24], the microlayer thickness near the outer boundary of the bubble base (Fig. 1b) at time t, reckoned from the start of bubble growth, is $\delta_M = C(\nu t)^{1/2}$. The constant C varies from 0.6 to 1.1, depending on the rate of bubble growth [15]; for estimates it is possible to assume simply that $C = 1$. The function $R(t)$ is taken in the form [1]

$$R = (12/\pi)^{1/2} Ja\,(a_2 t)^{1/2}.$$

The liquid velocity on the outer edge of the boundary layer for the conditions far from the critical ones can be assumed to be equal to $\dot{R}$ [25]. Then, the shear rate $\dot{\gamma}$ has the order of the ratio $\dot{R}/\delta_M$

$$\dot{\gamma} \sim \dot{R}/\delta_M = (3/\pi)^{1/2} Ja/(t\,Pr^{1/2}) \approx 4a_2\,Ja^3/(R^2\,Pr^{1/2}). \tag{2}$$

It is most difficult to estimate the number Pr in equation (2), since the effective viscosity of the solution is very sensitive to changes in the concentration of polymer occurring in an evaporating microlayer and shear rate. At the same time one should take into account that at small times, because of the relaxational behaviour and manifestation of physical nonlinearity in the expected range of shear rates, the effective viscosity in equation (2) should not greatly exceed η_s. Let $Pr \approx 10$–10^2, then $\dot{\gamma} \approx 10^4$–$3 \times 10^4$ for $R \approx 10^{-4}$ m and $Ja \approx 20$. As $\dot{\gamma}$ increases, the normal stress τ_n on the area parallel to the solid wall also grows and at large shear rates can be large. The criterion for the manifestation of the effect is

$$N_T \equiv F_a^{-1} \int_s \tau_n \, dS \gtrsim 1$$

$$F_a = 2/3\pi R^3 g(\rho_2 - \rho_1) \tag{3}$$

where F_a is the buoyancy acting on a semi-spherical bubble. Selecting as S the base of a bubble above the microlayer (Fig. 1b) and assuming [23] that $\tau_n \approx \beta\eta_p\lambda\dot{\gamma}^2$ ($\beta\eta_p$ is the contribution of a dissolved polymer to the effective viscosity, $\beta < 1$), equation (3) yields for $R \approx 10^{-4}$ m : $N_T \approx (\beta\eta_p\lambda) \times 10^8$. If it is taken into account that the last measurements of natural time for the solutions of macromolecules with $c \approx 100$ ppm and $M \approx 10^6$ give the value $\lambda \approx 5 \times 10^{-4}$ s (a solution of PEO in water at $T = 100°$C [26], which at a given value of $\dot{\gamma}$ corresponds to the Weissenberg number $We \approx 10$), as well as the factor of the concentration of solution in the microlayer by evaporating the solvent, then the fulfillment of the condition $N_T \approx 1$ turns out to be quite real despite the conventionality of the estimations made.

Taking account of the aforegoing, the result obtained in ref. [13] finds its explanation. Its reason is evidently associated with substantial differences in the conditions of boiling on a thin wire and a plate or a tube. Steam bubbles growing on a wire have a size commensurable with the wire diameter (in ref. [13] a developing bubble dragged around a wire) with the result that the role of the boundary layer decreases sharply, and also of the effects of normal stresses. Moreover, a substantial decrease in the rate of the growth of a bubble is also of importance as compared with boiling on a plane (for a wire $R \sim t^n$, where $n < 1/4$ [22]).

Note that the manifestation of elastic properties of the solution also occurs because of the stabilization of a spherical shape of vapour inclusions observed in boiling and cavitation experiments. Finally, a decrease in the tendency toward coalescence and an increase in the nonuniformity of the dimensions of the bubble phase, noted repeatedly in the literature, can also be attributed to the effects of normal stresses and longitudinal viscosity in thin films formed on the coalescence of bubbles.

