A thermomechanical model for the fragmentation of a liquid metal droplet cooled by water

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Abstract. A thermomechanical aspect of the fragmentation of a liquid metal droplet, solidified as it falls into cold water, is considered in the presented model. The formation of a solid phase in the form of continuous, fluid-tight and relatively rigid casting skin results in a pressure decrease inside the droplet due to the difference between liquid and solid metal density. Because of the high compression modulus of the melt, the pressure in the droplet becomes negative when the thickness of the solid skin achieves several microns. The tensile stress in the melt results in the deformation of the casting skin or the melt’s continuity violation in the form of a shrinkage pore. The rupture of the deformed solid crust results in the penetration of steam jets into the liquid part of the drop. Due to the difference in pressure in the surrounding steam and in the droplet, the casting skin is crushed and the melt is blown out. Both scenarios contribute to the hydrodynamic destruction of the droplet. The suggested thermo mechanical model gives a qualitative explanation for experimental data. In the experimental part of the work, droplets of molten Sn were solidified in water. The solidified pieces of the droplets usually include deformed, thin-walled shells and dispersed particles. On a qualitative level the composition and shape of the solid fragments can be explained within the bounds of the suggested thermo mechanical model.

1. Introduction

In the emergence of the prerequisites for a steam explosion, the fragmentation of drops of a metallic melt plays an important role. Over several decades of research into this process, two possible mechanisms of fragmentation have developed.

In the model of the hydromechanical fragmentation of droplets, the formation of a vapor layer between the surface of the melt and the surrounding water is considered. A sharp increase in pressure in the vapor layer, caused by intensive evaporation of water, facilitates the penetration of steam into the melt. Steam jets deform the "vapor-melt" interface, penetrate into the body of the drop and break it into fragments. The hydromechanical model considers the interaction of two liquids, one of which boils and evaporates, which leads to the fragmentation of the second liquid (metallic melt) [1].

In the thermomechanical model of fragmentation, solidification of the melt plays an important role. Within the framework of this model, it is assumed that the solid crust formed on the drop surface undergoes thermal contraction. Since the melt is an incompressible fluid, the formation of a crust leads to the occurrence of tensile stresses in the solid phase and to an increase in pressure in the melt.
Stresses in the crust lead to its rupture and the melt, under pressure, is ejected into the surrounding liquid [2].

The present study is devoted to another aspect of the thermomechanical model of fragmentation, which has not been previously considered. It is shown that the pressure inside the solidifying droplet of the melt falls due to the difference in density between the solid and liquid phases. The results obtained make it possible to evaluate some of the known experimental results by a new approach and open up new possibilities for their interpretation.

2. Melt droplet solidification model
In the process of solidification, two phases coexist in the volume of the drop: a melt and a solidified metal. Since the density of the solid phase, as a rule, is greater than the density of the melt, the solidification of the melt is accompanied by a so-called shrinkage. If the melt communicates with the environment, shrinkage results in the movement of the melt-environment interface. In the absence of contact with the environment, shrinkage leads to a pressure drop in the melt and the creation of internal interfaces i.e. macro and micropores.

For the volume fractions of phases in the elementary volume of the solidifying metal, the following relation holds:

\[ f_1 + f_s + \Omega = 1 \]  

(1)

where \( \Omega \) is the shrinkage volume fraction; \( f \) is the volume fraction of the phase; \( l, s \) - refer to the liquid and solid phases, respectively.

The mass balance in the solidification of the melt obeys the equation of continuity. As the droplet is an isolated volume of a melt, the continuity equation has the form:

\[ \partial (\rho f_1 + \rho_s f_s) / \partial \tau = 0 \]  

(2)

where \( \rho \) is the density; \( \tau \) is the time. From (2), using (1), one obtains

\[ \Omega = \beta (1 - f_1) = \beta \rho_s \]  

(3)

where \( \beta = (\rho_s - \rho_l) / \rho_s \) is the metal shrinkage factor, \( \beta = \beta / (1 + \beta) \).

3. Pressure in the solidifying droplet
After the solidification of a part of the melt, a melt and a solid phase are present in the isolated volume of the drop. Since the volume of the solid phase is less than the initial volume of the melt, the expression (1) is valid, in which \( \Omega \) is the volume fraction of the droplet, which can be occupied by the shrinkage pore.

We will proceed from the fact that solidification starts on the surface of the drop. A solid and impervious crust of the solid phase forms on the surface of the droplet. If the formation of the pore has not yet occurred, the melt occupies the entire space of the droplet, unoccupied by a solid phase. It follows that the melt is in a stretched state. The pressure in the droplet is given by the expression:

\[ P = P_a - E \Omega (f_1 + \Omega) \]  

(4)

where \( P_a \) is the external pressure; \( E \) is the compression modulus, \( \varepsilon = \Omega (1 + f_1) \) is the volumetric deformation of the melt. The second term (4) determines the pressure drop due to solidification and shrinkage of the metal [3].

