

WHY DOES MOLTEN ALUMINUM EXPLODE AT UNDERWATER OR WET SURFACES?^a

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Molten Al sometimes explodes when it inadvertently contacts a wet or underwater solid surface. With laboratory experiments, we investigated these initiations at seventeen different submerged surfaces. Surfaces nonwetable by liquid water were inert, while water-wettable surfaces initiated explosive molten Al-water interactions. We hypothesize that the water-wetted surfaces provide a thin layer of liquid water beneath the molten Al that can heat rapidly during transient liquid-liquid contact between melt and water. Because the surfaces of the wettable solid and the molten metal are both presumed to be poor nucleators of bubbles, substantial superheating of the water layer may occur. The resultant vaporization may be explosive, causing local fragmentation of the melt and pressure disturbances that initiate steam explosions elsewhere where melt and water are adjacent. Nonwetted surfaces, on the other hand, are essentially dry beneath the melt and produce little assistance to the explosive interaction.

kg quantities of molten metal were poured into water-filled containers. The inner bottom surfaces of these containers were treated in various ways. (Analogous experiments were performed by other researchers with molten copper poured identically into similar containers (7,8).)

Especially violent explosions occurred at certain surfaces, for example, rusted steel, gypsum, surfaces coated with lime, aluminum hydroxide sludge, iron hydroxide sludge; concrete surfaces were also active (3). Other surfaces were inert toward explosions. These included bare metals (steel and aluminum), surfaces that had been coated with organic materials, and, in some cases, normally active surfaces that had been exposed to a "swipe of oil" (9).

The activity of the explosive surfaces was attributed by Long (3) and Hess and Brondyke (4) to the vaporization of water "entrapped" beneath the melt. In neither of these papers, however, was there an explanation for why certain surfaces trap water while others do not. Also, no guidelines were set down for predicting whether a surface that had not been tested before would be active or not. Moreover, the field-scale experiments inherently had a degree of uncertainty that prevented firm conclusions about the action of the surfaces on these explosive interactions.

In this work, we have elected to perform laboratory-scale experiments because of their safety, rapid turnaround times, low cost and inherent ability to control parameters more carefully than in the field. We have performed a series of experiments with seventeen different surfaces to investigate the mechanism of explosions at underwater surfaces. We have combined our experimental results with those of the earlier industry experimenters and numerous literature references to provide an hypothesis for the initiation of explosive melt-water interactions at certain wet or underwater surfaces but not at others. Our proposed explanation is based on the ability of wettable solid surfaces to maintain a thin layer of liquid water beneath the molten aluminum that nonwetable surfaces cannot.

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INTRODUCTION

Molten aluminum sometimes explodes when it inadvertently contacts a wet or underwater surface, for example, in the direct chill casting process (1). These explosive interactions often are hazardous to nearby personnel and can cause extensive plant damage (2).

The aluminum industry has studied the causes and prevention of these explosive interactions for a number of years (3-6). These studies have been conducted primarily with field-scale experiments in which 5 to 25

EXPERIMENTAL

As shown in Figure 1, we used a tiltable, electrically heated furnace with a graphite crucible to melt approximately 12 g samples of 6061 aluminum alloy (1.0 Mg, 0.6 Si, 0.25 Cu, 0.25 Cr, balance Al (all weight percent)) in air. When the furnace was tilted, the mass of molten metal at ~800°C fell quickly into a water-filled cylindrical Plexiglas chamber 210 mm ID and 286 mm deep. About 75 mm below the surface of the water, we positioned a series of concave dishes 10-20 cm in diameter with ~18 cm radii of curvature, made of different materials. The surfaces of some of the dishes were treated to alter their wettability by liquid water. About 100 mm to one side of the dish, we positioned a closely spaced pair of electrodes through which we could pass a discharge from a 7 μF capacitor charged to 3 kV to produce a triggering pressure pulse in the water.

With side experiments, we showed that a transient pulse with a peak pressure of at least 5 MPa (measured at the melt with a PCB Model 138 A 01 tourmaline underwater blast transducer with digital oscilloscope

recording) was required to initiate a steam explosion in a molten 6061 aluminum globule falling freely through the water. In this work, we reduced the pulse height below the threshold to ~2 MPa to start the interaction at the surfaces. This pulse simulated the mechanical transients that might occur randomly in an accident situation where melt poured into water. After the molten metal was poured onto the surface, it was allowed to equilibrate quiescently underwater for ~0.8 s before the spark discharge was fired. If the surface did not assist the explosion, only uneventful freezing would occur; a single piece of metal with several bubble-like voids would be retrieved afterward. If the surface assisted the explosion, however, it would cause a vigorous interaction accompanied by the growth and collapse of one or more steam bubbles. The metal retrieved afterward would be fragmented extensively. Without the trigger pulse only dense frozen shapes were retrieved, regardless of the characteristics of the surface.