One should also pay attention to the fact that the connection of the effect of increase in boiling heat transfer of diluted polymer solutions with elastic properties of the system is confirmed by the existence of optimal concentration corresponding to α_{max}. As is known, a similar concentration also exists when polymer additives are used for suppressing turbulence where the elasticity of macromolecules plays the main role. Therefore, it is possible to expect that the factors favouring the formation of an expanded conformation of macromolecules and increase in the flexibility of chains, as well as an increase in the molecular mass, should lead to the strengthening of the effect.

New data [27] on heat transfer of aqueous solutions of PAA and HEC having the concentrations 60–1000 ppm and 500–5000 ppm, respectively, and boiling on a thin wire merit a special mention. The conclusions drawn in ref. [27] regarding the solutions of PAA in the entire concentrational range investigated agree with the data obtained earlier by oher authors and confirm the validity of the above arguments. At the same time, the data for the solutions of HEC point to the possibility of the manifestation of a fundamentally new effect, i.e. the superintensification of heat transfer when $c \gtrsim 1000$ ppm. The effect is described by the reserved branch of the curve of boiling on a plane $(\Delta T, q)$ occurring at $q = q \approx 13$ kW cm^{-2}, $\Delta T \approx 15$ K. It is characterized by a decrease in ΔT with an increase in the heat flux in the region with $q > q_c$. Unique observations made in work [27] stimulate an interest in more detailed experimental investigations of boiling of weakly concentrated solutions of HEC leaving unresolved the problem of the possible reasons for the phenomenon.

Analysis of the data on the boiling of concentrated solutions [17] shows that in such systems thermodynamic, diffusional and rheological factors are brought to the fore. The diagram of the phase liquid–vapour equilibrium for concentrated solutions ensures the decrease in the derivative $\mathrm{d}p/\mathrm{d}T$ (when $k_0 \to 0$, $\mathrm{d}p/\mathrm{d}T \to 0$). This leads to the growth of the work of the formulation of a nucleus (1), separation size (2) and, consequently, to a decrease in the frequency of generation of vapour bubbles. Note that in reality the critical work W_{cr} for a polymer fluid may exceed the value predicted by formula (1) because of the appearance of the elasticity of macromolecules.

As is known [28], the heat transfer coefficient in the case of developed bubble boiling of low-molecular fluids $\alpha = Aq^n$ where $n \approx 0.6$–0.7. For concentrated polymer solutions the exponent n is close to zero. The decrease in heat transfer is explained by the increase in the viscosity of the solution near the heating surface conditioned by the evacuation of the solvent with vapour phase. Another reason for the decrease of α, as follows from the results of ref. [1], is a decrease in the rate of growth of bubbles with a decrease in k_0 and the impossibility of attaining large Ja numbers by the growth of the superheat in solutions with a considerable polymer content.

Since α is small in the boiling of concentrated polymer solutions, the superheat of the wall at a fixed q increases and this may lead to the phenomena of structure formation, scorching, and thermal decomposition. Usually, the intensification of heat transfer is made by mechanical means. Note that one of the promising trends in this field may be the use of ultrasonic effects, the efficiency of which should be predicted with account of a considerable decrease in real losses in acoustic flows and pulsations of bubbles in an elastoviscous medium [2].

CONCLUSIONS

The available experimental results point to the enhancement of bubbles boiling heat transfer of diluted ($c \approx 0.01\%$) solutions of polymers on a heater with a characteristic size which is much higher than the mean size of bubbles. An explanation is suggested for the effect, taking into account the generation of normal stresses with deformation of a thin layer of a polymer fluid between a growing vapour bubble and a wall. The considerations given are confirmed by the evaluational calculation made on the basis of Cooper–Lloyd's model. As a result, the separation of bubbles from the heating surface accelerates, the frequency of generation increases and, as a consequence, the heat transfer coefficient becomes higher. The factors favouring the formation of an extended conformation of macromolecules (improvement of the thermodynamic affinity with a solvent, the growth of the degree of ionization of macromolecules, etc.), increase with the flexibility of chains, as well as increase in the molecular weight, must lead to strengthening of the effect.

In boiling weak ($c \approx 1\%$) solutions a decrease in the heat transfer coefficient is observed as compared with a pure solvent which is connected with the retardation of the growth of vapour bubbles and suppression of microconvection in the wall zone because of the growth of viscosity.

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