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THERMAL INTERACTION OF MOLTEN COPPER WITH WATER

WIKTOR ŻYSZKOWSKI*
Institute of Nuclear Research, Swierk, near Warsaw, Poland

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Abstract—The experimental work was performed to study the thermal interaction between molten copper particles (in the range of temperatures from the copper melting point to about 1800°C) and water from about 15–80°C.

The transient temperatures of the copper particles and water before and during their thermal interaction were measured. The history of the phenomena was filmed by means of a high speed FASTAX camera (to 8000 f/s). Classification of the observed phenomena and description of the heat-transfer modes were derived.

One among the phenomena was the thermal explosion. The necessary conditions for the thermal explosion are discussed and their physical interpretation is given.

According to the hypothesis proposed in this paper, the thermal explosion occurs when the molten metal has the temperature of its solidification and the heat transfer on its surface is sufficiently intensive. The "sharp-change" of the crystalline structure during the solidification of the molten metal is the cause of the explosive fragmentation.

NOMENCLATURE

a, β, γ,	quantities in formula (3);
$D_p, d_s, H_B, H_p, h_s, h_t, h_w$,	dimensions in Fig. 2;
F, F_1, F_2,	surface of the metal particle, and the surface of the contact of the particle with fluid and with the bottom of the vessel, respectively (Fig. 2);
k, ρ, c_p, L,	thermal conductivity, specific weight, specific heat, and latent heat of fusion, respectively;
Δl,	thickness of the bottom of the vessel;
M_{kr},	a large value of α (the "critical value" in the thermal explosion accident);
m (or m_{Cu}), r,	mass of the metal particle and its mean radius (calculated from the particle volume), respectively;
q''_k, α_k, D_k,	heat flux, heat-transfer coefficient and the slope of the temperature–time curve, respectively (k can be equal to: $B, L, LW, \ldots$, and it describes the value on the specified segment);
T_a, T_0,	temperature of the air (or inert gas), and water, respectively;
$T_{coil}, T_{ctc}, T_{bott}$,	volume mean temperatures of the particle; in the levitation coil, at the moment of contact with water surface, and with the bottom of the vessel, respectively;
T_{expl},	thermal explosion temperature;
T_I,	interface temperature between the hot and cold liquid at the moment of their contact;
$T_i, (T_m)_i, (T_{me})_i$,	indication of the thermocouple No. i ($i = 1, 2, 3, \ldots, 12$), maximum value of this indication, and its extrapolated maximum value, respectively (Fig. 4);
T_k,	temperature in the point k of the temperature–time curve ($k = B, C, E, \ldots$, e.g. T_L—Leidenfrost point temperature);
$T_M = T_M(\tau)$,	temperature of the metal particle where τ means time;
T_{melt}, T_S,	melting and solidification point of the metal particle, respectively;
u,	percentage of Cu, Cu_2O, CuO content or Cu/Cu_2O content in the metal particle, respectively;
α_t, E, μ,	thermal expansion coefficient, Young's modulus and Poisson's ratio, respectively;
ξ, ζ, η,	space variables according to the Fig. 13;
σ_r, σ_t, p,	radial end tangentid stress, and pressure induced in liquid phase inside the solid shell, respectively;
$\Delta\tau_{FB}$,	duration time of the film boiling phase;
$\Delta\tau_k$,	duration time where k describes the space in Fig. 4;
$\Delta\tau_l(\tau_{lev})$,	levitation time of the particle in the levitation coil ("l" can be equal to: $d, e, f, \ldots$, according to Fig. 3);

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$\Delta\tau_S, \Delta\tau_{Scl}, \Delta\tau_{Se}$,	duration time of the solidification of the metal particle, according to formulae (2). and (3), and extrapolated value from the temperature–time curve, respectively.

Subscripts

M,	metal particle;
o,	water;
m,	maximum value;
me,	maximum extrapolated value.

INTRODUCTION

EXPLOSIVE thermal interaction of the molten reactor fuel with coolant is one of the possible phenomena which can cause a serious destruction of the nuclear reactor structure. Similar a phenomenon occurs accidentally in metal industry and its destructive effects have been described in literature [6, 11].

The strong destructive power of the thermal explosion observed in the metal industry has resulted in many experimental and theoretical studies in this field [2, 6, 8, 19–22 and other] but the physical mechanism of the phenomenon is not yet completely understood.

The thermal explosion may occur when hot molten material comes in contact with relatively cold liquid. The phenomenon is characterized by sudden fragmentation of the molten material, fast energy conversion and the accompanying pressure wave.

A limited thermal explosion is rather simple to realize in a laboratory. A low melting metal is dropped in an open water tank and the following phenomena may be observed:

1. Violent boiling;
2. Spontaneous nucleation;
3. Hydrodynamic fragmentation;
4. Liquid surface instability;
5. Liquid entrainment and entrapment;
6. Pressurization in a shrinking shell;
7. Chemical reaction.

The following hypotheses based on these effects have been advanced to explain the thermal explosion phenomenon:

Violent boiling hypothesis [11–13, 17 and others]: The collapsing of the vapor film and the "explosive" transition from the film to the nucleate boiling (transplosion [17, 19]) generates forces which overcome the surface tension of molten metal, and causes its dispersal. Connected with the transplosion phenomenon, the following fragmentation mechanisms are considered: (a) "resonance fragmentation" [17] caused by the pulsation resonance frequency of the vapor film and (b) high speed liquid jets which penetrate and disperse in the molten material [13].

Spontaneous nucleation hypothesis [18]

Upon mixing of cold liquid with molten metal, some liquid is entrained and wets the metal surface. Because of lack of nucleation sites, its temperature is raised to the temperature limit corresponding to the spontaneous nucleation. When this temperature is reached, vaporization is rapid and can produce shock wave.

Hydrodynamic fragmentation hypothesis [14, 16]

The fragmentation occurs when the hydrodynamic inertial force exerted on the mass of molten metal overcomes the surface tension force at the contact of the metal with the cold liquid. This effect is described by the Weber number and should occur when the molten metal mass exceeds either the critical size, or the critical velocity. It can also appear above some critical temperature when the surface tension characteristic of the molten metal decreases with increase in temperature.

Liquid surface instability hypothesis [9]

According to Taylor's instability theory [1] small wavelike disturbances formed on the liquid surface when it is accelerated in a direction perpendicular to its planes, would tend to increase in size without bound. Liquid will tend to jet out into waves of large amplitude (liquid spikes). The formation and growth of these spikes may be the initiative mechanism of the thermal explosion.

Liquid entrainment and entrapment hypothesis [8, 9]

Cold liquid is trapped inside the molten mass of metal or covered by it at the surface of the container. A pressure pulse caused by steam generation results in fragmentation and thermal explosion of the molten metal.

Shrinking shell hypothesis [9, 10, 14]

The hot molten metal is encased in a thin shell on solidification. Pressurization due to induced thermal stresses causes the blowing of the enclosed molten mass apart.

Chemical reaction hypothesis

The thermal explosion is possible when free hydrogen is released from water during its chemical reaction with the hot molten metal. It has, however, been proved [9, 11] that such a chemical reaction is not the cause of the thermal explosion.

The foregoing hypotheses do not, however, fully explain the thermal explosion mechanism, and some questions arise when we compare these theories with experimental facts.

In this paper, the results of experiments about the thermal interaction between molten metal and water are presented. The necessary conditions for the thermal explosion are discussed and a physical interpretation is given.

EXPERIMENTAL TECHNIQUE

The purpose of the experiment is to explore the thermal interaction of small amounts of molten metal with a cold liquid and to measure their temperature history during the interaction for various initial conditions of both the substances. This is realized by means of the levitation coil [3,5], and a small vessel with thermocouples at its base (Fig. 1). The vessel is filled with cold water, the temperature and volume of which are varied.

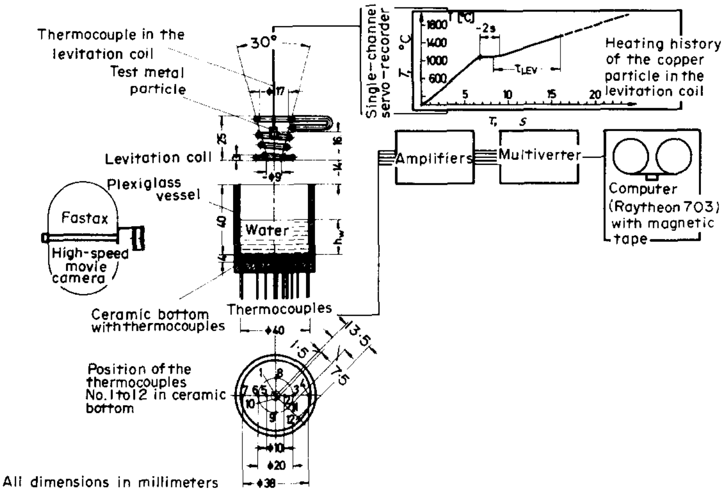

FIG. 1. Schematic drawing of the experimental apparatus and the recording equipment.

