

Minimum Film Boiling Temperatures for Spheres in Dilute Aqueous Polymer Solutions and Implications for the Suppression of Vapor Explosions

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폴리머 수용액에서 구형체의 최소막비등온도와 증기폭발 억제 효과

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Abstract

Pool boiling of dilute aqueous solutions of polyethylene oxide polymer has been experimentally investigated for the purpose of understanding the physical mechanisms of the suppression of vapor explosions in this polymer solution. Two solid spheres of 22.2mm and 9.5mm-diameter were heated and quenched in the polymer solutions of various concentrations at 30°C. The results showed that minimum film boiling temperature(ΔT_{MFB}) in this highly-subcooled liquid rapidly decreased from over 700°C for pure water to about 150°C as the polymer concentration was increased up to 300ppm for 22.2mm sphere, and it decreased to 350°C for 9.5mm sphere. This large decrease of minimum film boiling temperature in this aqueous polymer solution may explain its ability to suppress spontaneous vapor explosions. Also, tests with applying a pressure wave showed that the vapor film behaved more stable against an external disturbance at higher polymer concentrations. These observations together with the experimental evidences of vapor explosion suppression in dilute polymer solutions suggest that the application of polymeric additives such as polyethylene oxide as low as 300ppm to reactor emergency coolant be considered to prevent or mitigate energetic fuel-coolant interactions during severe reactor accidents.

요 약

폴리머 수용액의 증기폭발 억제 효과에 대한 물리적 현상을 이해하기 위해 폴리에틸렌옥사이드 수용

액에서의 풀비등 특성을 실험적으로 관찰하였다. 본 실험에서는 22.2mm와 9.5mm 직경의 두 구형체를 가열하여 여러가지 농도의 30°C 수용액에서 냉각시켰다. 그 결과, 순수한 물에서는 70°C 이상인 최소막비등온도(ΔT_{MFB})가 300ppm 농도의 폴리머 수용액에서 22.2mm 구의 경우 150°C 까지, 9.5mm 구의 경우 350°C 까지 낮아짐을 알 수 있었다. 이러한 폴리머 수용액에서 최소막비등온도가 크게 낮아지는 현상은 이 수용액에서 증기폭발이 억제되는 이유로 해석될 수 있다. 또한, 외부 압력파의 막비등에 대한 영향을 관찰한 결과, 수용액의 농도가 클수록 증기막의 안정도가 커짐을 알 수 있었다. 이러한 폴리머 수용액에서의 비등 특성과 증기폭발 억제에 대한 실험 결과들은 원자로 비상냉각수에 폴리에틸렌옥사이드와 같은 폴리머를 최소 300ppm 정도 소량 첨가하는 방법으로 중대사고시 폭발적 FCI 반응을 방지 또는 완화할 수 있음을 제시한다.

1. Introduction

When a hot, superheated surface is immersed into a volatile liquid, the liquid boils at the surface and, in general, such boiling exhibits one of the three modes of nucleate boiling, transition, and film boiling, depending upon a number of conditions such as surface and liquid temperatures. Film boiling is that the vaporization is so high that vapor film forms between the surface and the liquid, as first observed by J.G. Leidenfrost in water droplets hovering on a red-hot spoon. As the surface cools down, the vapor film collapses allowing contact of the surface and the liquid (transition boiling), and nucleate boiling follows. The minimum temperature that supports stable film boiling is often referred to as the Leidenfrost temperature, or minimum film boiling temperature.

In nuclear reactor safety, the minimum film boiling conditions are, on one hand, associated with the conditions considered necessary for initiation of energetic fuel-coolant interactions (FCIs), or vapor explosions. Such an event is a physical explosion in which the stored internal energy in the hot liquid is converted to produce work by a high pressure vapor. Since this process has the potential of destructive mechanical energy release and high pressure load to the system, it has been a safety concern in severe nuclear reactor accidents as well as in many industrial accidents of such as metal foundries and paper mills[1].

