SUPPRESSION MEASURES AND EFFECTIVE TRIGGERING RETARDANT OF STEAM EXPLOSIONS

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ABSTRACT

Steam explosion has been a potential threat during severe accident in light water reactors. We had reported that Polyethylene glycol (PEG) is a reliable retardant of steam explosions. Experiments were conducted to investigate the effects of concentration, molecular weight and salt additives on the controllability of steam explosions. Steam explosion was suppressed with a 0.03 wt% PEG solution for molecular weight of 4 million. This is because the cloudy-point phenomenon stabilizes vapor film and prevents the solution from mixing finely by the precipitated solute near the steam-water interface. The stabilizing effect of vapor film was confirmed in a solid stainless-steel sphere quenching experiment as well. The molecular weight must be selected in reference to the cloudy-point temperature to be lower than saturation temperature by a certain degrees at the target pressure. At atmospheric pressure, a molecular weight of 4 million is demonstrated to suppress steam explosions. The effective concentration became denser when large share stress and/or external force act on the vapor film. Steam explosion may occur in a PEG solution by adding 1wt% of sodium chloride, because such salts act as steam explosion promoter.

KEYWORDS
Steam explosion retardant, triggering, polyethylene glycol, additives, cloudy-point phenomenon

1. INTRODUCTION

Steam explosion has been causing disasters in chemical and metallurgy industries. In addition, Fletcher [1] reviewed the role of triggering in steam explosions in terms of nuclear reactor safety. Reliable countermeasures are crucial to cope with the industrial disasters by steam explosion. Large scale steam explosion is generally considered to proceed in the following four stages as shown in Fig. 1: (1) coarse mixing, (2) triggering, (3) propagation, and (4) expansion. Triggering is the event that initiates the rapid, local heat transfer and pressure rise that are necessary if a propagating wave is to develop. In spite of the industrial relevance, knowledge of the triggering mechanism is insufficient even for a simple molten-alloy droplet system since the experimental data are scattered even if initial conditions are set to be identical due to the random nature of steam explosions.

The authors [2] found that there are three factors that change triggering conditions sensitively during the premixing stage, resulting in the loss of reproducibility: (1) the heat transferred from a hot fluid to a cold fluid, (2) configuration and orientation of the interface where triggering occurs, and (3) the property of the surface, e.g., the oxide layer formation. The authors [3] have performed experiments in the droplet

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impingement configuration to suppress the premixing stage. Steam explosion occurred approximately one millisecond after a water droplet had impinged onto the molten alloy surface. In this configuration, changes in initial temperatures and surface conditions before contact affect the triggering phenomena directly without any interference with the premixing stage.

It has been reported that steam explosion was suppressed by means of surfactant [4]-[8] or polymeric [9]-[13] addition into water. As polymeric additives are known to increase the fluid viscosity, a viscosity ratio η_r (= $\eta_{solution} / \eta_{water}$) is referred to as the index instead of concentration. Dowling and Adbel-Khalik [9] and Ip *et al*. [10] found that steam explosion was entirely eliminated at $\eta_r = 2$. This upper limit viscosity ratio or concentration, however, differed among researchers. The mechanism to prevent steam explosion from occurring remains unknown. Possible factors are the restraint of relative motions in the premixing phase and prevention of fine mixing in the triggering phase.

Groenveld [5] was the first to address the suppression ability of a surfactant additive because the surfactant stabilizes the vapor film by depositing at the vapor-liquid interface. However, Chapman *et al*. [12] reported that representative surfactants did not exhibit suppression and mitigation effects. Experimental results differ among researchers. All the experiments were conducted in the system of the molten tin droplet impinging into a water bath. The surfactants may not have deposited on the liquid/vapor interface to exert their effect in such the configuration, since it took certain time to trigger a steam explosion at such a high impingement velocity (about a few meters per second).

The authors [14] have investigated the suppression effect of surfactant, polymeric and salt additives in the droplet impingement configuration, and found that polyethylene glycol (PEG) has substantial suppression effect at the concentration of only 0.02 wt%.

