Research of mechanisms of interaction of melt and coolant by means of small-sized experiments with solid and liquid metal samples

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Abstract. Several basic fragmentation hypotheses described in the literature for the case of fine-scale spontaneous vapor explosion, when single hot drops are falling in the coolant, are analyzed. It is shown that all of them, depending on the experimental conditions, describe the real physical phenomena. It is highlighted, that the process of fragmentation during the spontaneous vapor explosion is directly connected to the regime of the film boiling collapse of the underheated liquid, which is still underexplored. The experiments on the liquid drops fragmentation are complemented by those with surface boiling at the solid metal samples of semi-spherical form. The connection between the vapor explosion of water and the increase of the surface wettability is confirmed. The video material, indicating the possibility of formation of liquid jets that strike the hot surface during the collapse of the vapor shell, is obtained. The time interval (duration of the contact) between the first contact of the coolant with the hot surface and the explosion boiling of the liquid is experimentally determined. The dependence of the contact duration on the temperature of the overheated wall is determined and its comparison with the theoretical estimations is provided.

1. Introduction

Despite active research on vapor explosions for more than half a century, the detailed behavior of this complex and dangerous phenomena is not fully understood. In particular, in our opinion, the question of possible reason for hot melt fragmentation during its interaction with the low-boiling coolant, still remains open [1]. The fragmentation process plays the main role during the vapor explosion since it itself leads to the sharp increase of the heat-transfer surface and to the explosive increase of the vapor formation. In the standard approach to the behavior of the vapor explosions it is assumed that the intensive fragmentation of the coolant takes place at the three stages connected: firstly, with breakdown of the liquid-metal stream, falling in the cold liquid (so called pre-mixing state, linked with Relay-Taylor instability); secondly, with spontaneous fragmentation of a single drop during the collapse of the surrounding vapor shell; thirdly, with breaking of the multiple number of drops by the shock waves, generated during the sharp increase of the grow and collapse of the vapor objects. It should be noted that while in the first case relatively large drops surrounded by the vapor shell, with the sizes of several millimeters, are produced as a result of the fragmentation, for the second and the third processes the size of the produced fragments can be several micrometers. The latter facilitates the increase of the particle cooling speed and production of the materials with special properties due to the phenomenon of fragmentation.
The ways of fragmentation are usually divided into the hydrodynamic and thermo-hydrodynamic ones that take into account the thermal effects, particularly the coverage of the streams and particles by the vapor shell of the coolant. During the development of the numerical codes of the vapor explosion as a whole, the most difficult stage of its modeling is connected to the propagation of the explosive perturbation over the working area. The base of the description of this process is the model of so-called "physical detonation" (similar to the process of chemical detonation during the combustion) [1]. In this physical model it is assumed that the fragmentation of single drops, providing the energy to the shock wave front, takes place due to considerable gradients of velocity inside its volume, arising under the impact of the shock waves. These waves are produced either due to the external reasons, or due to the explosive fragmentation of the adjacent drops. As it was shown by the comparison with the experimental data, such approach is physically incorrect and for the reliable description of the process the finer thermal physical effects should be taken into consideration.

Such models are mainly based on the results of the simpler low-scale experiments, targeted at the research of fragmentation mechanisms of the single melt drops falling in the coolant (usually, in the cold water). It should be mentioned that the number of proposed hypotheses is quite big and, as it is rather witty noted in [1] corresponds to the number of carried out experiments. Let us focus on the main thermo-hydrodynamical models of the drop fragmentation putting special attention to the conditions of its applicability.

2. Applicability assessment of some thermo-hydrodynamic hypothesis of fragmentation

Among the first works explaining the fragmentation process there were theoretical studies based on the thermo-mechanical effect of ultrafast solidification of the surface layer of the drop during explosive destruction of the vapor shell [2]. This process is followed by the compression of the liquid core of the drop, formation of cracks in its hardened part and fragmentation. The fragmentation is carried out by ejecting liquid metal jets into the surrounding coolant. The occurrence of fragmentation by a similar mechanism is confirmed by the needle shape of fragments, which are obtained in numerous experiments including the ones carried out by our group. However, this hypothesis does not allow us to describe the fast-moving process associated with fine fragmentation of the melt. According to the results of theoretical estimations [3], the drop fragmentation time with a similar fragmentation mechanism is hundreds of microseconds, which is several orders of magnitude higher than the value observed in the experiment (tens to hundreds of microseconds) [4].