Our active diagnostics included a standard VHS video camera/recorder combination and high-speed photography with a Redlake 16 mm Hycam camera operating at ~5000 fps. Other diagnostics included examination

APPARATUS

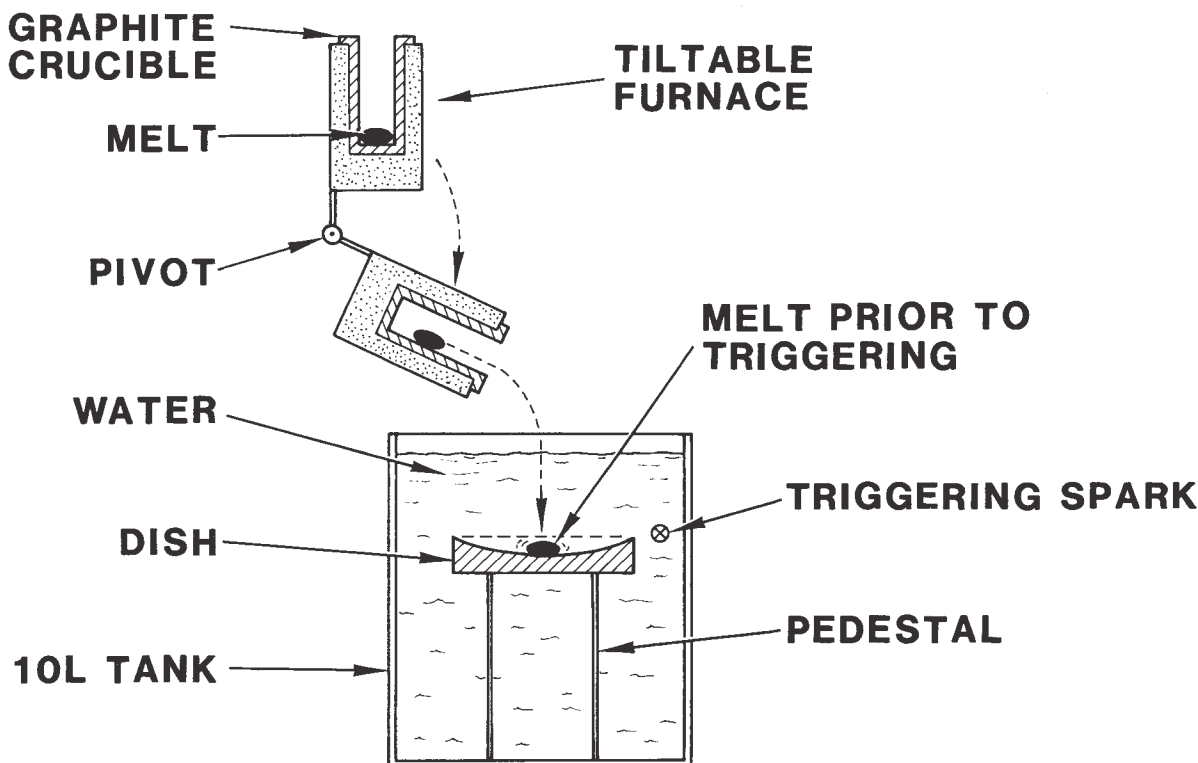


Figure 1. Schematic Diagram of the Experimental System Used to Study the Ability of an Underwater Surface to Assist a Melt-Water Explosion.

of the debris retrieved afterward and microscopic comparison of the various surfaces before and after the interactions.

We studied the wettability of the surfaces at room temperature in air by applying individual $23 \mu\text{L}$ drops of water from an automatic pipette. The wetting was estimated by the equilibrium area assumed by

the drop on the surface as measured with a photographic microscope. Typical photographs of the drops at equilibrium on a poorly and a strongly wetting surface are shown in Figure 2. Also, with drops on several of the surfaces, we simultaneously measured both their contact angles with an optical goniometer and their equilibrium areas with the photographic technique.