This paper addresses the additional concerns of using PEG as a steam explosion retardant. The experiments were conducted with tin droplets into aqueous solution pool to investigate the effects of molecular weight of PEG polymer and coexistence of a salt. In order to gain insight of the triggering mechanism, experiments were conducted with an aqueous solution droplet impinging onto molten lead bismuth surface. Finally vapor-film stability were investigated with a solid stainless steel sphere into aqueous solution pool.

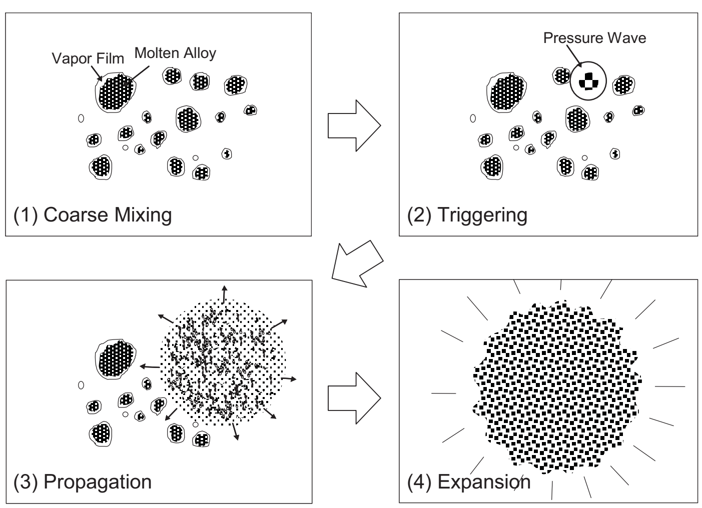

Figure 1. Four conceptual stages of the large-scale vapor explosion

2. EXPERIMENTAL FACILITY

Three different experimental setups were used. Steam explosion accidents have occurred frequently with molten alloy droplets (or jets) into water pool. In the same manner, the first setup is a facility to release the molten alloy droplet into a solution pool. In order to gain more visibility and reproducibility, the second facility (namely VECTOR) was used to release a solution droplet onto a molten alloy surface. The third facility is solid sphere quenching facility to investigate the vapor film stability.

2.1. Molten Alloy Droplet Immersion into Solution Pool

Fig. 2 is a schematic of steam explosion facility. The cylindrical crucible can contain molten alloy at a certain temperature level by electric heaters. Molten alloy droplets were released from the nozzle (i.d. 4 mm) at the bottom of crucible by withdrawing the stopper. The droplets injected into the solution pool. The pool temperature is 10 ^{o}C. The distance from the nozzle to the solution surface is 100 mm. The internal dimensions of the container are 100 mm^{W} × 100 mm^{L} × 200 mm^{H}. The solution depth is 180 mm. Steam explosion behavior was visualized on the basis of the backlighting method.

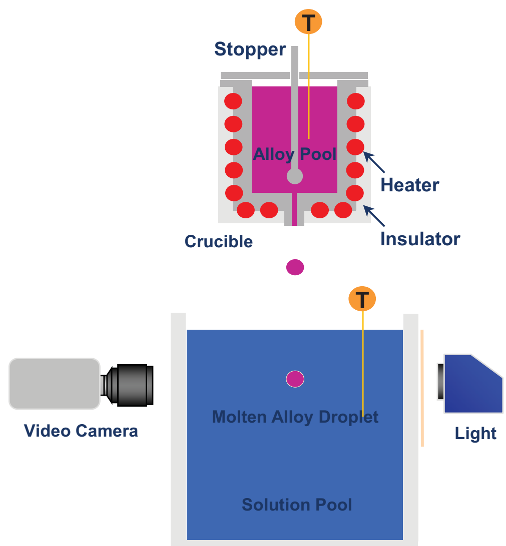

Figure 2. Schematic of vapor explosion facility. Molten droplets immersed into aqueous solution pool, which is the generally-observed configuration as industrial disasters.