From the point of view of the duration of explosive fragmentation process, the so-called cavitation-acoustic hypothesis [2] is the most suitable one. It assumes that when the coolant boils on the surface of a hot liquid metal drop, shock waves are generated due to the growth and collapse of vapor bubbles. These waves propagate in the droplet volume as well, and their internal reflection from the free surface leads to the appearance of a series of rarefaction pulses in the melt. Such impulses of negative pressure can be the reason of the formation of the cavitation cavities inside the drop of the hot coolant and its fragmentation during explosive expansion. The existence of the dissolved gases inside the melt can intensify this process of the drop fragmentation. The main limitation for the application of such hypothesis is associated with the necessity of assuming high-intense pressure pulses, which are required for producing cavitation in the liquid metal. As it is seen from Table 1, where the data about negative pressure, required for the development of the explosive fragmentation process is presented, the value of this quantity for the pure liquid metals exceeds one thousand atmospheres [2]. The gases and impurities dissolved in the melt considerably decrease this value (in absolute figures). However, for the liquid metals this question, due to complexity of the experiments, is practically unexplored. The experimental data of this quantity for mercury described in the literature, as can be seen from the table, have a very large experimental scatter. Therefore, the question of using the cavitation-acoustic hypothesis to explain the fine fragmentation of the melt requires additional investigations. The experimental and calculated data on the fragmentation of water drops [5] indicate the possibility of using (under certain conditions) a similar approach to describe the fragmentation process.
Table 1. Values of negative pressures required for cavitation in pure liquids.

| Material | Melting point, °C | Droplet temp, °C | Cavitation pressure, MPa |
|----------|------------------|------------------|--------------------------|
|          |                  |                  | Theoretical [2] | Experimental |
| Water    | 0                | 20               | -103.9          | -28.1 [2], -140 [6], -7.7 [7] |
| Hg       | 38.9             | 300              | -1190           | -46 [2]          |
| Hg       | 38.9             | 20               | -3000 [8, 9]   | -42.2 [8], -190 [9] |
| Sn       | 232              | 500              | -2450           | -               |
| Pb       | 237              | 500              | -1142           | -               |
| Al       | 660              | 800              | -3500           | -               |

Currently, among the theories of fragmentation of single drops described in the literature, the hypothesis of penetrating jets [10] is predominant. It is based (with slight variations) on the effect of the formation of coolant jets that strike into a hot drop. It is assumed that such jets are formed as a result of the collapse of vapor bubbles near a hot surface. As a result, the coolant penetrates into the volume of the melt, boils sharply and destroys the drop. The undoubted advantages of this hypothesis include the ability to describe the cyclical nature of the fragmentation process. However, as noted by many authors, this hypothesis is, firstly, too complex to be completely reliable, and, secondly, none of the experimenters have observed these penetrating jets. Therefore, a more detailed study of the process of collapse of vapor bubbles near superheated surfaces is necessary.

Another promising modern model of fragmentation is based on the effect of stepwise deformation and breaking of the surface of a liquid metal drop during the formation and explosive growth of vapor bubbles of a coolant on it [11, 12]. This hypothesis describes the cyclical nature of the breaking process. In addition, it allows one to calculate the fine fragmentation time, which is consistent with the experimental value (tens to hundreds of microseconds) [13].

All above-mentioned models to varying degrees depend on the insufficiently studied process of explosive boiling of the coolant on superheated surfaces. It is convenient to simulate such process by contacting water with a solid heated surface. Below some results of experimental studies on this problem are presented.

3. Experimental installation, technique and results

In experiments, the process of vapor film collapse was investigated. A detailed description of the experimental setup is given in [14]. We measured the temperature of water, heated sample and pressure pulses generated in the explosive boiling of a coolant. In addition, using the conductometric technique, the contact parameters of the coolant with an overheated surface were synchronously measured. The process of destruction of the vapor film in synchronization with other measurements was recorded with a video camera (200 fps). The temperature ranges of the sample and water were (150 - 500) °C and (20 - 80) °C, respectively. An important feature of our methodology is the synchronous measurements of the contact and pressure pulse. Such simultaneous measurements allow determining the contact time of the coolant with a hot surface, which is necessary for explosive boiling of a liquid. The time of explosive boiling was determined by the beginning of a sharp increase in pressure pulse. It must also be emphasized that the contact and pressure characteristics were measured in an almost inertialless manner with a sampling frequency of 10^5 meas./s. The purpose of the study is to detail previously discovered physical effects.

Figure 1 shows a sequential series of photographs of a typical explosive descent of a vapor shell near a heated hemisphere. The temperature of the water and the sample is 20°C and 250°C. The sample is made of stainless steel. The time interval between frames is 5 ms. The following points should be highlighted. Firstly, the linear expansion rate of the vapor cavity is several meters per second. Secondly, even at such short times (milliseconds), vortex structures have time to form around the vapor cavities, which deform the surface of the bubble. Thirdly, in the process of formation and deformation of the steam bubble, a strong steam jet is formed, directed first to the wall, and then from the heated surface to the coolant.
Figure 1. Photographs of the explosive destruction of the vapor shell. The time interval between frames is 5 ms.