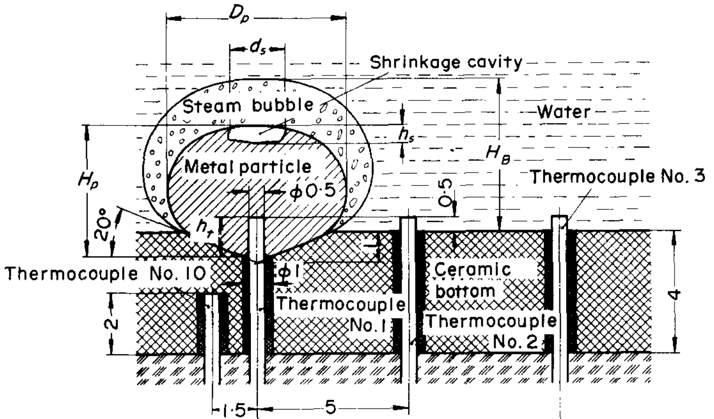

FIG. 2. Model of the metal particle on the bottom of the experimental vessel.

The small metal particle hangs on a thin wire or on a thermocouple in the levitation coil. The typical heating history of the metal particle is shown in Fig. 3. The levitation condition begins at the point *b*, and no mechanical support of the particle is then necessary. The temperature of the particle increases without any contact with its surroundings and the levitation time $\Delta\tau_l$ determines its final value (in some of the tests this method was used for temperature approximation of the metal particle in the levitation coil). In the levitation coil, the following metals are melted: Ag, Al, Au, Cu, Nb, Pb, Sn, Zn and stainless steel [22].

The hot molten metal particle is dropped down into the vessel containing the cold liquid on shutting-off of the current in the levitation coil resulting in the thermal interaction of both the substances.

In experiments with water, a plexi-glas vessel is used to make the observation of the phenomena possible. The bottom of the vessel is made from a ceramic (Thermostix 2000) in which twelve thermocouples are installed (Figs. 1 and 2). Thermocouple No. 1, placed in the center of the bottom (Fig. 2), measures the temperature history of the metal particle. The thermocouples Nos. 2–9 measure the temperature of the water, and thermocouples Nos. 10–12 measure the temperature of the ceramic bottom itself.

Standard Chromel–Alumel thermocouples of the grounded-type and 0·5 mm in O.D. encased in a steel sheath (4301) with MgO insulating material are used. Signals from the thermocouples are registered by a high speed digitizing system based on the Raytheon 703 Computer. The 500 Hz frequency response is used in the measurements. Signals from the thermocouple in the levitation coil (of the same type as at the bottom of the vessel) are indicated by the single-channel servo-recorder and registered on the paper tape. Photographic data are obtained using Fastax high-speed movie camera (to 8000 frames/s) and Bolex framing camera (32 frames/s).

A block diagram of the experimental arrangement is shown in Fig. 1.

RESULTS OF MEASUREMENTS

The measurements with copper are presented below. Technically pure copper (E–Cu 99·9 Din 1708) particles weighing about 0·5 g are heated in the levitation coil in the temperature range from its melting point to about 1800°C. For various copper particles, the shut-off temperature, T_{coil}, (or levitation time $\Delta\tau_l$) in the levitation coil, the temperature of the water from about 15 to 80°C, the volume of the water from 10 to 50 cm^3, and the distance between the levitation coil and the plexiglas vessel from 1 to 14 mm are varied.

The phases of heating and cooling through which the individual metal particle passes in the experiments are as follows:

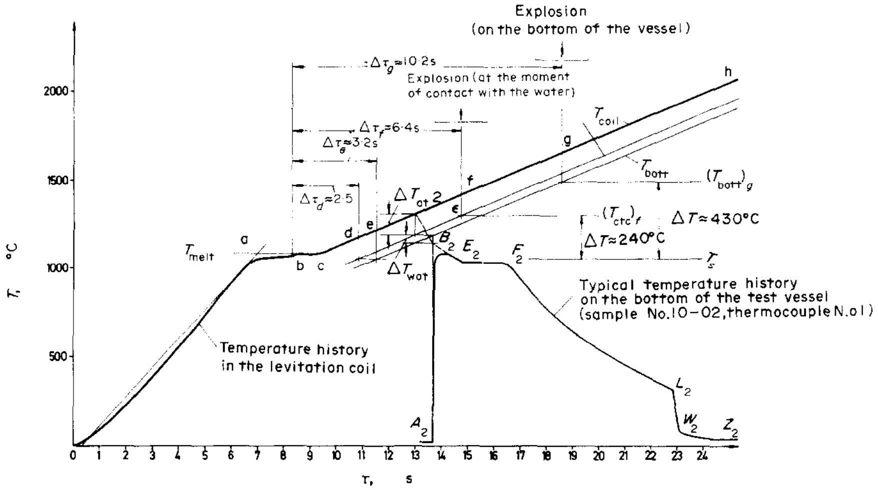

FIG. 3. Typical temperature history of the copper-particle in the experimental apparatus. (a) Start of melting of the particle. (b) Start of levitation. (c) End of melting. (d–h) Heating in levitation condition. Cut-off of heating current (drawn for the sample No. 10–02). $\Delta T_{at} = T_{coil} - T_{ctc}$, $\Delta T_{wat} = T_{ctc} - T_{bott}$, where T_{coil}, T_{ctc}, T_{bott} are temperatures of the particle: in the coil, at the moment of contact with water surface and with the bottom of the vessel, respectively.

1. Heating above its melting point in the levitation coil.
2. Fall in air (or inert gas).
3. Entry into and fall in the water.
4. Contact with the bottom of the vessel.
5. Cooling and equalization of the metal temperature with ambient temperature.

Solidification of the metal begins during one of the above mentioned 2–5 phases. Fragmentation or thermal explosion may also or may not occur during one of these phases depending on certain conditions which are discussed later in this paper.

In these experiments, the copper particle takes a spheroidal form in air, and during its fall develops a velocity of about 70 cm/s before it comes in contact with the water surface.

Entry of the particle into the water can be quiet and connected with simultaneous formation of a steam–air atmosphere around the particle (Fig. 6). The atmosphere protects the particle surface and forces the water in front of the particle out. Some flattening of the particle form is seen.

A short distance below the water surface, a sudden thermal explosion of the copper particle can also occur. The author observed such a thermal explosion only for the levitation time $\Delta\tau_l \cong 6{\cdot}4$ s ($T_{\text{coil}} \sim 1400°$C, diameter of the vessel—40 mm, distance between the vessel and the levitation coil—14 mm, volume of the water—20–30 cm^3, water temperature $\sim 20°$C).

Strong (Fig. 8) and weak (Fig. 10) thermal explosions below the water surface are distinguished. On the high speed films, no vapor film can be seen between the particle and the water. The thermal explosion happens, however, about 1·5–4 ms after the first contact of the particle with the water surface. This delay seems to be very important in understanding of the thermal explosion phenomenon. The front of the strong thermal explosion expands with the velocity of about 35 m/s. For the weak thermal explosion below the water surface, the copper particle can break up later at the bottom of the vessel (hydrodynamic partition) (Fig. 10). When, however, the particle splits into comparatively large particles during the thermal explosion, then these particles can again come together as they move further to the bottom.

The interface temperature T_I between the hot and cold liquid at the moment of their sudden contact is given by:

$$T_I = (T_o + aT_{\text{ctc}})/(1+a) \qquad (1)$$

where

$$a = (k_M \rho_M c_{pM}/k_o \rho_o c_{po})^{0{\cdot}5}.$$

For the copper–water contact at the respective temperatures 1100 and 20°C, $a = 14{\cdot}5$. In this case, the temperature T_I is about 80°C lower than the temperature T_{ctc}.

When no thermal explosion occurs during the fall in water, the particle moves quietly with velocity 80–90 cm/s. The vapor–air film moves ahead of the copper particle, and a steam–air "chimney" originates behind it. The "chimney" may remain till the copper particle comes to rest on the bottom of the vessel and even afterwards, and later on it pinches out. The copper particle flattens out at the bottom* and subsequently, it takes the spheroidal form again. Sometimes, the particle "dances" on the bottom and it touches the neighboring thermocouples before it settles on the thermocouple No. 1 (Fig. 4). The "dance" of the particle and delay in the thermocouple readings result in the temperature $T_M = T_{\text{bott}}$ of the first particle contact with the bottom of the vessel not being known exactly. However, a certain maximum temperature $(T_m)_i$ for the thermocouple No. 1 is obtained, and from this the extrapolated maximum temperature $(T_{me})_i$ is calculated (Fig. 4). It is assumed that $(T_{me})_i = T_{\text{bott}}$. The time which elapses between the shut-off of the current in the levitation coil and the first reading of the thermocouples at the bottom is about 60 ms.

The spheroidal molten copper particle subsequently cools at the bottom of the vessel. This period is the longest one in the cooling history of the particle, and is about 2–25 s for various particles tested (Fig. 4).