2.2. Solution Droplet Impingement onto Molten Alloy Pool

Fig. 3 shows a schematic of the VECTOR (vapor explosion of CRIEPI test with oxide layer removal devices) facility. The test vessel has a volume of 0.48 m^3 and contains a test section, which consists of a droplet nozzle, a molten alloy crucible and devices to remove the oxide layer. The internal dimensions of the crucible are 170 mm^{W} × 150 mm^{L} × 30 mm^{H}. Temperatures were measured at 2-mm-depth intervals in the molten alloy pool, and were found to show scatter within 0.7 K due to natural convection. Type-K thermocouples were immersed into the molten alloy pool to a depth of 8 mm to measure the initial molten alloy temperature.

Degassed water was used as the impingement droplet, which was passed through an ion exchange resin. Solutions were obtained by mixing the water with additives at room temperature using a stirrer for at least twenty hours. A smooth, round solution droplet was released slowly from the droplet nozzle due to the gravity head. Type-K thermocouples were inserted into the nozzle and attached at the nozzle exit to measure the initial droplet temperature. The temperature of water in the nozzle was maintained at a certain level by controlling the flow rate and the temperature of the water in the droplet-temperature-control piping. The droplet diameter is approximately 4.5 mm.

Although the air inside the test vessel was completely replaced by argon, a thin oxide layer was formed on the molten alloy pool surface due to the presence of small amounts of steam and oxygen. Devices to remove the oxide layer were operated in two directions to scrape off the oxide layer preceding each experiment.

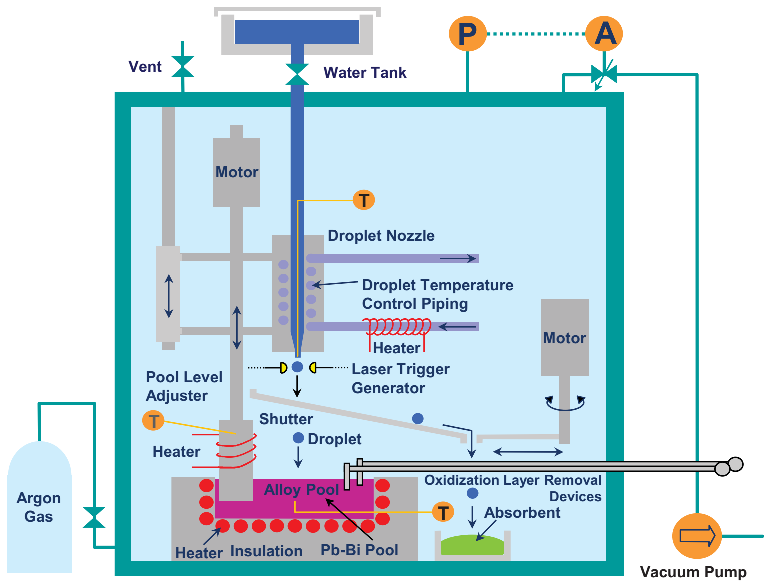

Figure 3. Schematic of VECTOR facility to gain insight of triggering mechanism. An aqueous solution droplet impinges onto an alloy pool. This configuration can attain good visibility and reproducibility.

2.3. Solid Sphere Quenching into Solution Pool

Fig. 4 shows a schematic of the solid sphere quenching facility. A type 304 stainless steel sphere of i.d. 30 mm was heated at 850 °C in the electric furnace, and then immersed into the solution pool. The tip of type-K thermocouples was attached at the bottom sphere and the surface was polished. The programed motor-controlled devise can travel the sheath of the thermocouples vertically with the sphere suspend. The internal dimensions of the container are 100 mm^W × 100 mm^L × 200 mm^H. The solution depth is 170 mm. The collapse of surrounding vapor film was visualized using the backlighting method.