In a typical vapor explosion when the two liquids first come into contact, the coolant begins to vaporize at the fuel-coolant liquid interface as a vapor film sep-

arates the two liquids. Then, vapor film destabilization occurs, triggering fuel fragmentation. The vapor film destabilization occurs spontaneously or by an external disturbance such as pressure pulses. For this reason, the mechanics of vapor film behavior with and without a pressure pulse has drawn a great attention in the area of vapor explosions research for understanding and modeling triggering mechanisms. Recently, it also led to the efforts of seeking a way of suppressing vapor explosions by changing coolant properties. This has been done by dissolving a small amount of polymers or surfactants into water[2–8].

One of the most useful features of polymers is their ability to increase the viscosity of a liquid in which they are dissolved, even at low concentrations. Also, dilute aqueous polymer solutions have been studied for many years in the area of drag reduction in turbulent flows and thermal systems. From this background, many kinds of water-soluble polymers have been studied for the effects of coolant viscosity on the suppression of vapor explosions and some polymers such as cellulose gum and polyethylene oxide were reported to have the ability to suppress vapor explosions[5–7]. However, the cut-off concentration depended on the type of polymers, and was as high as 240 times higher viscosity than pure water for triggered explosions of iron oxide and water[6]. Such high viscosity seems to discourage a practical application of the results.

The recent work of Ip et al. [7] showed that the dilute aqueous solution of polyethylene oxide(PEO) showed the ability to suppress spontaneous vapor

explosions at low viscosity increase. They conducted small-scale experiments of dropping 12g of molten tin into dilute PEO solutions and observed that spontaneous explosions were markedly suppressed when the polymer solution was twice viscous as pure water. This result of PEO solutions was tested again in a large-scale facility by Park et al. [8]. In this experiment, molten tin up to 5kg was dropped into the polymer solutions at room temperature and no explosive event was observed even at low viscosity increase (about two fold) and under external triggering.

The ability to suppress vapor explosions by the dilute PEO solutions seems to be a promising result for preventing energetic fuel-coolant interactions, however, the physical mechanisms of the suppression effect and the effect of an external trigger on the cut-off viscosity have not been clearly identified. The possible cause of the suppression of spontaneous explosions may be associated with a change in boiling characteristics of the polymer solutions. The past work on the boiling curves of aqueous polymer solutions were mostly limited to nucleate boiling on flat plates and wires [9–11]. Rouai and Abdel-Khalik [12] conducted quenching experiments of hot brass spheres in several polymer solutions at 90°C. They reported that the critical heat flux increased by more than 50% while the minimum film boiling temperature increased by more than 110°C when concentrations of 100ppm of polyethylene oxide or guar gum were used. However, these data seem not sufficient in explaining the explosion suppression effects of the polymer solutions due to the higher solution temperature compared to highly-subcooled room temperature that is considered here, nor one finds any clear relevance to the explosion suppression effect.

In the present study, quenching experiments of hot solid spheres in dilute aqueous polymer solutions, mainly polyethylene oxide, have been conducted for the purpose of investigating the physical mechanisms of the vapor explosion suppression effects. This paper reports the observations of pool boiling characteristics and minimum film boiling temperatur-

es of the dilute aqueous polymer solutions. Also the film boiling behavior under an external pressure pulse is presented.

2. Experiment

2.1. Experimental Apparatus

The apparatus consisted mainly of test chamber, furnace, trigger, and the test sphere assembly. Figure 1 shows a schematic of the experimental apparatus. The test chamber was an open-topped rectangular construction using clear polycarbonate plates for the purpose of visual observation. The inner dimensions of the chamber were 150mm in height and 90mm each side. The base of the chamber was a movable circular disk sealed with o-ring. A pressure wave was produced by tapping this plate using a pneumatic cylinder when the effect of an external disturbance was investigated. On a side wall of the chamber a