For the particle No. "i" ($i = 1, 2, 3, \ldots$), the following phases during its cooling history are distinguished:

(BL)$_i$—film boiling;
(BCE)$_i$—particle is molten;
(EF)$_i$—solidification of the molten particle;
(FLPWZ)$_i$—particle is solid;
(LP)$_i$—transition boiling (this phase is often reduced on Fig. 4 to one point, i.e. $L_i = P_i$);
(PW)$_i$—violent nucleate boiling;
(WZ)$_i$—end of the violent nucleate boiling and beginning of pure convection.

The heat-transfer history according to the above classification can be regarded as "normal" or "quiet". It takes place usually for these metals which have the solidification temperature higher than the Leidenfrost temperature of the cooling liquid (i.e. $T_M = T_S > T_L$). Deviations from such a "normal" course are however possible, and various courses of the phenomena are observed. For a fixed geometry, the main factor in these experiments is the temperature of the particle at the first contact with water ($T_M = T_{\text{ctc}}$).

It is observed, that when the molten copper particle has already taken a spheroidal form at the bottom of the vessel the "normal" course of heat transfer follows. However, the thermal explosion or related phenomena take place during the cooling process of the metal particle when the temperature of the particle at its first contact with the bottom of the vessel has a value which is very nearly equal to the temperature $(T_{\text{bott}})_g$ in Fig. 3. For such a case, at first, the "normal" heat transfer begins, and after a short period of time, we can observe: (i) thermal explosion, (ii) origination of "empty shell" or "fungus-form" particle, (iii) "shots" of molten metal.

The range of these irregularities is shown in Fig. 7 for the samples Nos. 13–10, 9–02, 13–35, 13–36 and 13–33. The corresponding temperature readings are shown in Fig. 4.

Figure 4 shows the temperature history of the copper particle for the thermal explosion at the bottom of the vessel for the sample Nos. 13–35. The maximum

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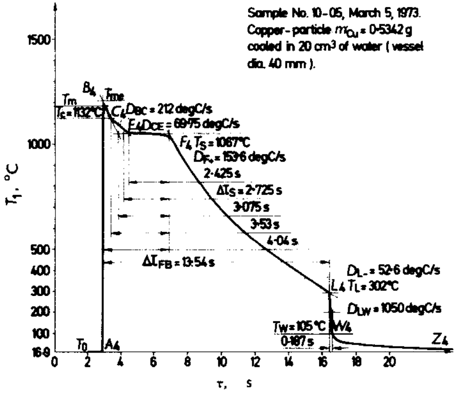
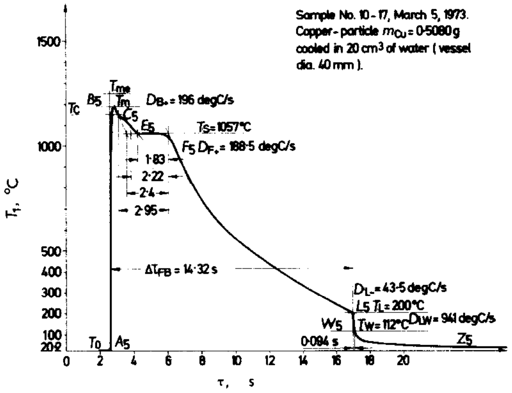
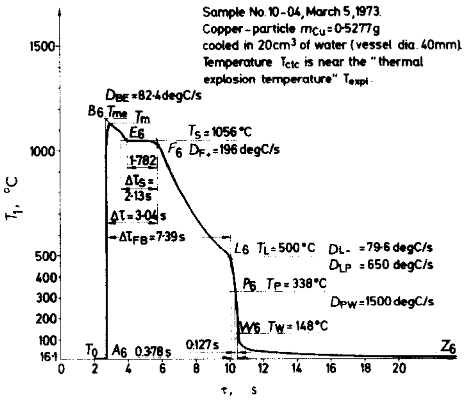
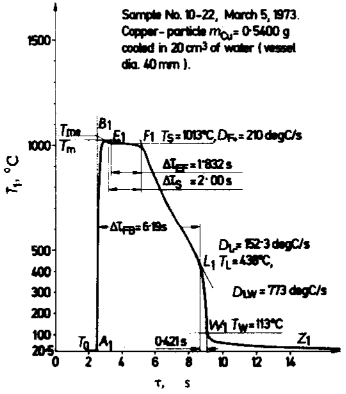
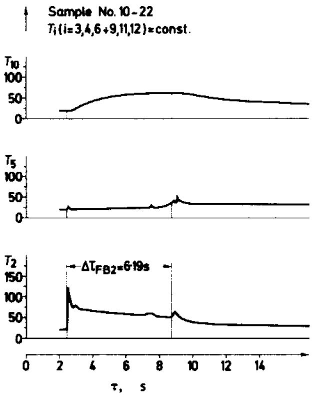
*For other metals, e.g. Ag, Pb, Sn, Zn, the molten metal particle solidified sometimes as a flat "plate" during this phase. Some rings (Helmholtz surface instability) [9] could be observed on the surface of such particles [22].

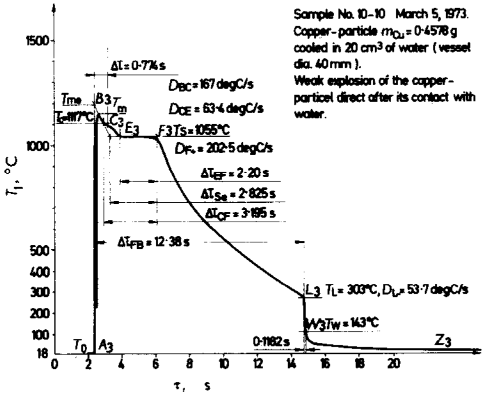

Sample No. 10-05, March 5, 1973.
Copper-particle m_{Cu} = 0·5342 g
cooled in 20 cm³ of water (vessel
dia. 40 mm).
B_4
T_{me}
T_m
T_c = 1132°C
C_4 D_{BC} = 212 degC/s
E_4 D_{CE} = 69·75 degC/s
F_4 T_S = 1067°C
D_{F+} = 153·6 degC/s
2·425 s
$\Delta\tau_S$ = 2·725 s
3·075 s
3·53 s
4·04 s
$\Delta\tau_{FB}$ = 13·54 s
D_{L-} = 52·6 degC/s
L_4 T_L = 302°C
D_{LW} = 1050 degC/s
T_W = 105 °C W_4
0·187 s
T_0
A_4
Z_4
T_1, °C
τ, s
16·9
0, 2, 4, 6, 8, 10, 12, 14, 16, 18, 20
100, 200, 300, 400, 500, 1000, 1500

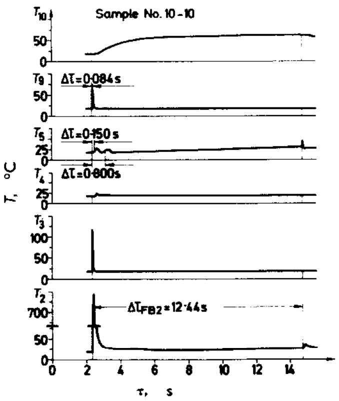

Sample No. 10-17, March 5, 1973.
Copper-particle m_{Cu} = 0·5080 g
cooled in 20 cm³ of water (vessel
dia. 40 mm).
T_{me}
B_5
T_m
T_C
D_{B+} = 196 degC/s
C_5
E_5
T_S = 1057°C
F_5 D_{F+} = 188·5 degC/s
1·83
2·22
2·4
2·95
$\Delta\tau_{FB}$ = 14·32 s
D_{L-} = 43·5 degC/s
L_5 T_L = 200°C
W_5
T_W = 112°C
D_{LW} = 941 degC/s
0·094 s
T_0
A_5
Z_5
T_1, °C
τ, s
20·2
0, 2, 4, 6, 8, 10, 12, 14, 16, 18, 20
100, 200, 300, 400, 500, 1000, 1500