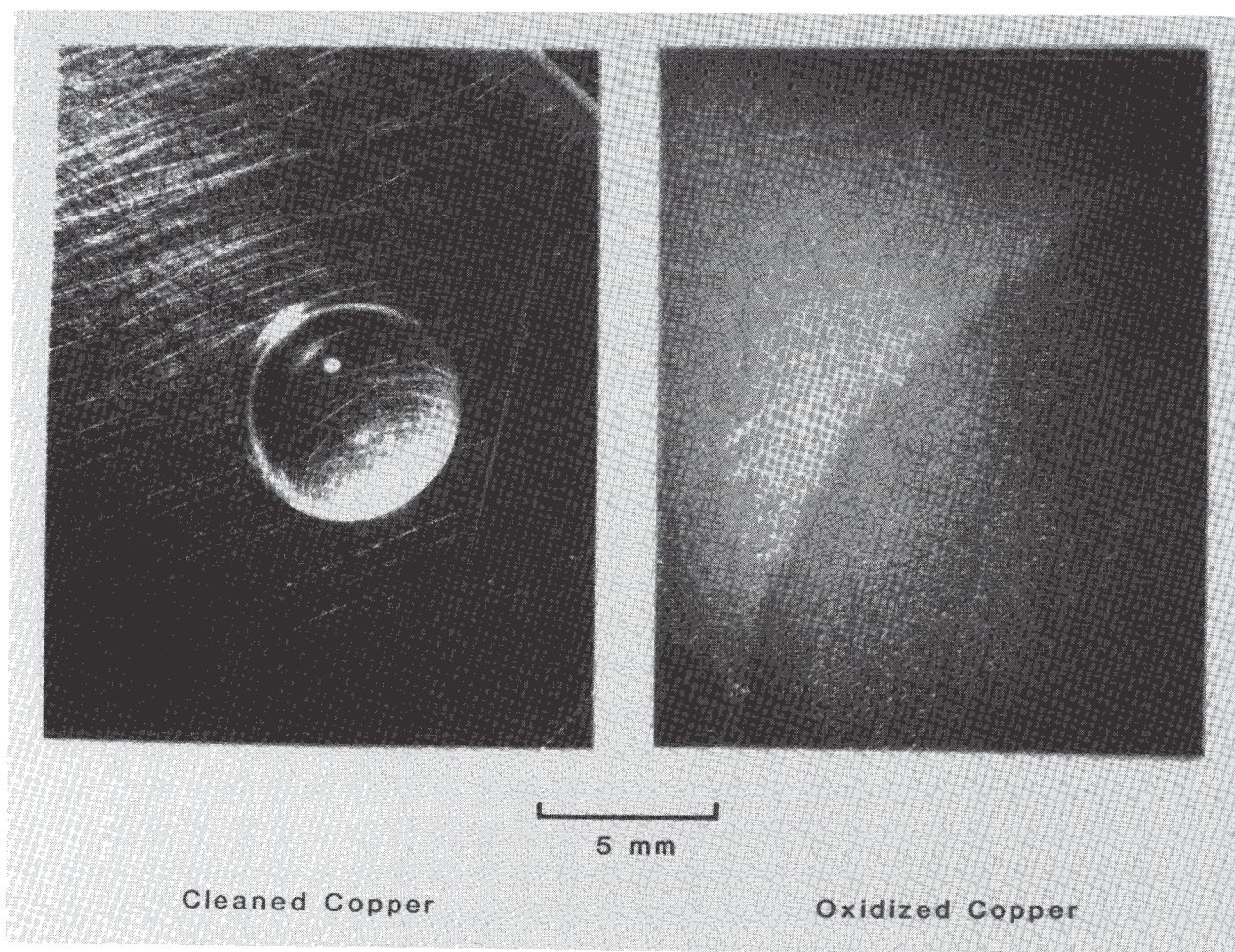


Figure 2. At Left, Photograph of a $23\mu\text{L}$ Drop of Water at Rest on a Copper Plate That Had Been Cleaned by Abrasion With Crocus Cloth; At Right, Photograph of a $23\mu\text{L}$ Water Drop at Rest on a Copper Plate That Had First Been Cleaned With Crocus Cloth and Then Oxidized in Air for 15 Minutes on a Hot Plate at 400°C to 425°C .

RESULTS

When individual globules of molten aluminum were poured onto the various submerged surfaces without the pressure transient, explosions never occurred. Instead, shortly after the globule came to rest on the surface, it would jump about 1 cm to one side. We attribute this to the sudden production of steam by vaporization of liquid water beneath or adjacent to the melt at the onset of transition boiling. Also, in the bottom of each globule that froze in this

manner, there always was an upward indentation about 10 mm across and 5 mm high.

The parameter we selected to study the influence of surface wettability on the initiation of molten aluminum-water interactions quantitatively was the sum of the maximum volumes of the steam bubbles produced above the surface in an interaction estimated from our high speed films. Outlines of typical images produced at wettable and nonwettable surfaces are shown in Figure 3.

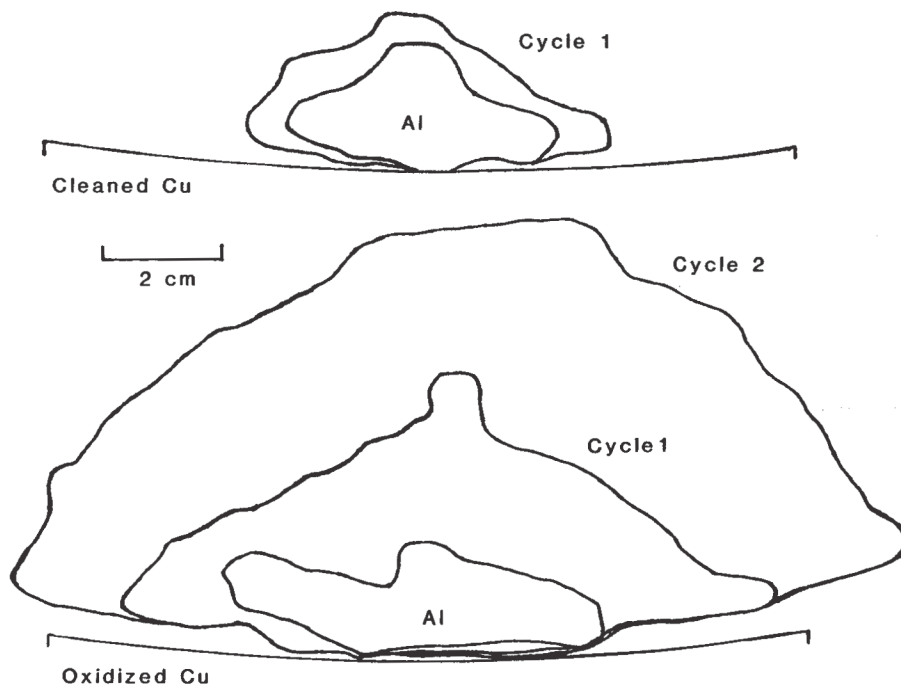


Figure 3. Outlines of Maximum Steam Bubbles Produced at (above) a Cleaned Copper Plate and (below) an Air-Oxidized Copper Plate by Melt-Water Interactions. As in most vigorous interactions, steam was generated in two bubble growth cycles (below); only one cycle was formed in the weak interaction (above).