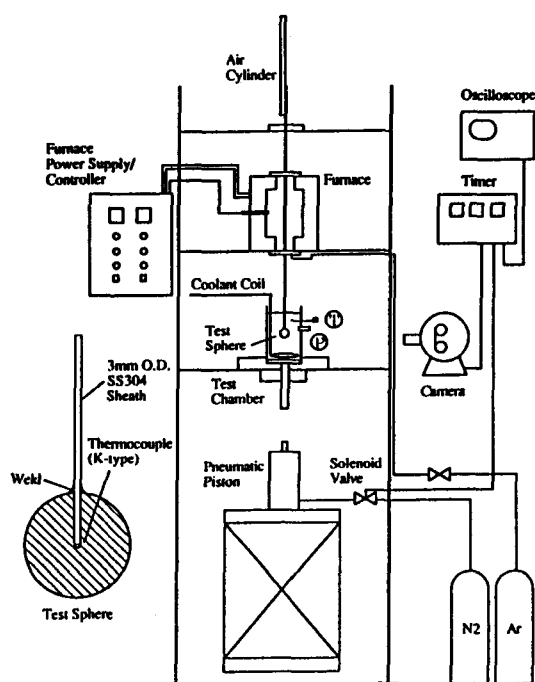


Fig. 1. Experimental Apparatus

T-type thermocouple was inserted to monitor pool temperature and a piezoelectric pressure transducer (PCB Model 112) was flush-mounted to measure dynamic pressure produced in transient boiling. A charge amplifier was used to amplify the signals from the pressure transducer. The pool temperature was controlled by circulating water through a copper coil immersed into the pool and a constant-temperature bath.

An electric furnace was used for heating the test sphere. It was fabricated using two half-cylinder heaters of 1.3kW capacity in total. A K-type thermocouple was inserted into the inner space of the furnace and connected to a temperature controller to regulate the furnace temperature at a desired temperature. The furnace was continuously purged with argon gas to minimize the surface oxidation of the test sphere.

The test spheres were stainless steel 304 balls and two sizes were tested; 22.2mm and 9.5mm in diameter. To measure the transient temperature of the spheres, the spheres were drilled to the center and a K-type thermocouple, sheathed with 3mm outer-diameter stainless steel tube, was inserted to the center and the sheath was welded at the sphere surface. To obtain fast sampling of the thermocouple signal, an electronic ice-point compensator and a millivolt amplifier were placed between the thermocouple and the digital oscilloscope. The sphere assembly was mounted on an air cylinder for easy and fast release from the furnace to the test chamber. The release of the sphere, data acquisition trigger, and all other motions in the apparatus were controlled using a set of electronic timers.

2.2. Data Reduction

To obtain the sphere surface temperature from the measured temperature-time trace at the center of the sphere, the inverse conduction problem was solved. Since for the stainless steel sphere the Biot number is greater (>0.1), the lumped capacity model is not

accurate for the present cases. Knowing the temperature of the center of the sphere as a function of time and the time of minimum film boiling point from the pressure signal, the surface temperature and heat transfer coefficient were calculated by solving 1-D transient conduction equation for the sphere with convective boundary condition[13].

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T \quad (1)$$

$$\Theta = \frac{T - T_L}{T_o - T_L} = f(\eta, Fo, Bi) \quad (2)$$

Hence, the dimensionless temperature, Θ , is a function of radial position $\eta (=r/R)$, Fourier number $Fo (= \alpha t/R^2)$ and Biot number $Bi (= hR/k)$. The solution was obtained by assuming constant thermophysical properties and uniform and constant heat transfer coefficient.

3. Results and Discussions

3.1. Polymer Solutions and Properties

The polymer solutions were prepared by adding slowly the dry polymer in powder form into distilled water in a large bucket while the water was stirred using a motor-driven stirrer at about 300rpm. Then, the stirring was continued for another 4 to 8 hours depending on the concentration until the polymer was dissolved completely.

The viscosity of the solution was measured with a size 50 Cannon-Fenske type capillary viscometer. Figure 2 shows the solution viscosity increases of the polyethylene oxide of the average molecular weight of 4×10^6 at various concentrations. In this figure, calculations of solution viscosity using the Mark-Houwink-Sakurada equation[14] are compared with the measured viscosity and a good agreement is observed. For a few tests with different type of polymers, solutions of polyvinyl acetate-vinyl alcohol (PVAA) with 12% acetate were prepared. The viscosity increases of these solutions are much smaller

than PEO's; it requires 4,000ppm compared to 300ppm of PEO to get the viscosity increase of 1.5.