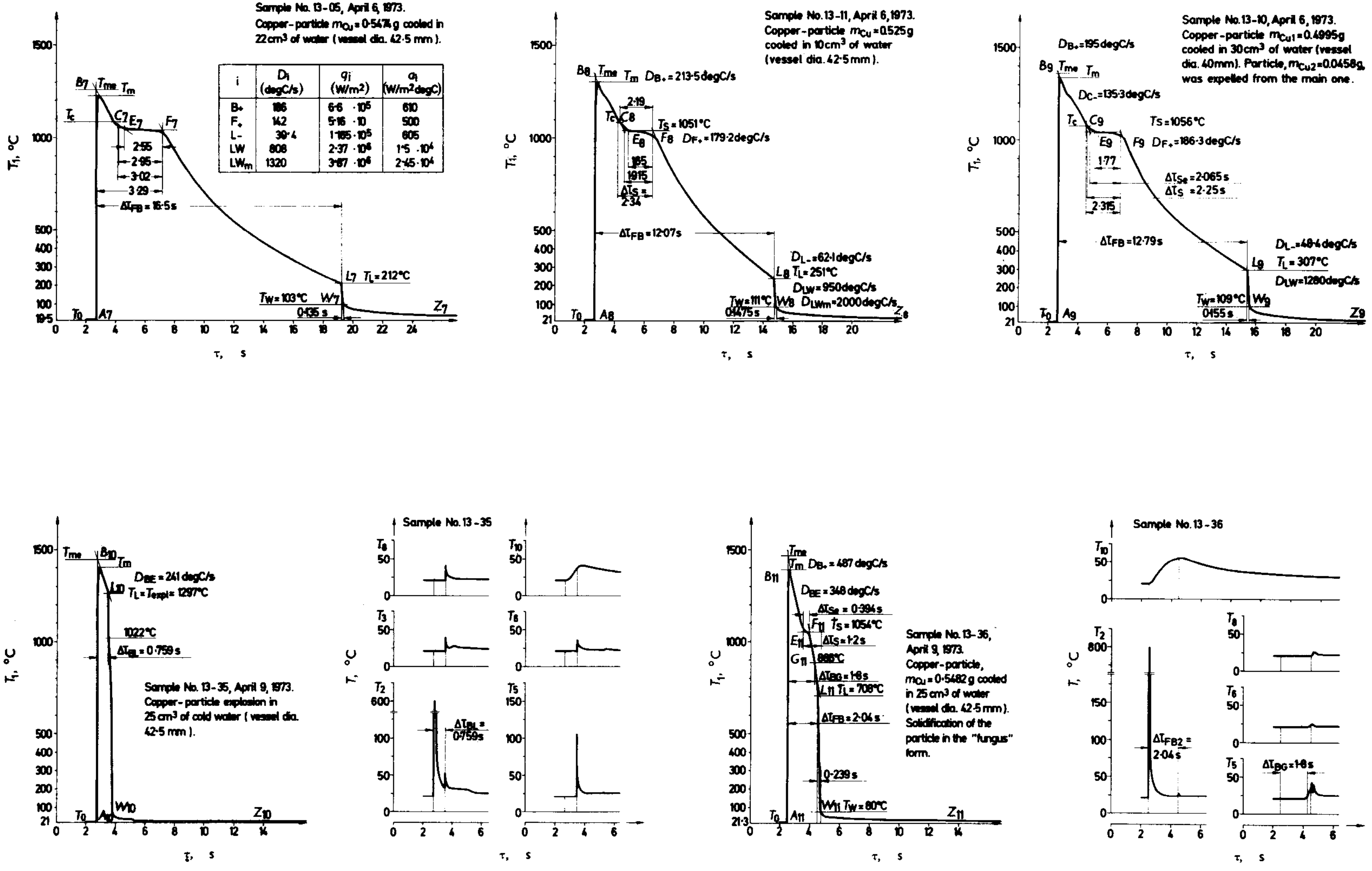

Sample No. 10-04, March 5, 1973.
Copper-particle m_{Cu} = 0·5277 g
cooled in 20 cm³ of water (vessel dia. 40 mm).
Temperature T_{ctc} is near the "thermal
explosion temperature" T_{expl}.
D_{BE} = 82·4 degC/s
B_6 T_{me}
T_m
E_6
T_S = 1056 °C
F_6 D_{F+} = 196 degC/s
1·782
$\Delta\tau_S$ = 2·13 s
$\Delta\tau$ = 3·04 s
$\Delta\tau_{FB}$ = 7·39 s
L_6 T_L = 500 °C
D_{L-} = 79·6 degC/s
D_{LP} = 650 degC/s
P_6 T_P = 338 °C
D_{PW} = 1500 degC/s
W_6 T_W = 148 °C
T_0
A_6 0·378 s
0·127 s
Z_6
T_1, °C
τ, s
16·1
0, 2, 4, 6, 8, 10, 12, 14, 16, 18, 20
100, 200, 300, 400, 500, 1000, 1500


Sample No. 10-22, March 5, 1973.
Copper-particle m_{Cu} = 0·5400 g
cooled in 20 cm³ of water (vessel
dia. 40 mm).
B_1
T_{me}
T_m
E_1
F_1 T_S = 1013°C, D_{F+} = 210 degC/s
$\Delta\tau_{EF}$ = 1·832 s
$\Delta\tau_S$ = 2·00 s
$\Delta\tau_{FB}$ = 6·19 s
D_{L-} = 152·3 degC/s
L_1 T_L = 438°C,
D_{LW} = 773 degC/s
W_1 T_W = 113°C
0·421 s
T_0
A_1
Z_1
T_1, °C
τ, s
20·5
0, 2, 4, 6, 8, 10, 12, 14
100, 200, 300, 400, 500, 1000, 1500


Sample No. 10-22
T_i (i = 3, 4, 6 ÷ 9, 11, 12) = const.
T_{10}
T_5
T_2
$\Delta\tau_{FB2}$ = 6·19 s
T, °C
τ, s
0, 50, 100, 150
0, 2, 4, 6, 8, 10, 12, 14


Sample No. 10-10 March 5, 1973.
Copper-particle m_{Cu} = 0·4578 g
cooled in 20 cm³ of water (vessel
dia. 40 mm).
Weak explosion of the copper-
particel direct after its contact with
water.
$\Delta\tau$ = 0·774 s
D_{BC} = 167 degC/s
D_{CE} = 63·4 degC/s
T_{me}
B_3 T_m
T_c = 1117°C
C_3
E_3
F_3 T_S = 1055°C
D_{F+} = 202·5 degC/s
$\Delta\tau_{EF}$ = 2·20 s
$\Delta\tau_{Se}$ = 2·825 s
$\Delta\tau_{CF}$ = 3·195 s
$\Delta\tau_{FB}$ = 12·38 s
L_3 T_L = 303°C, D_{L-} = 53·7 degC/s
W_3 T_W = 143°C
0·1182 s
T_0
A_3
Z_3
T_1, °C
τ, s
18
0, 2, 4, 6, 8, 10, 12, 14, 16, 18, 20
100, 200, 300, 400, 500, 1000, 1500


Sample No. 10-10
T_{10}
T_9
$\Delta\tau$ = 0·084 s
T_5
$\Delta\tau$ = 0·150 s
T_4
$\Delta\tau$ = 0·800 s
T_3
T_2
$\Delta\tau_{FB2}$ = 12·44 s
T, °C
τ, s
0, 25, 50, 100, 700
0, 2, 4, 6, 8, 10, 12, 14



Sample No. 13-05, April 6, 1973. Copper-particle $m_{Cu} = 0{\cdot}5474$ g cooled in 22 cm³ of water (vessel dia. 42·5 mm).

i	D_i (degC/s)	q_i (W/m²)	α_i (W/m² degC)
B+	186	$6{\cdot}6 \cdot 10^5$	610
F+	142	$5{\cdot}16 \cdot 10$	500
L−	39·4	$1{\cdot}185 \cdot 10^5$	605
LW	808	$2{\cdot}37 \cdot 10^6$	$1{\cdot}5 \cdot 10^4$
LW_m	1320	$3{\cdot}87 \cdot 10^6$	$2{\cdot}45 \cdot 10^4$

Sample No. 13-11, April 6, 1973. Copper-particle $m_{Cu} = 0{\cdot}525$ g cooled in 10 cm³ of water (vessel dia. 42·5 mm).

Sample No. 13-10, April 6, 1973. Copper-particle $m_{Cu1} = 0{\cdot}4995$ g cooled in 30 cm³ of water (vessel dia. 40 mm). Particle, $m_{Cu2} = 0{\cdot}0458$ g, was expelled from the main one.

Sample No. 13-35, April 9, 1973. Copper-particle explosion in 25 cm³ of cold water (vessel dia. 42·5 mm).

Sample No. 13-35

Sample No. 13-36, April 9, 1973. Copper-particle, $m_{Cu} = 0{\cdot}5482$ g cooled in 25 cm³ of water (vessel dia. 42·5 mm). Solidification of the particle in the "fungus" form.

Sample No. 13-36

FIG. 4. Temperature histories of the cooling of the various copper-particles in water.

registered temperature of the thermocouple No. 1 is $T_{m1} = 1408°C$ and corresponding temperature $T_{me1} = 1454°C$. After the time period $\Delta\tau_{BL} \cong 0{\cdot}759$ s of the film boiling mode, thermal explosion occurs. It is observed that the heat transfer mode changes abruptly at the temperature $T_L = T_{\text{expl}} = 1297°C$. The temperature T_1 sinks almost vertically to the temperature equal to 1022°C which is almost the solidification temperature of copper. Such observations recur in these measurements for the thermal explosions at the bottom of the vessel, i.e. the temperature $T_L \simeq 1300°C$ of the copper–water contact was measured several times. In Fig. 9 is seen the corresponding sequence of development of the thermal explosion at the bottom of the vessel. After 0·774 s from the first contact of the particle with the water surface (or 0·756 s from the first contact with the bottom of the vessel), the vapor film disappears and the first "shot" of the molten metal is observed. After a delay of about 5 ms, the full thermal explosion occurs. Thus, similar as in the case of the thermal explosion below the water surface, the delay between the direct contact of both substances and the thermal explosion is observed.

The thermal explosion products after the strong thermal explosion have, partially the spherical form with diameters of about 10–100 μ, and partially "shattered" form of about 10 μ.