TABLE 1. Sum of Maximum Steam Bubble Volumes Produced During Explosive Molten Aluminum-Water Interactions at Submerged Surfaces Tabulated with the Area Assumed by 23 μ L Water Drops on These Surfaces in Air

No.	Surface	Sum of Maximum Bubble Volumes (cm ³) ^a	Area of Drop (mm ²)
1	Grafoil	60	15
2	Gypsum (Hydrocal ^b), oiled	7.5	16
3	Graphite	2.6	20
4	Carbon ^c on Pyrex	48	22
5	Copper plate, 4mm-thick, polished	48	24
6	Copper ^c on Pyrex	104, 195	36
7	Tarset on steel	38	38
8	Aluminum, polished	55, 58	71
9	Steel, grit blasted	69, 4.0	76
10	Glass	50	76
11	Al, anodized, sealed	72, 221	118
12	Steel, rusted	1110	158
13	Gypsum (Supercal ^b)	1020	158
14	Gypsum (Hydrocal ^b)	235, 1430	161
15	Al, anodized, unsealed	69, 384	161
16	Copper, air oxidized	619	174
17	Copper ^c on Pyrex, air-oxidized	441	>1256 ^d

^aBecause we used a cylindrical vessel for our experiments, there was optical distortion inherent in our films. We measured a horizontal linear contraction that ranged from zero at the centerline to 10% at the outer edge of the vessel. When estimating the bubble volumes, numerous horizontal widths of the images were squared, which could produce a maximum uncertainty of 21% for the largest bubbles at their maximum width (the bubble from only one explosion approached the maximum width of the chamber). The underestimation for smaller bubbles would be considerably less, probably 5-10%. Although the volumes could be corrected for the optical distortion, the trends shown in Table 1 and Figures 4 and 5 and the conclusions drawn from them would not be altered, however.

^bTrade name.

^cVapor-deposited.

^dDrop evaporated before spreading was complete.

In order to vary the wettability of the triggering surfaces, we used two approaches: (a) to change from material to material, and, in several cases, changing the nature of the surface drastically by a one time treatment, and (b) progressively changing the wettability of one material by varying the extent of a single surface treatment.

Changing Materials

We studied the explosive interactions at the seventeen surfaces listed in Table 1. The sums of the maximum steam bubble volumes produced above each surface are presented in the third column of this table. These volumes are compared with the average areas assumed by 23 μ L drops placed on these surfaces, presented in the fourth column. The areas were calculated from the average measured diameters of three or more individual drops at room temperature in air after they had spread to their equilibrium diameters.

In Figure 4 we have plotted the sums of the maximum bubble volumes against the equilibrium areas assumed by the drops of water on the various surfaces presented in the third and fourth columns of Table 1. The surfaces at which the various interactions occurred are identified in Figure 4 by the numbers shown in the first column of Table 1. The contact angles for drops on several of the surfaces are shown along the top of Figure 4.

Progressively Changing the Wettability of One Surface

We were concerned that by changing from material to material to obtain different wettabilities (spreading areas of the water drops), we might change other significant parameters simultaneously. For example, changing the geometry or the thermal characteristics of the substrate beneath the surface might change the nature of the interaction. We therefore initially tried

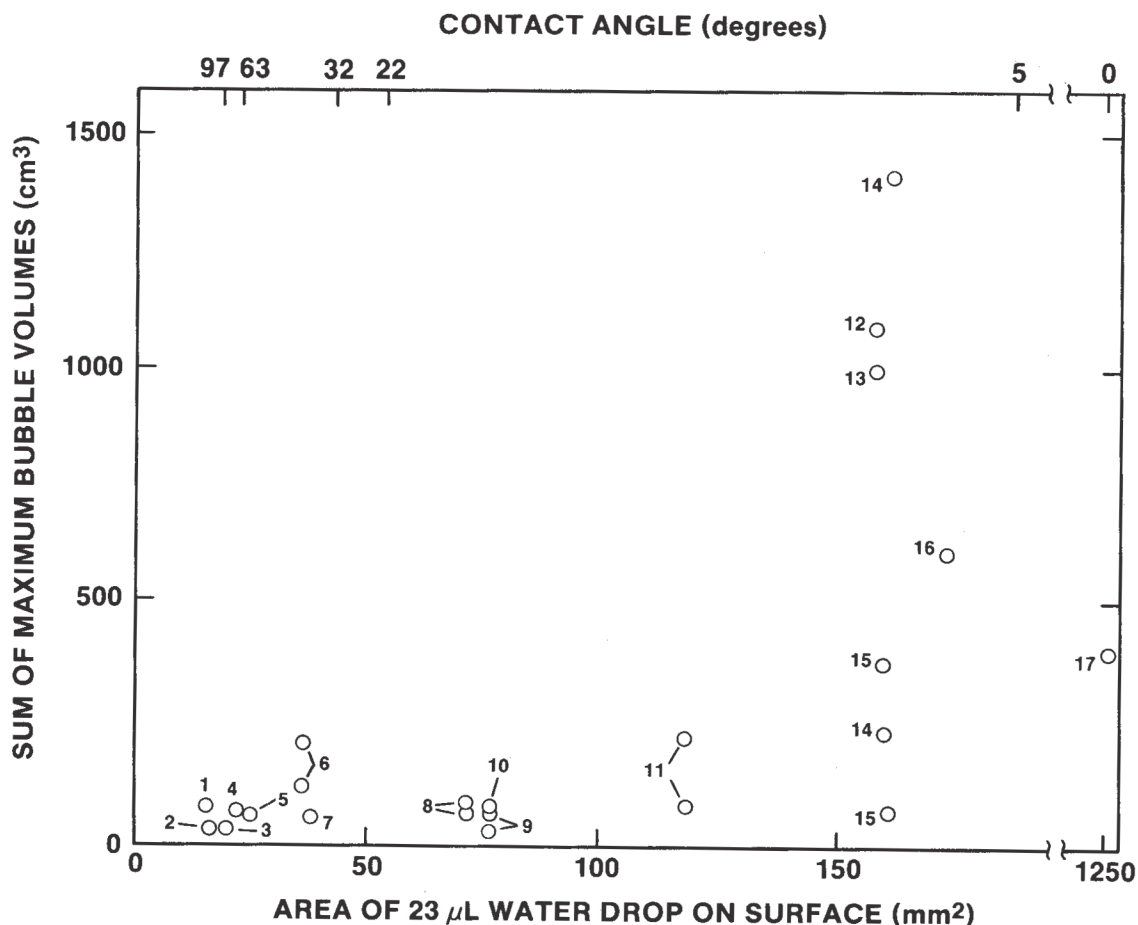


Figure 4. Sum of Maximum Steam Bubble Volumes Produced During Explosive Molten Aluminum-Water Interactions at Submerged Surfaces Plotted Against the Area Assumed by 23 μ L Water Drops on These Surfaces in Air. The numbers identify the various surfaces as listed in Table 1.