3. RESULTS AND DISCUSSIONS

3.1. Molten Alloy Droplet Immersion into Solution Pool

Fig. 5 shows successive stages of tin droplets injecting into solution pool. The above number is relative elapsed time. Fig. 5 (a) is water pool. Water triggered steam explosion at 6 ms. Fig. 5 (b) is 300 wtppm PEG aqueous solution. The average molecular weight of PEG used was 4×10^6. Throughout the experiment, no steam explosions were observed. Fig. 5 (c) is the same PEG concentration but reduced average molecular weight to 2×10^4. The solution triggered steam explosion at 6 ms in the same manner as for water. The molecular weight must be controlled to suppress the steam explosion.

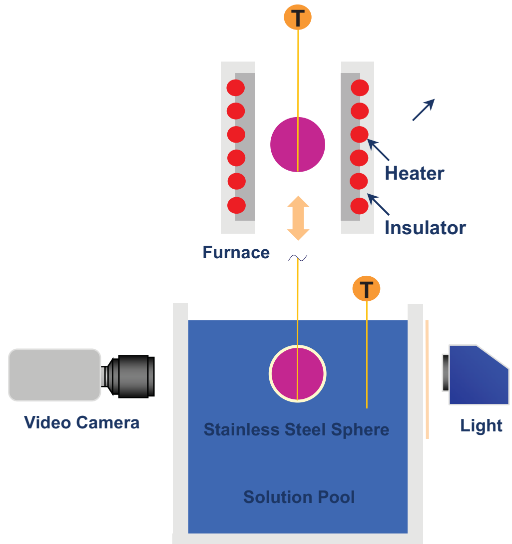

Figure 4. Schematic of quenching facility to investigate vapor film stability.

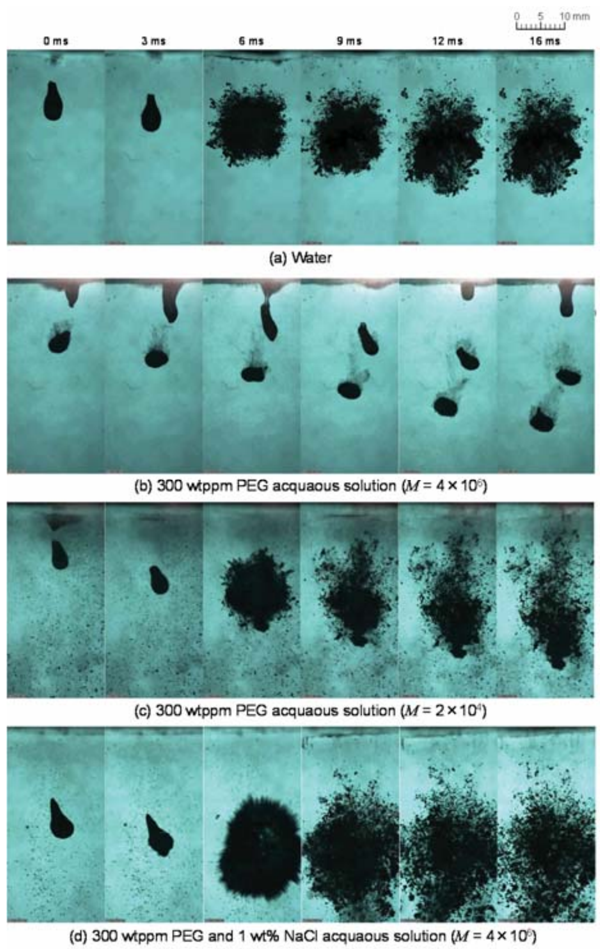

Figure 5. Successive stages of tin droplets injecting into solution pool.

Fig. 5 (d) is the same concentration with coexistence of 1 wt% sodium chloride. The solution triggered steam explosion at 6 ms. Steam explosion accidents occurs in a waste water or coolant pool, which occasionally contains salts. Such salts should be avoided since it may induce steam explosion.

3.2. Solution Pool Solution Droplet Impingement onto Molten Alloy Pool

In order to gain insight the triggering mechanism of solutions, a solution droplet impinged onto the molten lead-bismuth alloy. The author [15] reported that lead-bismuth shows the similar triggering behaviour as tin. Fig. 6 shows an evaporation curve that is usually employed in the droplet impingement system [16][17]. The following five contact modes can be identified in the droplet impingement system [15]. Fig. 7 gives examples of the contact modes.