At the temperature near to the temperature T_{bott} corresponding to the thermal explosion at the bottom, during the cooling history are observed: "shots" of molten metal (sample No. 13–10, and No. 9–02 in Fig. 7) or particles in the form of "empty shell" or in "fungus-form" (sample Nos. 13–33 and 13–36 in Fig. 7).

The temperature history of the "fungus-form" particle (Fig. 4, sample No. 13–36) has some similarities to that of the thermal explosion at the bottom, namely, $T_{m1} = 1390°C$, $T_{me1} = 1467°C$. The time of solidification is very short, and is equal to about $\Delta\tau_{Se} = 0{\cdot}394$ s. It proves that the increase of the particle size takes place during solidification. The calculated time of solidification without the increase of the particle size is about $\Delta\tau_{Scl} = 1{\cdot}2$ s. The sequence from the film (Fig. 11) shows the growth of the particle at the bottom. It seems that the vapor film exists partly on the surface of the particle for some time after the start of the solidification of the particle.

For the "normal" course of heat transfer, the vapor film exists around the particle for some time after the end of solidification of the particle. The solidified particle has the form of a spheroid with a small shrinkage cavity (sample Nos. 10–22, 13–05, 13–11 in Fig. 7).

The plateau of the temperature T_1 in Fig. 4 for the "normal" course is proportional to the time of solidification. For the pure copper particle which can be considered as a lumped system, the following times are distinguished: $\Delta\tau_{Se}$—the extrapolated value of the solidification time obtained from the "idealized" temperature measurement, and $\Delta\tau_S$—the calculated one obtained on the assumption of the continuity of the heat-transfer rate before and after the solidification of the particle:

$$\Delta\tau_S = Lm_{\text{Cu}}/[F_1\alpha(T_S - T_o) + F_2k(T_S - T_a)/\Delta l] \qquad (2)$$

where F_1, F_2 are surfaces of the particle in contact with the fluid, and with the bottom of the vessel, respectively.

These values are compared with the result of the simplified calculations according to the formula (for $\beta \gg \gamma$) [7]:

$$\Delta\tau_{Scl} = r^2L\beta/k(T_S - T_o)(1 + 2\beta) \qquad (3)$$

where $\beta = k/\alpha r$; $\gamma = c_p(T_S - T_o)/L$; k, c_p, L, r are respectively: thermal conductivity, heat capacity, fusion heat, and radius of the copper particle; and α is the heat-transfer coefficient.

For the copper particle heated in the air, the copper oxide layer originates on the surface of the particle. The solidification point of this layer is higher as the solidification point of the pure copper (see Appendix). This causes a deflection of temperature history on the CE-section in Fig. 4. Such a deflection is not observed in experiments when the copper particle is heated in an inert gas.

On the Fastax films, one can observe that the initial phase of the film boiling is unstable. The vapor film pulsates irregularly about the particle surface and a local direct contact of the molten metal with water is conceivable. The vapor film grows on a portion of the particle while it decreases in thickness at some other point. Next, the film boiling attains a stable character, but one can, however, distinguish between the following three types of courses: (i) one steam bubble of almost constant size surrounds the metal particle; (ii) one steam bubble exists which strongly increases in size or pulsates; (iii) several steam bubbles occur which depart from the metal particle one after the other.

The heat-transfer rate is computed from the temperature measurements as follows:

$$q_k'' = mc_pD_k/F \qquad (4)$$

and

$$\alpha_k = [mc_pD_k - F_2k(T_M - T_a)/\Delta l]/F_1(T_M - T_o) \qquad (5)$$

where $D_k = \mathrm{d}T/\mathrm{d}\tau$ is the tangent of the temperature curve.

The heat flux q_1'' as a function of the difference in temperature between the particle and the water is shown in Fig. 5. This graph is the summary of about 100 measurements. Heat flux decreases continuously

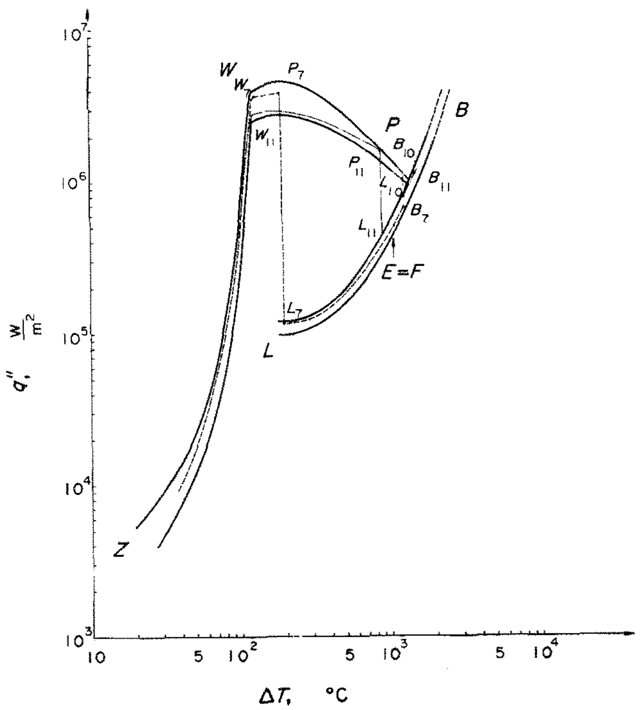


FIG. 5. Heat flux characteristic of the quenching of the molten copper-particles in cold water ($T_o \cong 20°C$). $B-L$, range of the film boiling; $P-W$, range of the intensive nucleate boiling; $W-Z$, range of the mild nucleate boiling and beginning of the pure convection; $L_i-P_i(i=1,2,3,\ldots)$, transplosion for the individual particle i during its cooling history ($B_iL_iP_iW_iZ_i$); B_i, entry of the copper-particle in water; Leidenfrost point.

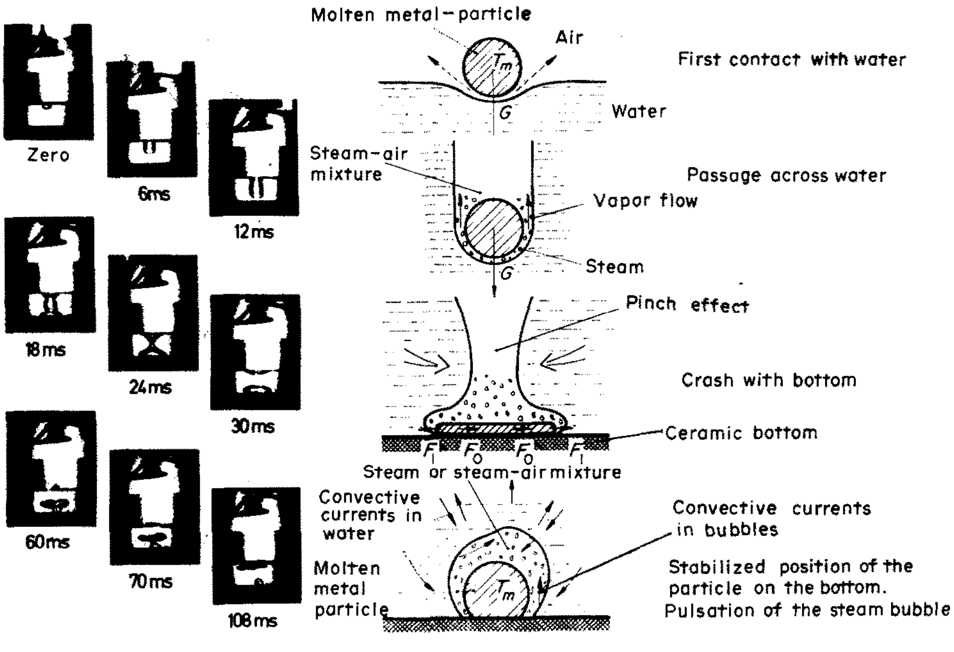


FIG. 6. Schematic model of particle-entry in water.

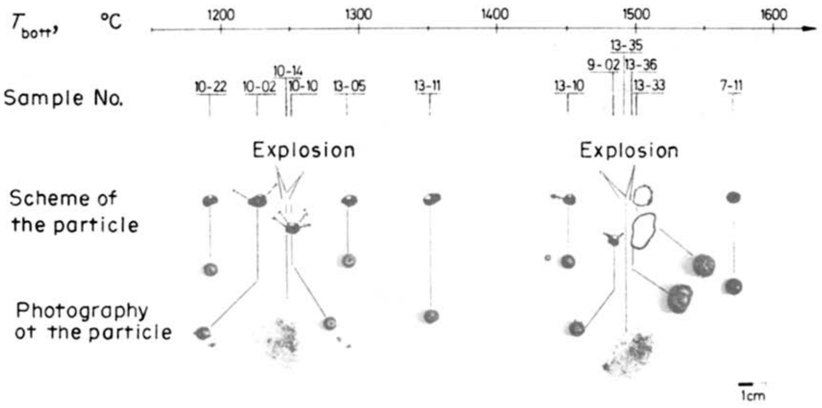


FIG. 7. Form of the metal particle after thermal interaction with water for various initial temperatures (T_{bott}) on the bottom of the vessel.