In addition to changing the solvent viscosity, it is known that surface tension of the PEO solutions is reduced by 10% even in high concentrations[10]. The effect of surface tension reduction by this amount is probably not significant in film boiling. Except for the solution viscosity and surface tension, it is known that all other physical properties of water remain unchanged at the low concentrations of polymers used in these experiments.

3.2. Pool Film Boiling

Pool boiling of hot solid spheres in dilute polyethylene oxide solutions at primarily room temperature (30°C) was investigated. In pure water at 30°C, the sphere at initially 700°C quenched rapidly and it was obviously in nucleate boiling regime from the beginning as confirmed also from the visual observation. Such difficulty of obtaining stable film boiling in this highly subcooled water was also observed in the data reported by Dhair and Purohit[15]. They con-

ducted film boiling experiments for spheres and obtained minimum film boiling data of pure water up to 50°C of subcooling. The extrapolation of these data indicates that more than 660°C of wall superheat ($T_{WALL} - T_{SAT}$) is required to obtain stable film boiling in 30°C water.

$$\Delta T_{MFB} = 101 + 8\Delta T_{SUB} \quad (3)$$

Here, ΔT_{SUB} is the liquid subcooling ($T_{SAT} - T_L$). The theoretical model of minimum film boiling temperature for spheres proposed by Gunnerson and Croneberg[16] predicts 1,250°C of wall superheat in this case. However, heating the sphere beyond 700°C was avoided in the present study due to the problem of possible phase change of stainless steel when heated over this temperature range, and hence change of material properties.

In the PEO polymer solutions at room temperature, however, the boiling pattern markedly changed. The sphere quenched rather slowly in film boiling regime and, for instance, this stable film boiling mode lasted for 35 seconds in 300ppm solutions for 22.2mm diameter sphere. Figure 3 shows the tempera-

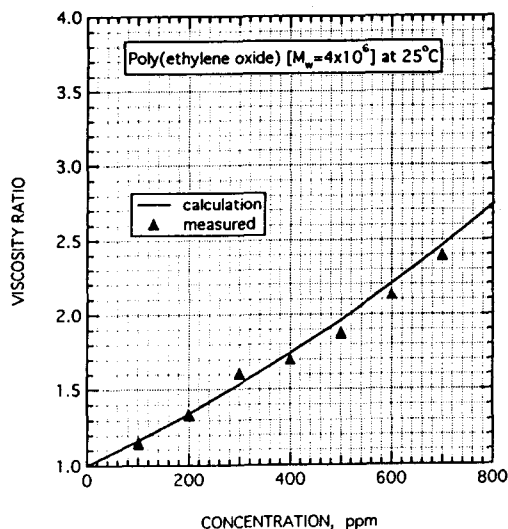


Fig. 2. Viscosity Increase of Aqueous Solutions of Polyethylene Oxide

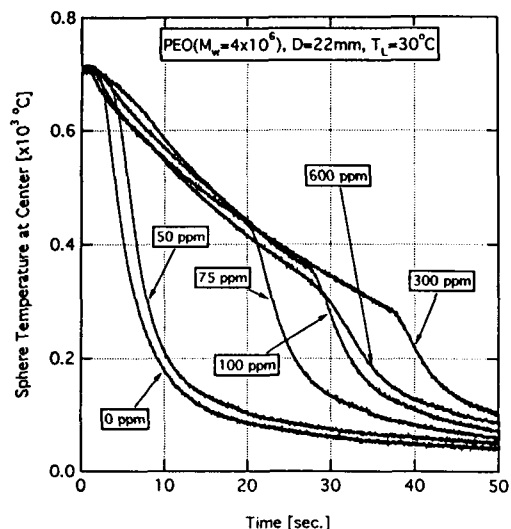


Fig. 3. Temperature-Time Traces of 22.2mm Sphere in Various Polymer Concentrations

ture-time traces of the 22.2mm diameter sphere quenched from the same initial temperature of 700°C, but in the polymer solutions of various concentrations. The period of film boiling rapidly increased when the concentration was increased from 50ppm to 75ppm. Above 300ppm, however, the film collapse tended to occur slightly earlier. This is because the film boiling heat transfer coefficient becomes larger when the concentration was further increased as shown in Fig. 8. For 9.5mm sphere, the similar behavior of film boiling was also observed, except that the transition boiling occurred at higher wall temperatures compared to 22.2mm sphere. This will be discussed later in terms of minimum film boiling temperature.