experiments on Pyrex and transparent fused silica, attempting to change the wettability by exposing them to various organic materials. Although we were able to vary the wettability of these materials significantly in air at room temperature, we encountered several major problems which led to erratic and nonreproducible results: (a) both Pyrex and fused silica reacted relatively rapidly with molten aluminum, (b) the organic materials used to vary the wettability of the surfaces apparently vaporized or decomposed nonreproducibly in proximity to the molten aluminum, and (c) the Pyrex and fused silica surfaces usually shattered when they participated in an explosion, preventing their reuse for subsequent experiments.

In order to avoid the difficulties with the Pyrex and the fused silica, we switched to a 10 mm-thick copper plate that could be repeatedly cleaned and oxidized. This procedure is similar to that reported in the wettability studies of Horsthemke and Schröder (10). A typical clean and oxidized surface, respectively, are shown in Figure 3. (We altered the spreading by progressively oxidizing the plate by heating in air at approximately 400°C for various lengths of time.) In order to return the plate to its nonoxidized state we cleaned the surface by abrasion with fine crocus cloth.

When the variously wettable clean and oxidized copper plate surfaces were used in tests of the explosivity of the molten aluminum globules, the vigor of the explosive interaction (again estimated by the sum of the maximum steam bubble volumes) increased as the wettability increased. This is shown in Figure 5. The fragmentation of the melts also increased as the wettability increased. This is indicated by the descriptions of the debris shown along the left edge of Figure 5.

Overall Description of the Phenomena

In Table 1 and Figures 4 and 5, we notice the following:

1. The smallest volumes of steam generated, i.e., the mildest interactions, were produced when the areas assumed by the 23 μL water drops were less than about 100 mm^2 . Note that some of the volumes of steam generated were extremely small, for example, on graphite, where the volume was only a few cubic centimeters.
2. The largest volumes of steam, up to about 1400 cm^3 , were generated when the wettability was large, where the drop areas were greater than about 150 mm^2 .