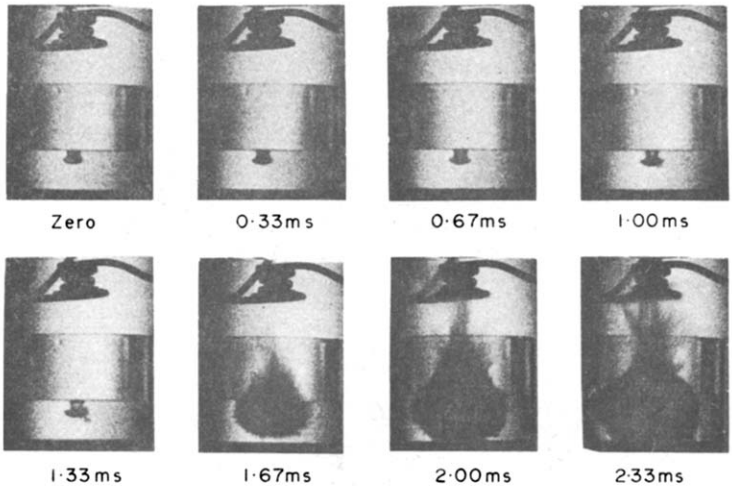

Zero
0·33ms
0·67ms
1·00ms
1·33ms
1·67ms
2·00ms
2·33ms

FIG. 8. Sequence from a film showing development of a thermal explosion. Copper in water. $T_{ctc} \sim 1300°\text{C}$, $T_o = 19{\cdot}5°\text{C}$, $m_{Cu} \cong 0{\cdot}5\,\text{g}$. Sample No. 13-04.

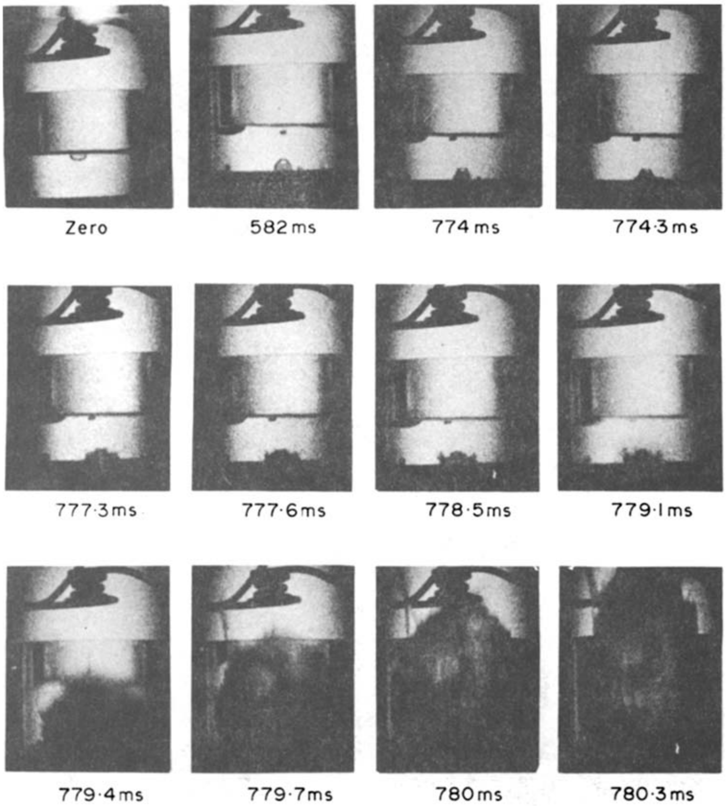
during the film boiling regime from its initial value $q''_B \sim 10^6\,\text{W/m}^2$ in these experiments and it can approach the value $q''_L \sim 10^5\,\text{W/m}^2$ (the range BL in Fig. 5). Afterwards, stable film boiling ceases abruptly (transplosive [17]) and violent nucleate boiling begins.

These measurements indicate that the transplosion can happen in any point (Leidenfrost point) of the film boiling regime during the cooling history of the individual particle "i". In Fig. 5, one can see, e.g. the points L_7, L_{10}, L_{11} for sample Nos. 13–05, 13–35, 13–36, respectively. The other values can be found in Fig. 4.

The transplosion from the film to nucleate boiling seems to have random character and it depends, e.g. on the geometry of the interaction zone, hydrodynamic conditions and the external disturbances during the boiling phase. Before the transplosion occurs, some destabilization of the vapor film is observed. Next, the vapor film is broken in the duration time well below 0·3 ms. A "cloud" of small air bubbles may disperse from the particle surface. Accoustic effects and strong disturbances of the surrounding water are observed. The thermocouples in the water record some temperature peaks at that moment (Fig. 4).

Sometimes, the transition boiling regime occurs between the film and nucleate boiling.

The heat flux jumps from its value at the Leidenfrost point to its value in the violent nucleate boiling

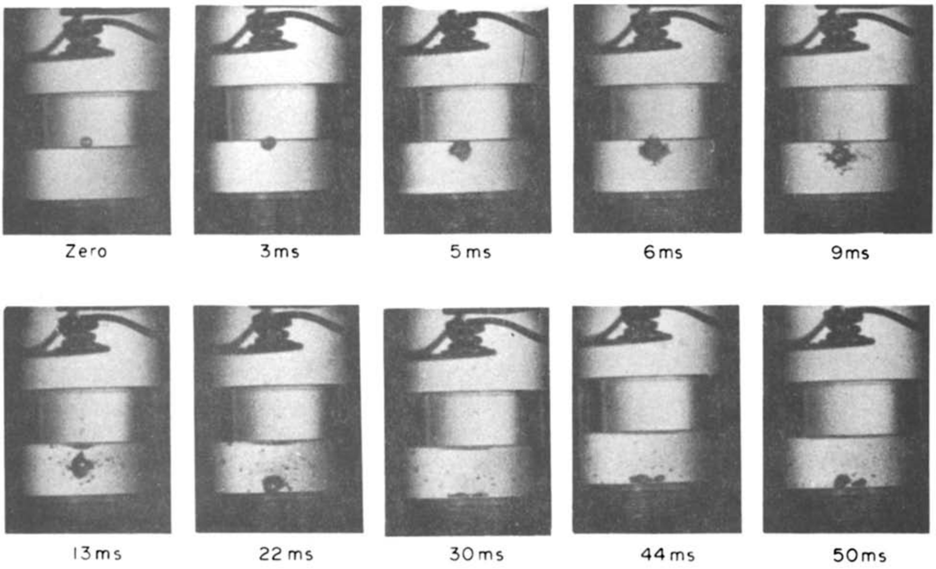


Zero 582 ms 774 ms 774·3 ms

777·3 ms 777·6 ms 778·5 ms 779·1 ms

779·4 ms 779·7 ms 780 ms 780·3 ms

FIG. 9. Sequence from a film showing development of a thermal explosion on the bottom. Copper in water. $T_{\mathrm{bott}} = 1454°\mathrm{C}$, $T_o = 21°\mathrm{C}$, $m_{\mathrm{Cu}} \cong 0{\cdot}5\,\mathrm{g}$. Sample No. 13–35.



Zero 3 ms 5 ms 6 ms 9 ms

13 ms 22 ms 30 ms 44 ms 50 ms

FIG. 10. Sequence from a film showing development of a weak thermal explosion. Copper in water. $T_{\mathrm{bott}} \sim 1300°\mathrm{C}$, $T_o = 19{\cdot}5°\mathrm{C}$, $m_{\mathrm{Cu}} \cong 0{\cdot}5\,\mathrm{g}$. Sample No. 8–03.

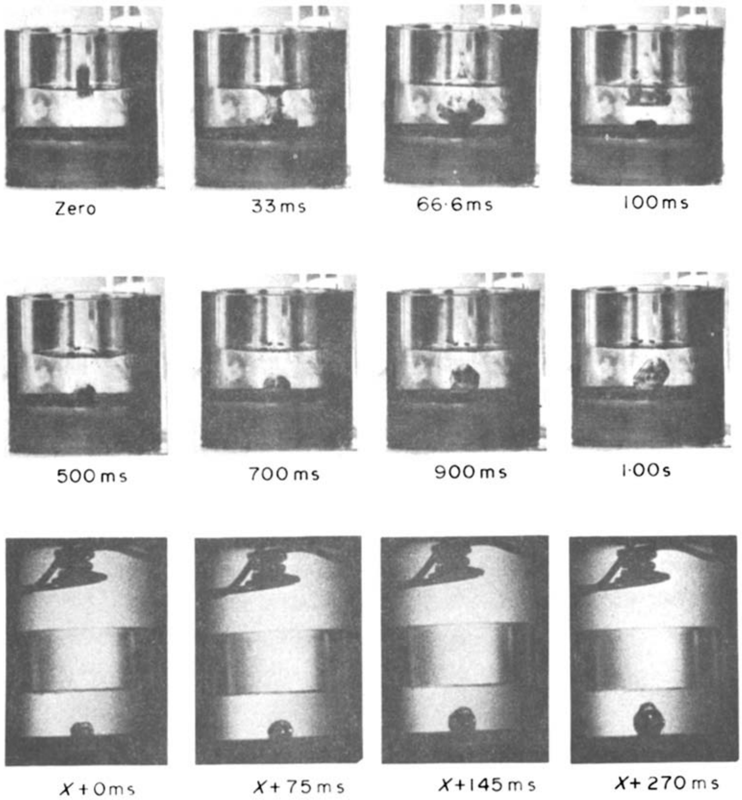


FIG. 11. Sequence from the frame camera film and high speed film showing growth of the "empty shell" particle. Copper in water. $T_{bott} = 1467°C$, $T_o = 21·3°C$, $m_{Cu} = 0·5482$ g. Sample No. 13–36.

phase ($q''_{LW} \sim 1$–$5\ 10^6$ W/m^2, the range PW in Fig. 5). This value stays almost constant during this boiling regime.