The visual observations showed clearly the three modes of boiling; (a) stable film boiling, (b) film collapse, and (c) nucleate boiling. Figure 4 shows the typical photographic views of these three boiling modes. Also in the polymer solutions, the collapse of vapor film around the sphere was clearly identified as a violent event. A big sound was heard at this point, and a pressure peak was recorded in the pressure transducer signal as shown in Fig. 5. This pressure peak is a more accurate indication of the minimum film boiling point than the point of the beginning of rapid temperature decrease, which past investigators often assumed for the transition point.

In Fig. 5, the calculated surface temperature and the center temperature of the sphere are plotted together with the measured center temperature in 300ppm PEO solutions. It shows a good agreement between the measurement and calculation of the center temperature except the early time. This is because the heat transfer rate was relatively higher in this early period of initial contact between the sphere and the pool.

In the quenching tests in the solutions of polyvinyl acetate-vinyl alcohol at the concentrations of 1,000 and 4,000ppm, which increases the solution viscosity by 1.1 and 1.5 respectively, the long-lasting stable film boiling did not occur although the viscosity

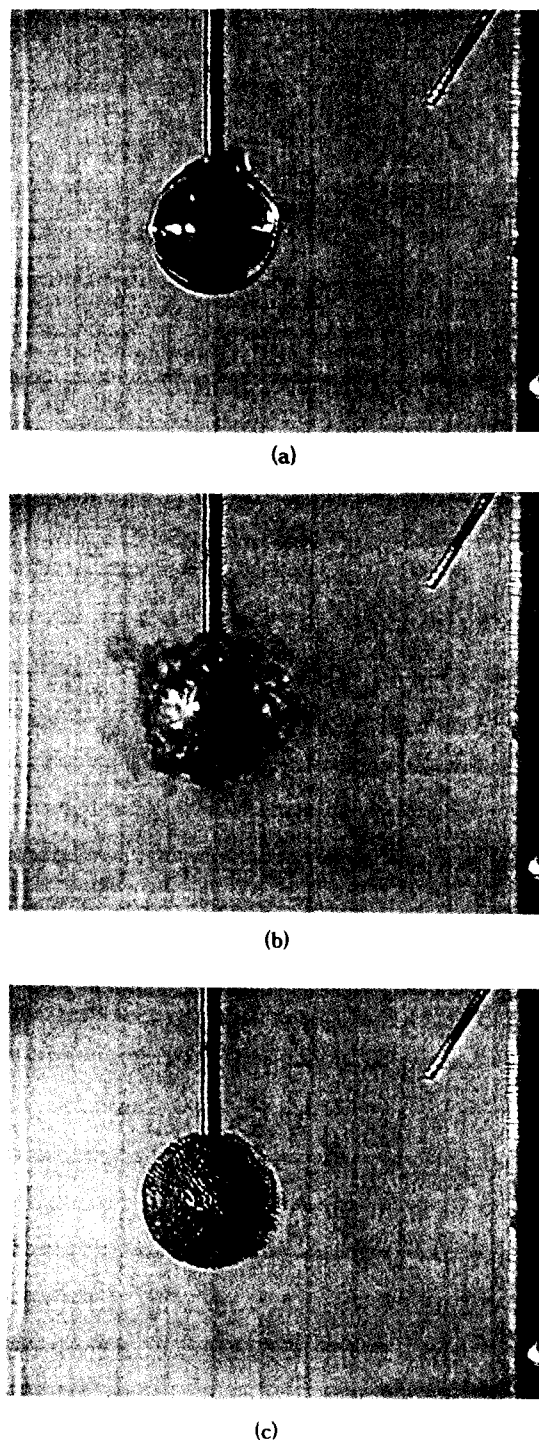


Fig. 4. Photographic Illustration of Pool Boiling Modes : (a) film boiling, (b) transition, and (c) nucleate boiling

increases were in the same range of the PEO cases. It was observed that nucleate boiling occurred from the beginning with thicker cloud of bubbles around the sphere than in pure water.