(1) Wetting State: A hemispherical droplet sits on and touches the molten alloy pool surface. It evaporates from the free surface of the droplet. Boiling is observed if the molten alloy temperature is high.

(2) Transition State: The intermediate condition between the wetting and the spheroidal state. Bouncing motion can be observed due to intermittent contact with the molten alloy pool surface.

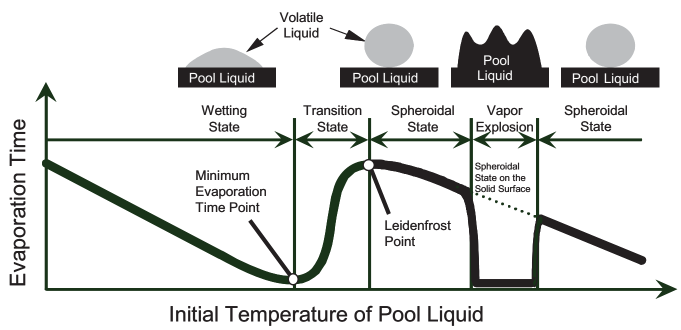

Figure 6. Representative evaporation curve [15]

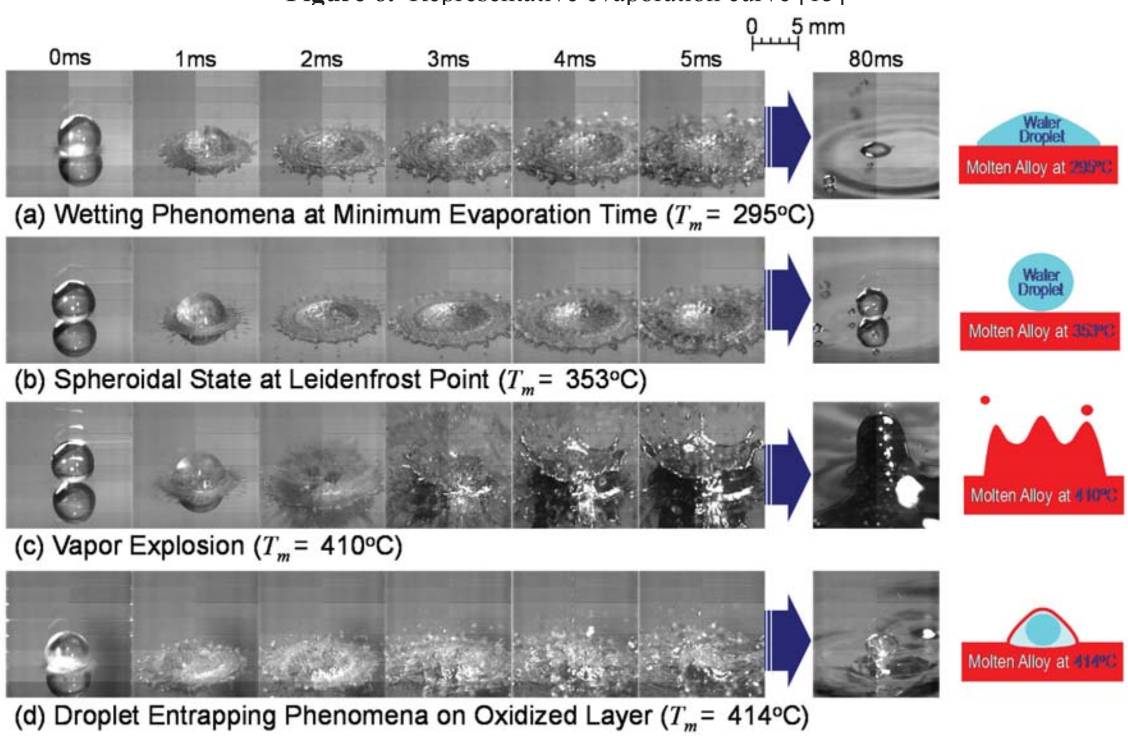

Figure 7. Successive stages of water droplets impinging onto molten lead-bismuth pool [15]