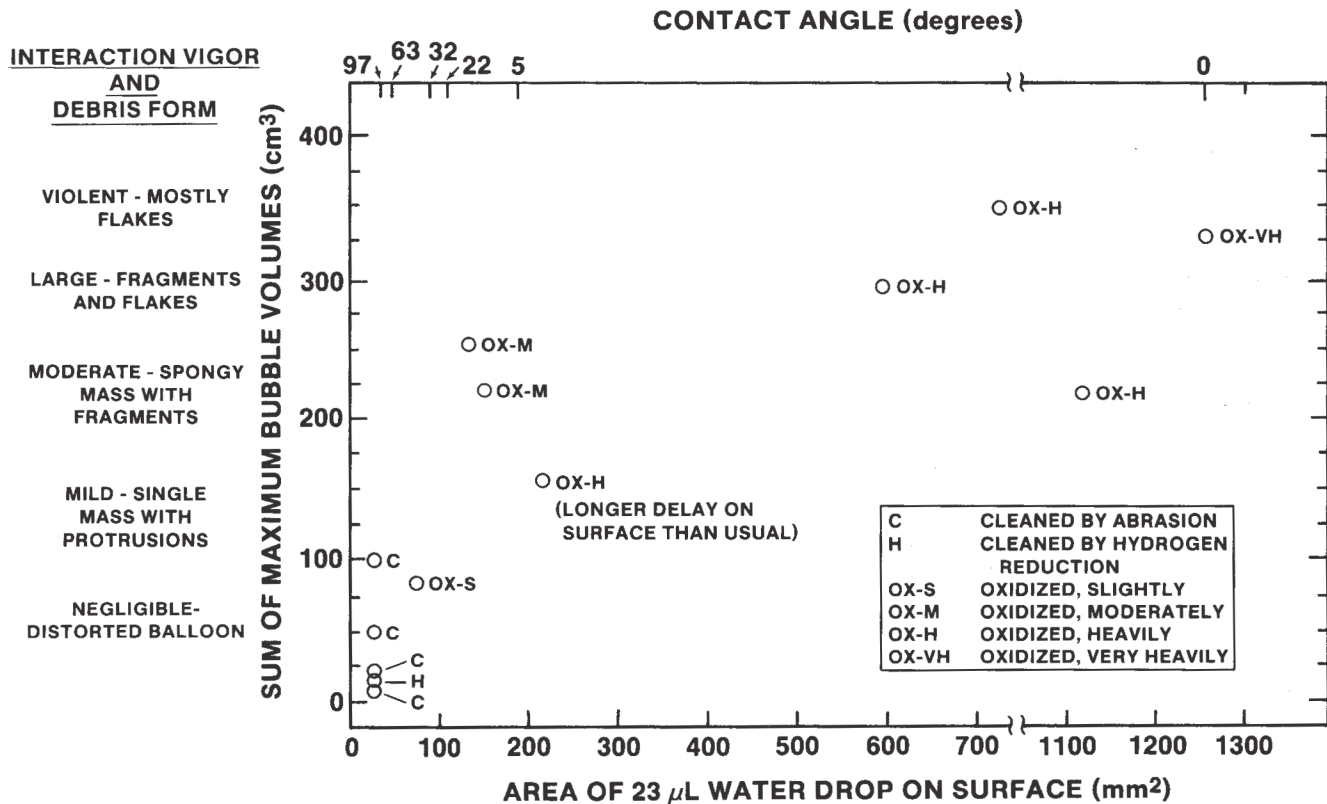


Figure 5. Sum of Maximum Steam Bubble Volumes Produced During Explosive Molten Aluminum-Water Interactions at Submerged Progressively Oxidized Copper Surfaces Plotted Against the Area Assumed by 23 μL Water Drops on These Surfaces in Air. The nature of the surface is indicated next to each data point. The vigor of the interaction and the nature of the debris are indicated at the left of the figure.

3. There was more than an order of magnitude increase in the volume of steam generated when the nonwetable metallic surfaces, Cu, Al, and steel, were treated to produce wettable oxide surfaces--i.e., air-oxidized copper, anodized aluminum and rusted steel.
4. More than an order of magnitude decrease in the maximum volume of steam generated occurred when a wettable gypsum surface was treated with an organic oil. This treated surface produced almost as small an interaction as the graphite surface mentioned above. The behavior of the oil-treated gypsum surface should be compared with the inhibitory action of a "swipe of oil" on the rusted steel surfaces reported by earlier aluminum industry researchers (9).

On the basis of the data presented in Table 1 and in Figures 4 and 5, we conclude that the ability of a submerged or wet surface to assist an explosion is related to its wettability by liquid water.

DISCUSSION

The initiation of molten metal-water explosions at underwater or wet surfaces has been described by a number of authors (3-8); the mechanism is poorly understood, however (11). In the following sections, we propose an hypothesis that we believe rationalizes our experimental observations and many of the aluminum industry's earlier studies with the literature of heat transfer, boiling and bubble nucleation.

Our hypothesis is consistent with these observations:

1. If molten aluminum is released onto a submerged surface, liquid water is retained beneath the melt for relatively long periods, perhaps on the order of seconds. The existence of this water beneath the melt is inferred from the upward indentations ("steam domes") found in the bottom of aluminum that has frozen on a submerged surface without explosion. This phenomenon has been observed both in our work and in earlier work at Battelle Institute (12). We also see evidence of this liquid water by the abrupt jump of the globule of melt as it freezes on a submerged surface, presumably caused by the suddenly increased generation of steam as transition boiling sets in.
2. There is strong similarity between the initiation of explosions with both molten aluminum (3-6) and molten copper (7,8) when poured into water-filled tanks with rusted steel bottoms. Both melts trigger only at the bottom of the tank. Moreover, the interactions are apparently suppressed in both cases when the bottom of the chamber is coated with certain organic materials. The similarity of behavior of two metals as different as copper and aluminum also strongly

suggests that the triggering action is physical, rather than related to the chemistry or metallurgy of the specific metals.

3. Initiation and suppression of the melt-water explosions at underwater surfaces do not seem to involve chemical phenomena. For example, thermite-type interactions have been proposed as initiators by Maischak and Feige (13). At none of the thermite-related surfaces that initiated explosions in our work, for example, rusted steel or air-oxidized copper, was there any sign of a surface reaction that could be detected afterward with a microscope at magnifications up to 100X. Such chemical reactions are also deemed unlikely because of the similarity of the explosive interactions with both molten aluminum and molten copper described in the previous section, even though the chemical reactivities of the two metals are very different.

Furthermore, a chemical explanation is not needed for the inactivity of submerged surfaces, as suggested by Hess and Brondyke (4). These authors attribute the inertness of surfaces with organic coatings to the evolution of pyrolysis gases at the interface in the presence of the hot metal. They postulate that these gases physically separate melt and water, prevent thermal contact, and thereby suppress the steam explosion. In our experiments, we also saw large quantities of such gases form when molten aluminum was poured onto Tarsel-coated steel; much of it bubbled out of the water and produced a "tarry" odor. And indeed, no explosions occurred in our experiments on the Tarsel surface.

This explanation certainly seems viable, especially in industry where organic coatings are used extensively on submerged surfaces. It does not explain, however, the inactivity of other surfaces, such as clean copper, aluminum, grit-blasted steel, graphite and vapor-deposited carbon (see Figure 4 and Table 1), where evolution of pyrolysis (or other) gases seems unlikely. Our mechanism hypothesized below does not require submerged surfaces to evolve gases to achieve inertness in the presence of molten metal. Their presence, of course, would add an extra measure of safety in industrial situations.

4. In order to test the influence of the thermal properties of the solid substrate, we prepared clean copper surfaces both on a polished 10 mm-thick copper plate and by vapor depositing copper onto a Pyrex surface. We also oxidized both surfaces in air at elevated temperatures in order to increase the spreading of water on the surface (for example see Figure 2). As shown in Table 1 and Figure 4, both types of clean copper surfaces were similarly inert, while both oxidized copper

surfaces similarly assisted the steam explosions, even though the thermal properties of the substrate were vastly different.

These experiments were performed to compare the behavior on these two substrates with the hypothesis of Dewing (9) (see also (11)) which attributes the initiation of metal-water explosions at submerged surfaces to the conduction of heat from the melt through the substrate to produce a region of transition boiling at the substrate surface a short distance from the edge of the melt.