3.3. Minimum Film Boiling Temperatures

From the calculated surface temperatures of the spheres, the minimum film boiling temperature differences ($\Delta T_{\text{MFB}} = T_{\text{WALL,MIN}} - T_{\text{SAT}}$) were obtained and shown in Fig. 6 as a function of polymer concentration. In this figure, the value for the pure water is shown together for comparison, which is extrapolated from the data obtained by Dhir and Purohit[15]. For 22.2mm diameter sphere, the minimum film boiling temperature decreased rapidly from over 700°C for pure water to about 150°C as the concentration was increased up to 300ppm, and it did not decrease further from this level when the concentration was further increased. Such trend persisted also for 9.5mm diameter sphere except that the reduction is smaller, down to about 350°C. This is the major unique observation in the present study for the effect of dilute aqueous solution of polyethylene oxide on pool boiling,

and this seems to be the key mechanism in explaining the vapor explosion suppression in this polymer solutions.

The present results are, however, contrast to the results obtained by Rouai and Abdel-Khalik[12] in the past. They conducted similar experiments using 4.76mm and 6.38mm-diameter brass spheres at C pool temperature. Their results showed that the PEO solutions increased minimum film boiling temperature by 110°C. It is noted that in these two experiments the pool temperatures are largely different; 90°C compared to 30°C of the present study.

In order to identify a possible cause of the disagreement, a series of tests using 9.5mm-diameter sphere were conducted in heated pool to 80°C and Fig. 7 shows the results of temperature-time traces. Again in these heated pool tests, the sphere quenched in film boiling much longer than in pure water. Also, the heat transfer rates seem to be almost same regardless of the polymer concentration by noting the nearly same slopes of temperature drop in film boiling regime. For heated solutions, one must note that the PEO in aqueous solutions retains to a greater or lesser extent the helical conformation and the

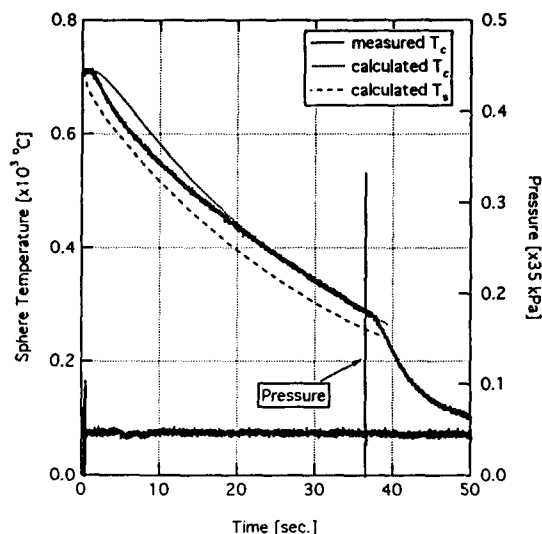


Fig. 5. Comparison of Measured and Calculated Temperature Traces

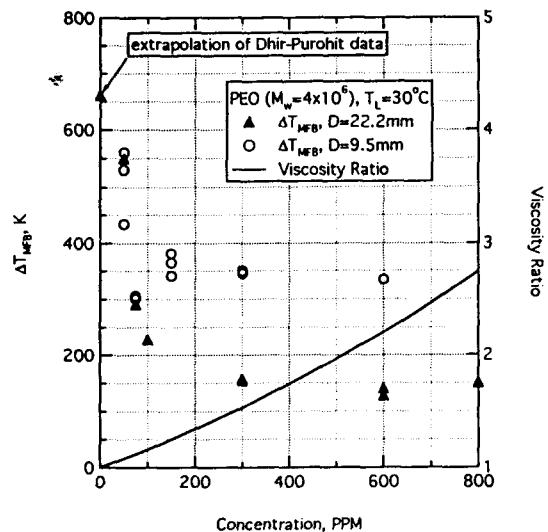


Fig. 6. Minimum Film Boiling Temperatures v.s. Polymer Concentration

melting point of these helical sections is around 40°C[14]. This may alter the properties of PEO aqueous solutions kept at high temperature for a long period of time, causing uncertainties in the data.