(3) Spheroidal State: A spheroidal droplet floats on the molten alloy pool surface. The evaporation time is long since a vapor film at the interface prevents the droplet from being in contact with the surface.
(4) Steam (vapor) explosion: Deformation of the molten alloy pool surface is induced by strong thermal interaction. Steam explosion can be seen only if the pool material is a liquid.
(5) Droplet Entrapping: Unevaporated droplet is entrapped in a molten alloy dome, unlike steam explosion in which the entire droplet evaporates instantaneously. Droplet entrapping often occurs on an oxide surface, since the oxide layer prevents coherent vaporization and mixing.

Fig. 8 is a contact mode map of PEG concentration and initial molten bismuth-lead pool temperature. Two bold lines indicate the conditions when the contact temperature, T_i, agrees with the homogeneous bubble nucleation temperature (left line), $T_{hn,water}$, and critical temperature of water (right line), T_{cr}. These temperatures refer to the properties of water, not the solution. The homogeneous bubble nucleation temperature was estimated on the basis of semi-theoretical correlation [14]. The contact temperature was calculated on the basis of the conduction theory of two semi-infinite solid bodies in contact with each other.

The average molecular weight of polyethylene glycol, MPEG, was 4×10^6. The temperature region where steam explosion occurred became narrower with increasing concentration. Steam explosion was suppressed by a 200 wppm solution. The viscosity ratio of an aqueous solution to pure water is 1.27 at 20 ºC for a 200 wppm polyethylene glycol solution. The author [10] reported that steam explosion did occur for hydroxyethyl and carboxymethyl cellulose aqueous solutions at this viscosity ratio and much higher than this.

When the temperature of polyethylene glycol solution exceeds a certain point, the solution becomes cloudy due to steep reduction in solubility. This is referred to as a cloudy-point phenomenon. The cloudy-point phenomenon causes a deposition of the solute resulting in a condensed solution or gel. For instance, the viscosity ratio becomes thousands by a 1 wt% polyethylene glycol solution. Thus, steam explosion did not occur, since the vapor film was stabilized and fine-scale mixing was suppressed.

The cloudy point (temperature) is a function of molecular weight. Qualitative dependence on the molecular weight was investigated on the basis of visual observation of the solution in a beaker in an oil bath. The concentration of the polyethylene glycol solution was fixed at 200 wtppm. The cloudy-point phenomenon was observed at 98 ºC for an average molecular weight of 4×10^6. The phenomenon was, however, not observed for average molecular weights of 1×10^3 and 2×10^4, although heating was performed up to 110 ºC. Fig. 9 is the contact mode map in terms of the average molecular weight. The concentration of polyethylene glycol solution was fixed at 200 wtppm. Steam explosion would be suppressed only when the cloudy point is lower than the saturation temperature at a target pressure.

The lower limit of effective concentration may depend on the external force acting on the liquid-liquid interface. The effect was examined experimentally by changing the impingement velocity. Fig. 10 shows a contact mode map in terms of the impingement Weber number. The diameter, the concentration, and average molecular weight are identical at 4.5 mm, 200 wtppm, and 4×10^6, respectively. As investigated previously, the base case is plotted in the figure as a Weber number of 10^6. When the Weber number was 295, the droplet-entrapping phenomenon was observed within the region where steam explosion occurred for pure water as shown in Fig. 8. When the Weber number was 484, steam explosion frequently occurred in the same region. Thus, one should use a larger molecular weight and/or higher concentration when the mixture scale is large and/or when intensive external forces act on the liquid-liquid interface.