The violent nucleate boiling is followed by quiet nucleate boiling when the temperature decreases to about 120°C and next by pure natural convection of the nonboiling water. The heat flux ceases quietly to zero.

The measurements with water up to 80°C demonstrate the same character of the phenomena [22]. The observed thermal explosion has however some lower destructive energy, but it takes place under similar conditions as in the case of water at 20°C.

DISCUSSION OF MEASUREMENTS

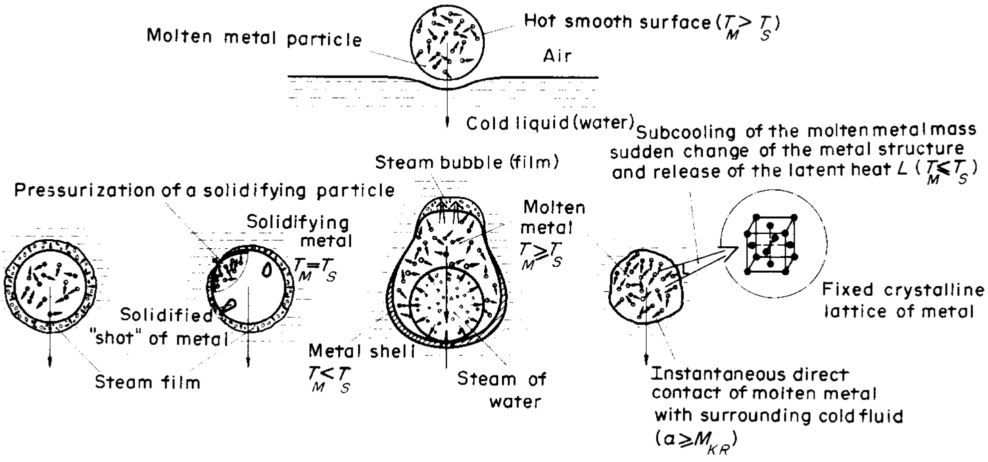
The experiments, performed so far, have shown several possible modes of heat transfer which may appear during the thermal interaction of the molten metal with cold water. The most destructive of them and at the same time not explained completely is the thermal explosion. Some of the effects mentioned in the introduction to the paper are observed in these experiments.

The entrainment and entrapment of cold water are indeed observed in these experiments (particularly with Al). But on the sequence of photographs of the thermal explosion below the water surface no such effects have been observed before the event. Hence the particles only flatten out, but the size of them remain constant.

Because of the large surface tension and small size of the examined particles, the entrapment of the water is not very likely. Thus, the hypothesis based on these effects cannot explain the thermal explosion at a short distance below the water surface.

The hydrodynamic fragmentation and the liquid surface instability are observed in some situations but they do not explain, e.g. the "shots" of the liquid metal and the thermal explosion at the bottom of the vessel.

The transplosion phenomenon (and connected violent boiling theory) does not explain the delay between the direct contact of the molten metal particle with cold water and the thermal explosion which is observed on the high speed films. The transplosion occurs indeed rapidly and is really connected with some pressure pulse but the observation of the thermal explosion sequence suggests more "intrinsic" character of fragmentation.* The spontaneous nucleation hypothesis [17] can probably explain the thermal explosion below the water surface but does not explain the thermal explosion at the bottom of the vessel. Namely, the vapor film around the particle contains air, and it is unlikely that after the film collapse, no air nuclei remain and that the necessary water superheating is possible.



1. Quiet film boiling.
2. Local direct contact of cold water with molten metal. "Shots" of molten metal.
3. Origination of "empty shell" or "fungus-form" particle.
4. Thermal explosion after direct contact of all (or almost all) surface of the molten particle with cold liquid (1°, $T_M \leqslant T_S$, 2°, $\alpha \geqslant M_{kr}$).

FIG. 12. Various possible modes of heat transfer during interaction of molten copper with cold water. T_M, T_S—temperature of the metal particle, and solidification temperature, respectively; α—heat-transfer coefficient, M_{kr}—a large value of α.

The liquid surface instability phenomenon can cause transplosion of the vapor film and leads to the direct contact of both hot and cold liquids, but the existence of the delay between the event and the thermal explosion as well as such effects as the "shots" of small metal particles from the main one and the origination of the "fungus-form" particle are still not explained.

The pressurization of the molten metal in a shrinking shell and the induced stresses can cause the ruptures of the shell and jetting of the small metal particles into the surrounding coolant. In these experiments in air atmosphere such a pressurization can be explained by the existence of the copper oxide in the surface layer of the metal particle. The examination of the high speed films and temperature measurements show that these "shots" appear in the temperature range between about 1250°C and the solidification point of copper. Only small "shots" and no thermal explosion was observed for copper–water interaction in these experiments when an inert atmosphere (Ar or N_2) was used. This supports the pressurization model.

Also, the origination of the "empty shell" or "fungus-form" particle can be explained by the expansion effect of the water steam in the partly solidified shell. Water could be sucked into the shell through one of the ruptures in the metal shell (Fig. 12).

But on the other hand, the oxide layer on the surface of the particle has the lower thermal expansion coefficient than copper itself and this contradicts the facts mentioned above. However, the temperature gradients in a thin surface layer of the metal particle should be examined in detail. Additionally, preliminary experiments of copper–sodium interaction by use of this method in inert gas atmosphere show intensive fragmentation of the copper particle with limited expulsion of sodium from the experimental vessel.

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*It is suggested [16], that some resonance fragmentation mechanism connected with the pulsation frequency of the vapor film exists, but in Figs. 8 and 10 no vapor film can be seen before the thermal explosion.

These observations indicate that the thermal explosion is caused by some internal process in the molten metal during the rapid solidification of the molten metal mass in a relatively cold liquid. The heat transfer at the surface of the molten metal mass should be sufficiently high ($\alpha \geqslant M_{kr}$) to subcool this metal below the solidification point. In such a case, when rapidly produced latent heat of fusion in a confined space cannot be transferred smoothly, the "swelling out" forces originate which involve the fragmentation of the metal. It should be noticed that if the molten metal mass is subcooled then both substances hot and cold exist in an unstable condition.

The geometry of the reaction zone can have such a thermohydrodynamic characteristic that the direct contact of both liquids is possible only for some defined set of the thermohydrodynamic parameters. One of these may be the "thermal explosion temperature", as it is in these experiments.

The direct contact of the molten metal with water can originate, e.g. by the entry into the water or by the collapse of the vapor film at the surface of the molten metal. Both such situations are observed in these experiments.

The "direct contact" between both liquids when the temperature of the "hot" one exceeds the critical temperature of the "cold" one should be examined in further experiments. The "shots" of metal from the main particle and the formation of the "bridges" through the vapor film can produce such a contact. Also, the liquid surface instability and the following local changes of the heat-transfer intensity at the metal surface can favor such phenomena.

The solidification of a substance is attended by changes of the physical properties such as the specific volume, the specific heat, the thermal conductivity and others, and by release of the latent heat of fusion. The unconstrained, disorder position of atoms in liquid state changes during the solidification. The atoms are arranged in the form of the fixed crystalline structure (Fig. 12).

We can distinguish the "slow" and "fast" solidification [4]. The slow solidification process characterizes by the continuous growth of the solid phase and quiet change to the crystalline structure of the body considered [20].

The fast solidification may be connected with a subcooling of the liquid metal mass.

The internal process which initiates the fragmentation of the metal, can be the abrupt change of the crystalline structure during the solidification of the molten metal.

After fragmentation of the metal comes to rapid vaporization of the surrounding cold liquid, and the thermal explosion appears.

The delay between the "triggering shot" and the vapor explosion supports this hypothesis.

The local thermal explosion can also cause further destruction of the vapor film when it is destroyed at one point on the surface of the molten metal mass, and this may trigger the full thermal explosion.

CONCLUSIONS

The measurement techniques used, permit an examination of the interaction of high temperature materials with a cold liquid. The transient heat flux and heat transfer coefficients can be derived from such measurements.

According to the hypothesis proposed in this paper, the thermal explosion occurs when the molten metal solidifies and some internal mechanism in the metal itself causes the fragmentation of this metal. Such mechanism of fragmentation may follow, e.g. sufficiently high heat transfer at the surface of the molten metal mass. Structural changes and occurring of the intercrystal forces can cause stresses which initiate this fragmentation process. Increasing of the contact surface between the metal and the superheated water which is connected with the fragmentation of the metal, initiates the vapor explosion.