Proposed Mechanism

Our hypothesis for the assistance of melt-water explosions at certain underwater surfaces is based on a suggestion by Long (3): "It is believed that the triggering action [at a surface] is a minor explosion due to the sudden conversion to steam of a very thin layer of water trapped below the incoming metal...With grease, oil or paint on the container bottom, the explosion is prevented because the thin layer of water required for the preliminary triggering explosion is not trapped beneath the molten metal."

We extend his idea by substituting a "wetted layer of water" for his "trapped water". Thus the initiation occurs on a wettable surface because it provides a thin layer of water beneath the molten metal, while a nonwettable surface does not provide this layer.

To the concepts suggested by Long, we add the behavior observed in studies of both boiling (14) and bubble nucleation (15) that wettable surfaces act as poor nucleators of bubbles, while nonwettable surfaces provide copious nucleation sites for the generation of bubbles. Thus wettable surfaces tend to promote superheating of water while nonwetting surfaces tend to prevent superheating.

We envision the behavior of the molten metal at surfaces submerged in water as shown in Figures 6 and 7 for the nonwettable and wettable situations, respectively. These figures are based primarily on the comments of Long but augmented with the observations in our experiments and in the literature.

The nonwettable situation depicted in Figure 6 will consist primarily of dry areas beneath the melt. At these dry areas, there probably will be considerable direct contact between the molten metal and the substrate surface. However in some situations, there may be globules of liquid water which are essentially "balled up" in relatively large thicknesses. The presence of these globules is consistent with the steam domes observed both in earlier experiments (12) and in our work.

If the substrate surface is wetted by liquid water, however, we hypothesize that a

thin layer of water is held beneath the molten metal by surface forces. This layer of water is separated from the melt by a boiling film (that is, the melt is levitated a short distance above the liquid water layer). This is shown at the left side of Figure 7. Initiation of the interaction occurs as the result of random spontaneous contact between the melt and the thin layer of water; this is depicted at the right side of Figure 7. (This random contact was simulated in our experiments by the low energy, subthreshold pressure pulse supplied by the underwater spark.) This mechanical motion of the melt forces liquid-liquid contact between the melt and the thin layer of water, permitting it to heat rapidly. Because of the poor nucleation characteristics of the water-wettable substrate surface, the thin layer of water is hypothesized to heat rapidly during this brief contact period and probably superheat far above its normal boiling point. We further hypothesize that this superheated water subsequently flash-vaporizes to fragment melt locally and to create both movement of the melt and pressure disturbances that can initiate steam explosions elsewhere where the melt and the water are contiguous.

If the molten aluminum is initially separated from the layer of water on the wetted substrate by a boiling film, the heat transfer from melt to the liquid water layer across the vapor gap is relatively slow. From traditional boiling curves, the heat flux lies in the region perhaps 10^3 - 10^5 W/m² for normal film boiling (14).

In the case where liquid-liquid contact is forced transiently, the heat transfer will rise dramatically. Recent estimates have been made by Makino and Michiyoshi (16) and in older work by Baines and Board (17) with the transient behavior of water drops falling onto hot solid metal surfaces. Their work indicates that the right hand configuration in Figure 7 might exist metastably for many milliseconds. Moreover, their work indicates instantaneous heat transfer rates from the hot side to the liquid water several orders of magnitude greater than in the case of normal film boiling. This then supports an hypothesis that rapid heating of a thin layer of water sandwiched between melt and a wettable solid substrate might occur on the time scale required for triggering a steam explosion, i.e., about a millisecond. Approximately 0.2 ms is reported as the initiation time for surface triggering of both molten aluminum (6) and copper (7, 8).

We can speculate about the pressure disturbance that might be produced if the layer of liquid water were to superheat to the limit of homogeneous nucleation, approximately 300°C, which is the maximum superheating possible in liquid water as predicted thermodynamically (18). If we assume that the instantaneous vaporization of the highly constrained layer of water sandwiched between melt and substrate surface will produce a pressure pulse equal to its

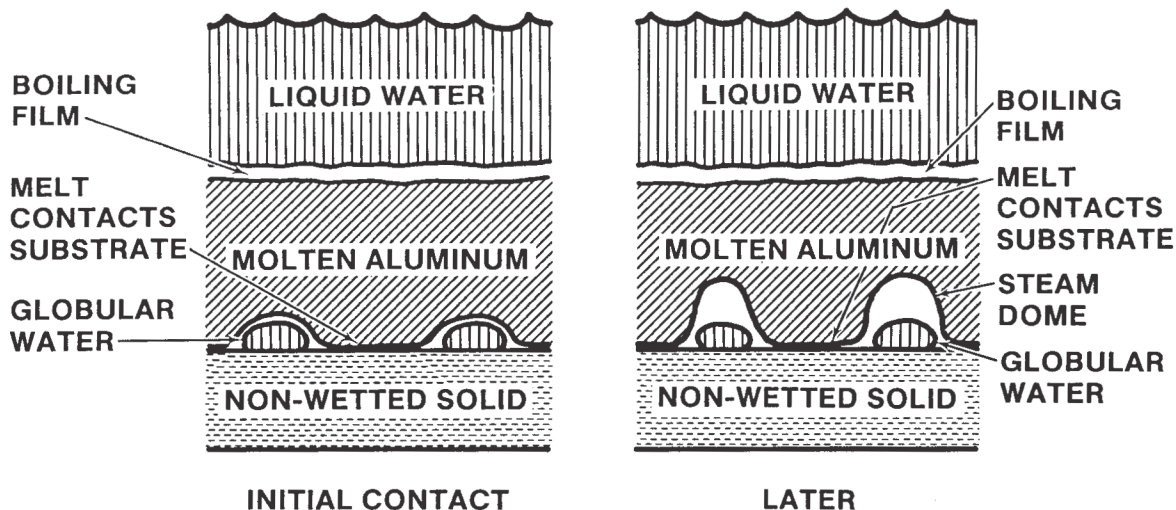


Figure 6. Hypothesized Diagram of Water Entrapped Beneath Molten Aluminum Resting on a Submerged Surface That Is Not Wetted by Water. Some dimensions have been exaggerated for clarity.