It is noted that the solution viscosity increases nearly linearly with the concentration, while the minimum film boiling temperature in the solutions drops rapidly within the range of 100ppm and stays unchanged even at higher concentrations. This indicates that the viscosity may not be the only contributing parameter for this change of minimum film boiling temperature in the polymer solutions. One may also note that currently existing models of minimum film boiling temperatures[16,17] are not significantly affected by liquid viscosity or surface tension in the ranges considered here. This may imply that the interfacial phenomena at the vapor-liquid interface at which polymer is dissolved play a greater role in such changes of minimum film boiling temperature, particularly with long linear-chain polymers like PEO.

3.4. Heat Transfer Coefficients

The average heat transfer coefficients of pool film

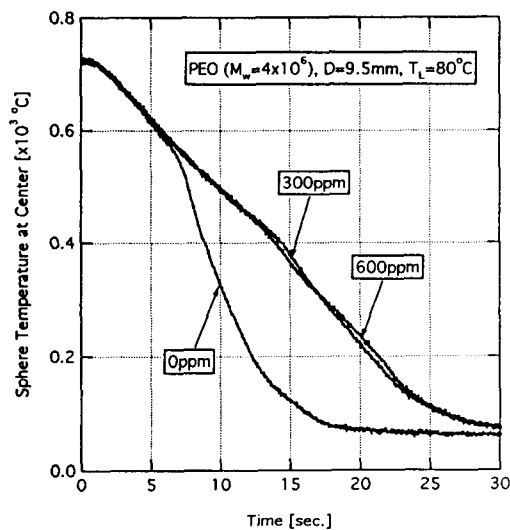


Fig. 7. Temperature-Time Traces of 9.5mm Sphere in 80°C Solutions

boiling in PEO polymer solutions are shown in Fig. 8. Although it varies slightly as concentration changes, the heat transfer coefficients are not changing significantly while the solution viscosity increases linearly as the concentration increases. The correlations of pool film boiling heat transfer for water are scarce, but one proposed by Dhira and Purohit[15] is given as

$$Nu = \frac{hD}{k_v} = Nu_o + Nu_{nc} \frac{Pr_v Sc}{Pr_l Sh \mu} \quad (4)$$

In this equation, radiation contribution is neglected, which is less than 10% of total heat transfer in the cases considered here. The expressions for Nu_o , Nu_{nc} and nomenclatures are given in Ref. [15], hence omitted here. One notes that this correlation was made with data obtained up to 50°C of liquid sub-cooling.

The prediction of heat transfer coefficients by this equation is given in Fig. 8 for comparison purpose. In these calculations only liquid viscosity was altered as concentration changes. From this figure it is observed that the measured and predicted heat transfer coefficients are of similar magnitude while the boiling

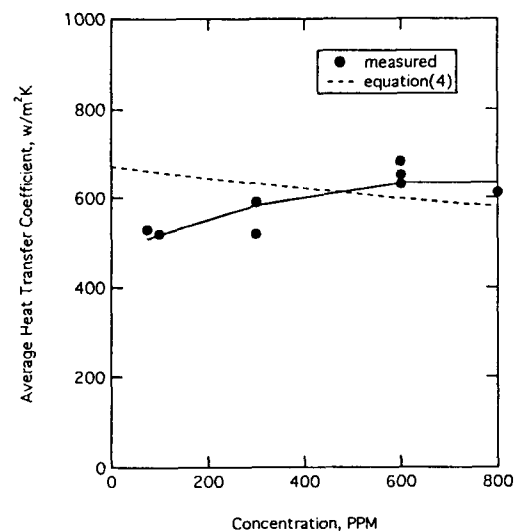


Fig. 8. Average Heat Transfer Coefficients in PEO Solutions

behaviors are significantly different between pure water and polymer solutions. This may imply that in polymer solutions the stability of vapor film is enhanced by the dissolved polymer chains at the interface beyond the thermal and hydrodynamic balances, possibly causing a violent motion of the interface when the film collapses at much lower wall temperatures than pure water.