As follows from (4), the pressure in droplets of different diameters is described by a single curve; however, the thickness of the solid crust depends on the fraction of the solid phase and the size of the droplet. Calculations performed for drops with a diameter of 2, 5 and 10 mm at \( E = 2000 \) MPa and \( \beta = 0.05 \) show that the formation of a solid crust of 10-50 \( \mu m \) in thickness (depending on the droplet size) can lead to a drop in pressure of up to -3 MPa. In principle, this value does not contradict the
known experimental data on the measurement of negative pressure in liquids, for example, given in [4, 5, 6]. However, there are two reasons why such low pressure can be unattainable. The first of these is the deformation of the hard crust, which compensates for the solidification shrinkage. This circumstance can be taken into account by a reduction of the compression modulus $E$. Therefore, in what follows we shall speak of an effective compression modulus that takes into account the deformation of a solid crust. The second reason is that a pressure drop below the melt's tensile strength can lead to the creation of a new interface, i.e. to the nucleation of a shrinkage pore.

4. Nucleation of a stable pore
Since certain work to create the melt-pore interface is required, the pressure in the melt at the time of shrinkage pore nucleation is a negative value. Nucleation of a shrinkage pore occurs when the pressure in the melt drops below a critical value $P_{cav}$. We will assume that the pore has a spherical shape. The condition for pore nucleation in a solidifying droplet can be written as

$$P_a - E \Omega^* (\Omega^* + f_1^*) = P_{cav}$$

(5)

where $f_1^*$, $\Omega^*$ are the volume fraction of the liquid phase and the shrinkage defect at the moment of pore nucleation, respectively.

The fraction of the liquid phase at the moment of pore nucleation is obtained from (5) and (3):

$$f_1^* = \beta \left[ \beta + (P_a - P_{cav}) (E - (P_a - P_{cav})) \right]^{-1}$$

$$\Omega^* = \beta \left[ \beta E (P_a - P_{cav}) + 1 \right]^{-1}$$

(6)

(7)

The maximum pore volume cannot exceed the volume of the shrinkage defect $\Omega^* V$, therefore the pore radius must obey the condition $R_p < (\Omega^*)^{1/3} R$, where $R$ is the droplet radius. On the other hand, the radius of the pore must be large enough so that the surface tension will not be able to collapse the pore, i.e. $R_p > -2\sigma P_{cav}^{-1}$. From this, one obtains the condition for the existence and growth of the pore:

$$R > -2\sigma (\Omega^*)^{-1/3} P_{cav}^{-1}$$

where $\sigma$ is the surface tension.

Figure 1. The boundary of the range of parameters for the possible creation of pores as a function of the droplet diameter and the compression modulus at $\sigma = 0.8 \text{ n/m}$. 1. - $E = 2000 \text{ MPa}$; 2. - $E = 200 \text{ MPa}$.

Figure 1 shows the $P_{cav}$ curves calculated from (5) dependent on the droplet diameter, for two values of the melt compression modulus of 200 and 2000 MPa. The range of parameters at which pore formation is possible lies below these curves. Above these curves the pore can not exist, since it is not
capable of overcoming the surface tension. In addition, to create a pore, the pressure in the melt must be below the tensile strength $P_b$. The boundary $P = P_b$ is shown in figure 1 in dashed lines.

The smaller the droplet diameter, the lower the pressure in the melt, at which a shrinkage volume $\Omega^*$ sufficient to create the pore capable of overcoming the surface tension will be accumulated. In the case of large droplets ($D > 1$ mm), pore nucleation pressure is equal to $P_b$. Therefore, in such droplets a pore occurs, when $P \leq P_b$. In the pressure range $P_b < P < P_{cav}$ the shrinkage pore can exist, but can not arise. In the case of small droplets ($D < 1$ mm), the pressure of the appearance of a stable pore $P_{cav}$ is lower than the tensile strength $P_b$. In this case a stable pore can nucleate and exist only if $P < P_{cav}$. Therefore, the critical pressure of pore formation is not a physical property of the melt, but a characteristic of the solidification process: $P_{cav} = \min[P_{cav}, P_b]$.

5. Condition for the existence of a stable pore
After pore nucleation, a level of tensile stress in the melt is defined by curvature of the interface between the pore and the melt. At homogeneous nucleation a pore apparently has a shape close to a sphere. The equilibrium equation for a spherical pore can be written as

$$P_a - E\left(\Omega^* - f_p\right)\left(\Omega^* - f_p + f_p^*\right) = -2\sigma/R_p$$

(8)

where $R_p$ is the pore radius; $f_p = (R_p/R)^3$ is the pore volume fraction.

In the equations (5), (8), the compression modulus $E$ and the pore nucleation pressure determine the shrinkage volume $\Omega^*$, which can be converted into a pore. With a small compression modulus, the pressure in the droplet drops more slowly and the pressure $P_{cav}$ necessary to nucleate a pore is reached at a larger shrinkage volume $\Omega^*$.