Further experimental and theoretical works to prove this hypothesis would be useful.

To explore the mechanism of the thermal explosion in a fast reactor, the tests with liquid sodium and molten structural materials and reactor fuel (stainless steel, UO_2, etc.), similar to that described above, are in preparation.

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APPENDIX

The copper particle, heated in the levitation coil oxides on the surface of the metal when heated above its melting point. The thickness of the oxide surface layer and an amount of oxide (Cu_2O at the inner and CuO at the outer zone) depends on the atmosphere, temperature and time of heating. The chemical analysis of the metal particle after thermal interaction in water shows the following weight percentages of the oxygen depending on the surrounding atmosphere during the heating phase: in argon ~0·04 per cent, in nitrogen ~0·05 per cent and about 2 per cent in air (~1·85 per cent for the levitation time $\tau_{lev} = 5{\cdot}6$ s, and 2·12 per cent for $\tau_{lev} = 9{\cdot}6$ s).

The copper oxide has a higher melting point as copper itself, namely

$$T_{S/CuO} = 1336°C \quad \text{or} \quad 1447°C, \; T_{S/Cu_2O} = 1235°C. \tag{23}$$

The temperature history of solidifying particles shows that in the range *CE* (Fig. 4), the solidification proceeds at the variable temperature. The percentage of the particle mass which has the variable solidification point can be calculated as follows:

$$u_{CE} = c_p(D_{BC} - D_{CE})\tau_{CE}/L_m \tag{A.1}$$

where L_m is an average value of the latent heat of fusion between 49·2 cal/g for copper and 94·2 cal/g for Cu_2O. The values of u_{CE} reach ~30 per cent (e.g. for the test 10–05, $u_{CE} \simeq 26$ per cent) and show a variation of the percentage of the copper oxide in the surface layer of the copper particle.

Assuming a spheric form of the copper particle, a linear dependence of the copper oxide percentage along the particle radius beginning from some intermediate value $x = r_1$ of this radius (or dimensionless beginning from $\xi = \zeta$ where $0 < \zeta < 1$, Fig. 13) and furthermore assuming that all the oxygen is in Cu_2O, the formula for the mass of the copper oxide in the particle follows:

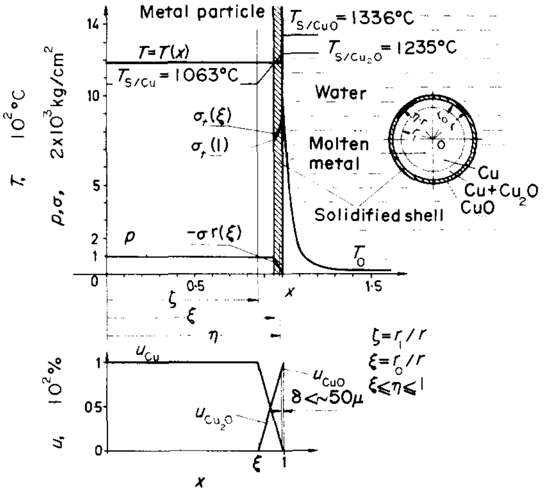

FIG. 13. Origination of metal shell on the surface of the molten metal mass. $T = T(x)$—temperature distribution, u_i—percentage of Cu, Cu_2O, CuO, respectively, p, σ_r, σ_t—pressure induced in liquid phase inside the solid shell, nedial and tangential stress, respectively.

$$m_{Cu_2O} = \frac{\omega}{1-\zeta}\int_{\zeta}^{1} x^2(x-\zeta)\,dx \tag{A.2}$$

where

$$\omega = 4\Pi r^3 \sigma_{Cu_2O}.$$

The mass m_{Cu_2O} can be calculated from the percentage of the oxygen in the particle and the formula (A.2) can be used to obtain the radius ζ:

$$\zeta = \sqrt[3]{[-\phi + \sqrt{(\phi^2 + \psi^3)}]} + \sqrt{[-\phi - \sqrt{(\phi^2 + \psi^3)}]} + 1/9 \tag{A.3}$$

where

$$\psi = 2/9$$
$$\phi = -44/27 + 2m_{Cu_2O}/3\omega.$$

The numerical values calculated by means of both formulae (A.1) and (A.3) are in good agreement, e.g. for the 10–05 by means of the formula (A.3) $\zeta \simeq 0{\cdot}9$ and $u_{CE} \simeq 23$ per cent.

The pressurization effect in the shrinking shell can be approximately calculated as follows [10]:

$$p = \frac{2\Delta\alpha_t E}{1+2\xi-\mu(4\xi^3-1)}(1-\xi^3)\Delta T$$

$$\sigma_r = p\frac{\xi^3}{\xi^3-1}\cdot\frac{1-\eta^3}{\eta^3}$$

$$\sigma_t = -p\frac{\xi^3}{\xi^3-1}\,\frac{2\eta^3+1}{2\eta^3}$$

where: ΔT—sudden change of the particle temperature.

Assuming hypothetically, the value $\Delta\alpha_t = 10^{-6}$ 1/deg and $\Delta T = 10°C$ the curves shown in Fig. 13 are obtained. It should be, however, noticed that the calculation model is based on thermoelastic considerations in the solidified shell which for the temperatures near the solidification point and for such extreme stresses is not valid. The model does not also regard exactly the conditions at the metal particle boundary. Additionally, the difference $\Delta\alpha_t$ between Cu and Cu_2O has a negative sign and it should be proved when such a pressurization effect can be observed in the reality, e.g. between CuO and Cu_2O.

INTERACTION THERMIQUE DU CUIVRE FONDU ET DE L'EAU

Résumé—Un travail expérimental étudie l'intéraction thermique entre des particules de cuivre fondu (dans le domaine de température compris entre le point de fusion et 1800°C) et de l'eau entre 15 et 80°C.

Les températures des particules de cuivre et de l'eau sont mesurées avant et pendant leur interaction thermique. On filme l'histoire du phénomène au moyen d'une caméra à grande vitesse FASTAX (jusqu'à 8000 images/s). On en déduit une classification des phénomènes observés et une description des modes de transfert thermique.

Un phénomène parmi les autres est l'explosion thermique. On discute les conditions nécessaires de l'explosion thermique et on donne leur interprétation physique.

Selon l'hypothèse proposée dans ce texte, l'explosion thermique a lieu lorsque le métal fondu atteint la température de solidification et quand le transfert thermique à la surface est suffisamment intense. Le "changement brutal" de la structure cristalline durant la solidification du métal fondu est la cause de l'explosion par fragmentation.

THERMISCHE WECHSELWIRKUNG GESCHMOLZENEN KUPFERS MIT WASSER

Zusammenfassung—Die experimentelle Arbeit wurde durchgeführt, um die thermische Wechselwirkung zwischen geschmolzenen Kupferpartikeln (im Temperaturbereich zwischen dem Kupferschmelzpunkt und über 1800°C) und Wasser zwischen 15°C und 80°C zu untersuchen. Das zeitliche Temperaturverhalten der Kupferpartikel und des Wassers vor und während ihrer thermischen Wechselwirkung wurde gemessen. Der zeitliche Ablauf wurde mit einer Hochgeschwindigkeitskamera (FASTAX) gefilmt (bis 8000 Bilder/s). Eine Einteilung der beobachteten Phänomene und eine Beschreibung der Wärmeübergangstypen wurde erreicht. Eines der Phänomene war die thermische Explosion. Die dafür notwendigen Bedingungen werden diskutiert und physikalisch interpretiert. Der hier vorgeschlagenen Hypothese zufolge tritt die thermische Explosion auf, wenn das geschmolzene Metall seine Restarrungstemperatur hat und der Wärmeübergang an seiner Oberfläche genügend intensiv ist. Der plötzliche Wechsel der Kristallstruktur während der Erstarrung des geschmolzenen Metalls ist die Ursache der explosiven Zersplitterung.

ТЕПЛОВОЕ ВЗАИМОДЕЙСТВИЕ РАСПЛАВЛЕННОЙ МЕДИ С ВОДОЙ

Аннотация — Проведено экспериментальное исследование теплового взаимодействия расплавленной меди (в диапазоне температур от точки плавления меди до 1800°С) и воды при температуре от 15 до 80°С.

Проводились измерения неустановившихся температур частиц меди и воды до и во время их теплового взаимодействия. Весь процесс был снят на пленку с помощью высокоскоростной камеры FASTAX (до 8000 фут/сек). Проведена классификация наблюдаемых явлений, и сделано описание видов теплопереноса.

Обсуждаются необходимые условия для одного из наблюдаемых явлений, теплового взрыва, и дается их физическая интерпретация.

Согласно предлагаемой гипотезе тепловой взрыв происходит при температуре затвердевания расплавленного металла и достаточно интенсивном теплообмене на поверхности металла. Быстрая кристаллизация при затвердевании расплавленного металла является причиной взрывного диспергирования.