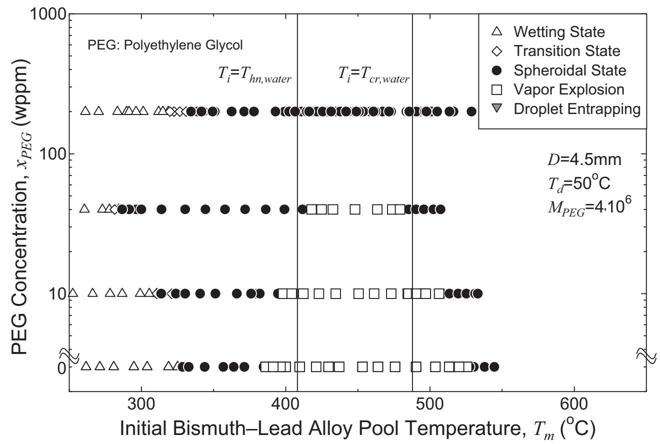

Figure 8. Suppression Effect of Polyethylene Glycol Concentration [15]

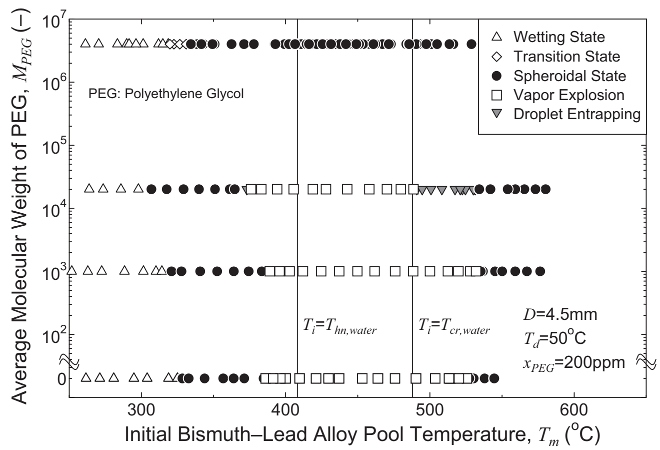

Figure 9. Suppression Effect of Molecular Weight of Polyethylene Glycol [15]

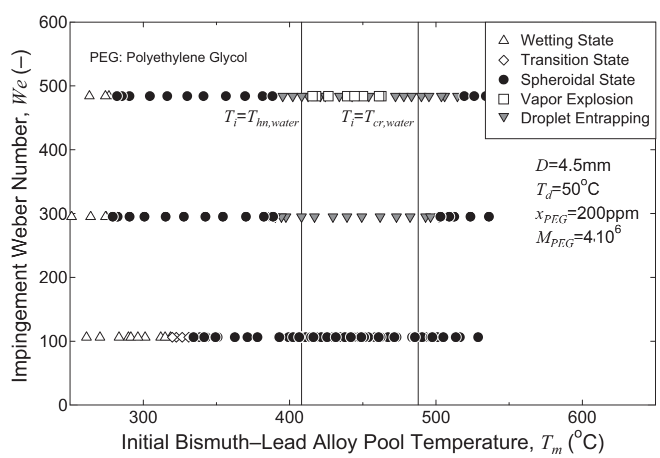

Figure 10. Effect of Impingement Weber Number on Severity of Vapor Explosion [15]

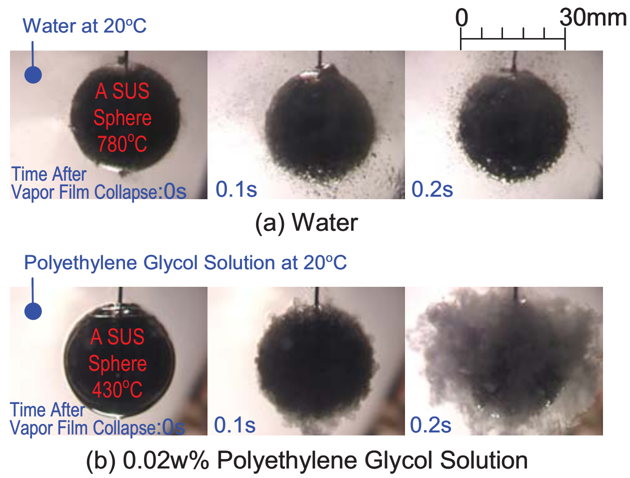

Figure 11. Stabilizing Effect of Polyethylene Glycol Solution on Vapor Film [14]