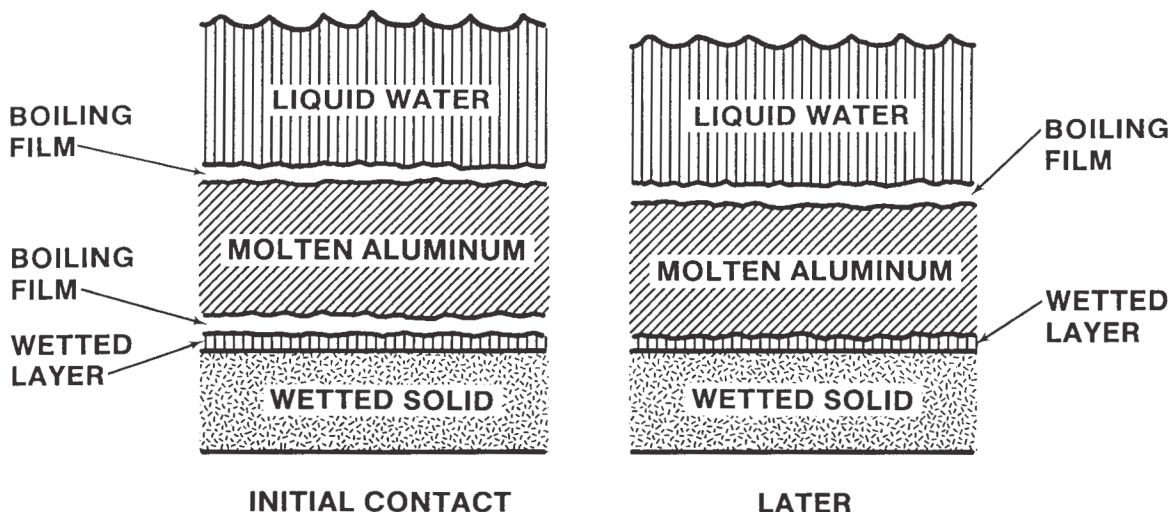


Figure 7. Hypothesized Diagram of Water Entrapped Beneath Molten Aluminum Resting on a Submerged Surface That Is Wetted by Water. Some dimensions have been exaggerated for clarity.

equilibrium vapor pressure at the homogeneous nucleation limit, the flash vaporization-produced trigger transient would be about 8.5 MPa. This is significantly higher than the 5 MPa pulse needed to initiate a steam explosion in a globule of molten aluminum falling freely through water (see above).

There is some indirect chemical evidence that the water held beneath the molten aluminum is superheated. This is derived from the observations of Battelle workers (6) that flashes of white light were sometimes emitted from beneath molten aluminum that had been poured into a water-filled tank with a transparent bottom. The molten aluminum was at a temperature of approximately 1000°C in these experiments. This temperature is below the temperature between 1400°C (19) and 1750°C (20) at which molten aluminum is

reported to ignite in water. The best explanation for the flashes of light emitted downward beneath the molten aluminum is that small patches of aluminum are ignited in water. But since the aluminum does not ignite normally at 1000°C, we assume that ignition occurred because the water was at high temperature. Molten metals, including aluminum, ignite and burn more readily in superheated than in cold water, as discussed by Baker and Liimatainen (21). We conclude, therefore, that the ignition occurred because the thin layer of water had been superheated. (Note, however, that the ignition of aluminum is probably not necessary for the surface initiation of steam explosions because of the similar behavior with molten copper at water wettable surfaces (7, 8). It is unlikely that the molten copper would ignite under these circumstances.)

CONCLUSIONS

On the basis of our laboratory-scale observations, the extensive field experiments of workers in the aluminum industry (3-6, 9, 12), and the literature of boiling and bubble nucleation (14, 15), we hypothesize that the initiating action at a wet or underwater surface is caused by a thin layer of liquid water enclosed beneath molten aluminum and held by surface forces at a water-wettable solid surface. Relatively large patches of direct liquid-liquid contact (see right side of Figure 7) could be produced transiently for times on the order of milliseconds. This contact (in this inherently hydrodynamically unstable configuration) could be forced by normal mechanical perturbations in the system produced, for example, by sloshing during pouring of melt, by vigorous boiling, or intentionally by application of a triggering transient, as in our work. During this brief period of contact, the water layer would superheat significantly, then flash-vaporize. This violent vaporization would fragment melt locally, and send a mechanical disturbance to other regions where melt and liquid water are contiguous. This disturbance would initiate larger steam explosions at those distant locations.

On the other hand, we hypothesize that nonwettable surfaces do not have the thin layer of water between melt and substrate, and offer little or no assistance to triggering during the interaction. Water may be present beneath the melt in some instances, but only as "balled up" globules that cannot superheat rapidly to assist the explosion process.

Although the explanation proposed here seems to rationalize our observations with a large body of literature, our experimental evidence is not extensive enough at this point to solidly substantiate (or negate) any mechanism of surface-assisted explosions. More experiments and theoretical modeling will be required to rigorously test our hypothesis.

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