A change in the pressure in droplets with a diameter of 1 mm and 3 mm at $E = 2000$ MPa, $\sigma = 0.8$ n/m and $P_b = -0.1$ MPa calculated from (5) and (8) is shown in figure 2.

![Figure 2. Calculated change in pressure in solidifying droplets with a diameter of 1 mm and 3 mm at $E=2000$ MPa and $\sigma = 0.8$ n/m.](image)

|    |    |
|----|----|
| 1  | D = 1 mm; 2 - D = 3 mm. |

The initial pressure in the melt is equal to atmospheric pressure. Due to the large compression modulus, the pressure in the droplets drops very rapidly with the appearance of a solid phase. As can be seen from figure 1, the nucleation of pores in the droplets with a diameter of 1 and 3 mm occurs with a decrease in pressure below $P_b$. The nucleation of the pore leads to an increase in the pressure in the droplet up to -0.04 MPa (for the droplet of 1 mm diameter). Subsequent solidification of the melt leads to the growth of the pore and an increase in pressure in the droplet. However, the pressure in the droplet remains negative all the time, which is necessary to compensate for the surface tension, which tends to collapse the pore. In figure 2 bold arrows show the solidification path.
The curves of the solid crust thickness for droplets with a diameter of 1, 3 and 6 mm depending on the effective compression modulus of the melt are shown in figure 4. In the case of a rigid crust of solid metal \((E = 2000 \text{ MPa})\), the pressure in the droplet decreases so rapidly that even at a crust thickness of less than 1 μm, a pore is created. If the crust is not rigid enough, the pressure does not decrease so quickly, since the tensile stresses arising in the melt are partially compensated for by the deformation of the crust. Decrease in the rate of pressure drop leads to nucleation of the pore at a slightly thicker crust.

Figure 3. The thickness of the solid shell at the time of nucleation of a stable pore as a function of the compression modulus. 
1 - \(D = 1 \text{ mm}\); 2. - \(D = 3 \text{ mm}\); 3. - \(D = 6 \text{ mm}\).

Figure 4. Photographs of different types of fragments of the droplet after vapor film detachment.

6. Comparison with the experiment
The fragmentation of hot droplets immersed in cold water has been investigated in many works (see, for example, publications [7 - 11]). In our experiments on this subject, a droplet of liquid metal (material: tin, ball bearing steel) was either suspended by the original method at the end of the stainless steel tube-heater, or placed on a ceramic substrate and heated by an inductor.
The results of the experiments carried out using various measuring probes and video cameras confirm the assumption of a variety of mechanisms of droplet destruction. Figure 4 shows typical shapes of fragments of the droplets of tin formed as a result of various mechanisms of crushing. It was found that fine droplet fragmentation (Fig. 4a) occurs in the case of maximum pressure pulsation and a sound signal. In this case, the dimensions of the frozen fragments after crushing a droplet with a diameter of ~ 4 mm vary from less than 1 μm to 1000 μm. If the interaction was relatively weak and probably proceeded in a different way, the solidifying droplet either did not change its shape, or its body was permeated with pores and caverns (figure 4b). In some cases, cooling of the droplet was accompanied by the formation of frozen liquid jets, emerging from a droplet (figure 4c) and air cavities in the volume of the melt (figure 4d). The scheme of the development of processes during the fall of hot tin droplets into cold water observed in [9] and in the experiments carried out in the framework of this paper can be fully interpreted as the collapse of a solidifying droplet due to the loss of stability of the solid crust.

7. Conclusion
The appearance of a solid phase in the form of a solid, impermeable and relatively rigid crust on the surface of the drop leads to a drop in pressure inside the drop due to the difference in the density of the solid and liquid phases. Because of the large value of the compression modulus of the melt, the pressure in the droplet reaches negative values at a thickness of a solid crust of several microns. A thin and, apparently, very frail crust of the solid phase is under the influence of a significant difference in pressures-increased pressure in the steam shell at the outer surface and negative pressure in the solidifying melt. The pressure drop in a droplet can be larger then . The rate of pressure drop is so high that the process can be considered as a hydraulic shock initiating the destruction of the solid crust.

Since the real droplet is not an ideal sphere, the result of the force action will depend very much on its shape. An important role in the fragmentation of the droplet is played by the angle of incidence of the droplet and the uneven thickness of the vapor film. In addition, it should be remembered that decrease in pressure in the droplet is possible only with full tightness of the hard crust. Otherwise, the shrinkage of the solidifying metal will lead not to a drop in pressure, but to the movement of the free surface of the melt. Loss of stability and crushing of the hard crust can lead to two main scenarios of fragmentation of the droplet: 1) - extrusion of melt jets into the environment and 2) - penetration of steam and cold water into the liquid metal inside the drop.

8. References
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