3.5. Implications for Suppression of Vapor Explosions

The large decrease of minimum film boiling temperature in the polyethylene oxide aqueous solutions can explain its ability to suppress spontaneous vapor explosions. That is, in the polymer solutions, a drop of molten metal undergoes more stable film boiling at even low temperature. Thus at the time that the vapor film eventually becomes unstable, which is generally considered as one of the plausible triggering mechanisms of spontaneous vapor explosions, the melt drop surface is already frozen, preventing disintegration of the drop. This is why a spontaneous explosion is less likely in the polymer solutions.

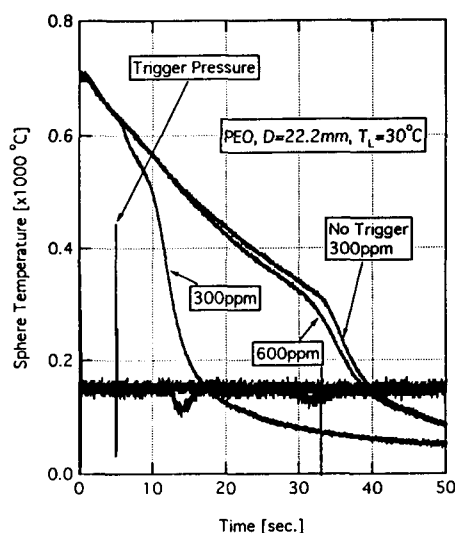


Fig. 9. Effect of External Disturbance on Film Boiling

In the case when an external disturbance like a pressure wave exists, the vapor film can be destabilized before the surface cools down to minimum film boiling condition. In order to investigate the effect of an external disturbance, a series of tests were conducted in which the base of the test chamber was tapped to produce pressure waves. Figure 9 shows the temperature traces of the 22.2mm sphere when the pressure wave was applied five seconds after the sphere was dropped into the pool. The pressure pulse was typically 50kPa in peak. In 300ppm solutions, the vapor film collapsed by the disturbance. In 600ppm solutions, however, stable film boiling was sustained after a short period of vapor film disturbance. Therefore, it is likely that the polymeric additives make the vapor film more stable against an external disturbance.

In reactor accident cases, molten materials like corium at much higher temperatures of around 2,500°C are concerned and this melt has relatively a small amount of superheat from the melting point; typically a few hundred degrees. Because of this, corium is likely to freeze at the surface soon after it contact the coolant and an initiation of energetic fuel-coolant interactions may be less likely. However, laboratory experiments of vapor explosions of corium and water have shown the occurrence of vapor explosions in this system[1]. Therefore, applications of polymeric additives such as polyethylene oxide to reactor emergency coolant may greatly contribute to the suppression of energetic fuel-coolant interactions due to the enhancement of vapor film stability in the polymer solutions. However, for the practical applications of dilute polymer solutions to reactor safety systems, it must be tested for wider ranges of conditions such as melt types and fuel and coolant temperatures. Also the aging effect of polymer solutions must be taken into consideration.

4. Conclusions

Quenching experiments of hot solid spheres in di-

lute aqueous solutions of polyethylene oxide polymer have been conducted for the purpose of investigating the physical mechanisms of the suppression of vapor explosions in this polymer solution. Two spheres of 22.2mm and 9.5mm-diameter were tested. It was heated to 700°C in a furnace and immersed into polymer solutions of various concentrations at 30°C.

The key observation in this study was that the minimum film boiling temperature (ΔT_{MFB}) in this highly-subcooled liquid rapidly decreased from over 700°C for pure water to about 150°C as the polymer concentration was increased up to 300ppm for 22.2mm sphere, and it decreased to 350°C for 9.5mm sphere. This large decrease of minimum film boiling temperature in the PEO aqueous solutions may explain its ability to suppress spontaneous vapor explosions. It was also observed that the vapor film behaved more stable against an external disturbance at higher polymer concentration. This may imply that in polymer solutions the stability of vapor film is enhanced by the dissolved polymer chains at the interface beyond the thermal and hydrodynamic balances, possibly causing a violent motion of the interface when the film collapses at much lower wall temperatures than pure water.

In order to apply such dilute polymer solution technique to the prevention or mitigation of energetic fuel-coolant interactions during severe nuclear accidents, it must be tested for wider ranges of conditions such as melt types and fuel and coolant temperatures. Also the aging effect of polymer solutions must be taken into consideration.

Acknowledgment

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