We have previously shown [14] that the collapse of the vapor coolant shell around a heated hemispherical surface generates pressure pulses of intensity ~ MPa and a duration of several tens of microseconds. Such pressure pulses may well trigger spontaneous steam explosions. In this work, it is also noted that explosive boiling in experiments with stainless steel samples is observed only after several preliminary immersions in water, accompanied by quiet (not explosive) boiling on their surfaces. That is, the explosive collapse of the vapor film was observed only on oxidized surfaces. In order to elucidate the causes of this effect, additional experiments were carried out to study the influence of the surface state and, primarily, wetting, on the process of explosive boiling. Analysis of surfaces using optical microscopes (optical amplification ×500 and higher) has not revealed any significant differences between fresh and repeatedly involved (“explosive”) surfaces. In part, this can be explained by the transparency of metallic oxides for optical radiation. However, experiments to study the wetting of these surfaces with water droplets have revealed a significant difference. As can be seen from Figure 2, reused (oxidized) surfaces are much better wetted by water (wetting angle \( \theta \approx 45 ^\circ \)) than specially polished “fresh” surfaces (wetting angle \( \theta = 83 ^\circ \)).

Figure 2. Photos of the water drop spreading on a steel surface: (a) – “fresh” polished surface without oxides; (b) – surface reused in experiments (coated with an oxide film).

It is known [1] that wetting has a significant effect on coolant vaporization near a heated wall. The intensity of this process (the rate of formation of vapor nuclei — \( J \)) can be estimated from the modified Dering – Volmer relation [1]:

\[
J \sim \exp \left( \frac{-f(\theta)W}{kT} \right)
\]  

(1)

where \( W \) is the work spent on the formation of a vapor bubble of critical size; \( k \) is the Boltzmann constant; \( T \) is the temperature; \( f(\theta) = (2 + 3 \cos \theta - \cos^3 \theta)/4 \) is the function that takes into account the
wetting of the hot wall coolant. Taking into account the values of the contact angle $\theta$ obtained from experiments, we assume that for the “fresh” and oxidized surfaces $f(\theta) \approx 0.5$ and 1, respectively. Therefore, based on relation (1) for a fixed value of the surface temperature, the rate of nucleation on the “fresh” surface should be $\approx e^{0.5}$ times higher, which contradicts the experimental results. Therefore, the question of possible causes of explosive boiling only on oxidized surfaces requires further comprehensive experimental study.

The time between the primary contact and boiling of the liquid is an important parameter in the process of interaction of the coolant with a heated surface. In addition to theories of steam explosion, this value is important for constructing theories of jet cooling and the transition boiling regime. As mentioned above, the developed technique, based on the simultaneous measurement of pressure and electrical contact, allows us to determine this time quite reliably. The dependence of this quantity obtained in experiments on the temperature difference between the working sample and the saturation $T_s$ is shown in Figure 3. The following important points should be highlighted. The time of heating the liquid in this temperature range of measurements is relatively small (units - tens of microseconds). This circumstance explains, in particular, the available scatter of experimental data at short delay times. At the same time, the graph clearly shows a tendency to increase the warm-up time in the temperature range (130–150)$^\circ$C of surface overheating. The obtained result correlates with the data on the intensity of pressure pulses obtained earlier [14] and indicates that a longer heating time $t_{del}$ corresponds to more intense boiling of the coolant. The nonmonotonic dependence of the coolant heating time on the surface temperature $T_w$ observed in the experiments differs (Figure 3) from the relation proposed in [15], in which this value increases monotonically with decreasing wall temperature according to the law $t_{del} \sim (T_w-T_s)^{-8/3}$. Clarification of the reasons for this discrepancy requires further experimental and computational research.

**Figure 3.** The dependence of time between the primary contact of the liquid and its boiling in contact with a hot surface on the overheating temperature of the hemisphere surface.

1. $t_{del} \sim (T_w-T_s)^{-8/3}$;
2. $t_{del} \sim (T_w-T_s)$;
3. experiment.

**Conclusions**

The analysis of the fragmentation hypotheses indicates that the available theoretical studies describe different types (coarse and fine fragmentation) of melt crushing in a small-scale steam explosion. One of the most attractive theories is the cavitation-acoustic hypothesis of fine fragmentation, which gives the potential to control the crushing process by controlling the amount of dissolved gases and impurities present in the melt. However, the question of the occurrence of cavitation phenomena in liquid metals has not been fully studied, and the available data are contradictory. In order to increase the reliability, all the fragmentation hypotheses discussed above must be supplemented with new information about the process of interaction of the coolant with a heated surface. The obtained video
materials confirm the assumption of the formation of steam jets, accompanying the process of collapse of steam shells and also directed to a heated surface. The data on the intensification of explosive boiling on wetted surfaces are in some contradiction with traditional ideas about this process and require a more detailed analysis.

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