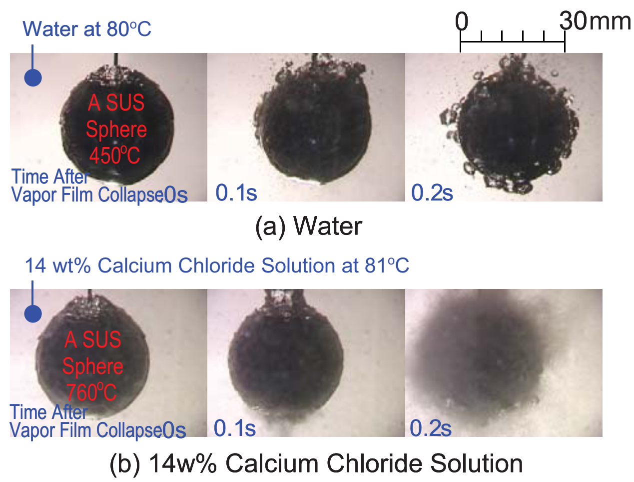

Figure 12. Destabilizing Effect of Calcium Chloride Solution on Vapor Film [14]

3.3. Solid Sphere Quenching into Solution Pool

In order to validate the stabilizing effect of the polyethylene glycol solution, solid sphere quenching experiment was conducted. Fig. 11 shows the successive stages of vapor film collapse in a 200 wtppm polyethylene solution. The average molecular weight of polyethylene glycol was 4×10^6. A type 304 stainless steel sphere at 850 ºC was immersed into a water pool at 20 ºC. The vapor-film collapse, or quench temperature was 780 ºC for the water pool, while it was 430 ºC for a 200 wtppm polyethylene glycol solution. The polyethylene glycol solution has a stabilizing effect on the vapor film by such small concentration. This is because polyethylene glycol precipitates at the vapor-liquid interface due to the cloudy point phenomenon.

The cloudy point (temperature) drops when salt is added into a polymeric aqueous solution. One may think that the addition of salts emphasizes stabilizing effect of polymeric aqueous solution on steam explosion, which exhibits a cloudy point. Fig. 12, a stainless steel sphere at 850 ºC was immersed in the water pool at 80ºC. The quench temperature was 450 ºC for the water pool, while it was 760 ºC for a 14 wt% calcium chloride solution. At the quench point, minute bubbles were dispersed from the surface of the sphere at such low subcooling. The author [14] reported that such salt solution has a destabilizing effect. This may be because a rate of evaporation to condensation mass flux decreases with increasing solute concentration.

As seen in Fig. 5 (d), experimental results indicated that the steam explosion may occur even for a 200 wtppm polyethylene glycol solution by adding sodium chloride. Therefore, one should not expect a suppression effect to occur after the addition salts.

4. CONCLUDING REMARKS

(1) Polyethylene glycol (PEG) is found to suppress steam explosion for tin drops immersed in a water pool by adding 300 wtppm of average molecular weight of 4×10^6.
(2) The cloudy point phenomenon stabilize vapor film by increasing viscosity thousands times near the vapor-liquid interface by precipitated PEG.
(3) The effective concentration became denser when large share and/or external force act on the vapor film.
(4) The molecular weight must be selected for the cloudy point by a certain degree lower than saturation temperature at a target pressure. At atmospheric pressure, molecular weight of 4×10^6 is demonstrated to suppress steam explosion.
(5) Steam explosion may occur in a PEG solution by adding 1wt% of sodium chloride. Such salts act as steam explosion promoter.

ACKNOWLEDGMENTS

The authors are grateful for many constructive suggestions made by emeritus professor Akira Inoue of Tokyo Institute of Technology and emeritus professor Takeo Takashima of Oyama National College of Technology. They thank to Mr. Takeo Yoshioka of Ceres, Inc. for conducting these experiments.

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