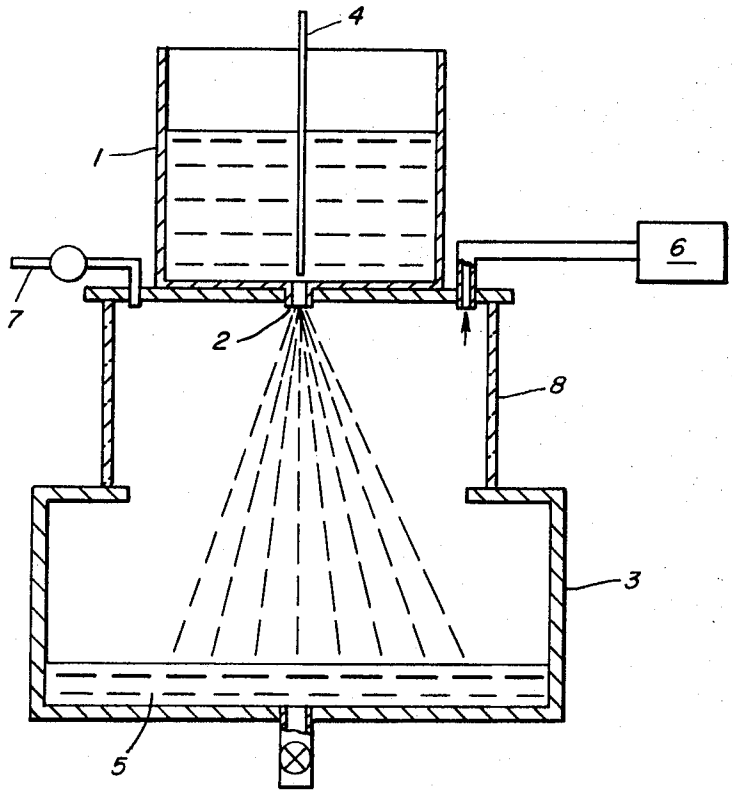
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ATOMIZATION OF LIQUID MATERIALS AND THE SUBSEQUENT QUENCHING THEREOF

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INVENTORS.
ROBERT G. OLSSON &
ETHEM T. TURKDOGAN
By Alf Greif
Attorney

United States Patent Office

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ATOMIZATION OF LIQUID MATERIALS AND THE SUBSEQUENT QUENCHING THEREOF

Robert G. Olsson, Edgewood Borough, and Ethem T. Turkdogan, Pittsburgh, Pa., assignors to United States Steel Corporation

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ABSTRACT OF THE DISCLOSURE

A method for producing both metallic and non-metallic droplets, by causing a stream of liquid entering a chamber of lower pressure to form a spray of fine droplets. The desired material is converted to a liquid, such as by melting or by solvation. The liquid is forced through an orifice into the chamber, wherein prior to its entrance in the chamber, fine bubbles of gas are entrained within the liquid. After the entry into the chamber, the individual bubbles expand radially, causing the liquid stream to be broken into sheets of extremely fine droplets, which are subsequently quenched to form powders, or agglomerated to form sheet or slab material.

Finely divided powders have numerous industrial and practical applications. Thus, metal powders are employed in powder metallurgy for the production of compacts of intricate shapes, as well as roled sheet products. These powders are also utilized in pyrotechnics and as solid fuel components. They may be oxidized or otherwise reacted to form high surface area catalysts. Resin or plastic powders are similarly employed in the formation of compacts and as insulating fillers.

A number of methods have been proposed for the conversion of liquid materials into fine droplets to produce powders. These methods often require intricate mechanical devices, and are difficult to control. Thus, in atomization processes, the liquid is aspirated through a nozzle into a high pressure jet of gas, and blown into a collecting chamber. Shot particles are also formed by passing the molten material through openings in vibrating or rotating screens. In both methods, clogging of the openings is a serious problem.

Resin powders have also been formed by causing the liquid resin, having an expansion agent dissolved therein, to flow from a pressurized chamber through a passage to a zone of lower pressure. Upon release of the pressure, the normally gaseous expansion agent vaporizes and expands the resin to form a spray of droplets. Recently, metal powders have similarly been formed (U.S. Pat. No. 3,510,546) by dissolution of a high vapor pressure material in the metal bath or by saturation of the bath with an absorbed gas, prior to its release into the chamber of reduced pressure. The instant invention is directed to an improvement of these latter expansion processes, utilizing a simplified method which provides a degree of control and versatility not otherwise obtainable. The instant method totally eliminates the time-consuming and expensive requirement for saturating the molten bath with the expansion gas, prior to discharge of the liquid into the low pressure region. This invention is based on the discovery that a relatively small amount of gas, in the form of minute bubbles which can be entrained in the liquid just prior to its being discharged in the region of lower pressure, will break the liquid stream into a spray of fine droplets even when there is no dissolved gas evolution from the liquid. The degree of liquid stream expansion and hence droplet size, may be controlled with

comparatively minor variations of for example, gas injection rate, bubble size and pressure ratios.

The so formed liquid droplets may then be quenched in a fluid medium to form powders; or they may be employed to strike a substrate and be quenched thereby, so as to produce a casting of sheet or plate material. In the latter embodiment, the substrate may be employed solely as a quench plate from which the cast material is then separated, or it may be in the form of a sheet or plate article which is coated by the molten droplets.

These and other objects and advantages of the instant invention will be better understood by reference to the following description and figure, in which:

The Figure is a schematic diagram of the apparatus which was employed to carry out the method of this invention.

Initial experiments on droplet formation were performed employing Wood's metal, a low melting point alloy (≈72° C.), containing 50% Bi, 25% Pb, 12.5% Sn and 12.5% Sb. With reference to the Figure, the molten metal flowed from the reservoir **1**, through nozzle **2**, into the vacuum tank **3**. Helium gas was injected into the metal at a metered rate through a hypodermic needle **4** (other experiments employed a porous plug) at a point 0.3 cm. above the orifice. The thus formed spray droplets were quenched in chilled, low vapor pressure oil **5** at the base of the tank and were subsequently sized in U.S. Standard Sieve Series. Prior to each experiment, the oil was chilled to about 0° C. with dry ice and the nozzle **2** sealed with a thin coating of Wood's metal. The chamber **3** was evacuated with a two-stage vacuum pump **6**; the desired pressure being maintained with metered gas bleed **7** (in this case, also He) directed into the vacuum chamber. The metal was melted in a separate vessel and at the beginning of a run, about 14 kg. of molten metal was poured into the reservoir. Within a few seconds, the nozzle plug melted through and a stream formed. The average liquid flow rate was about 3.1 liters per minute with a head of 12 cm. Temperature rise of the quench oil, during the about 28 seconds duration of each run, was of the order of 4° C. The upper portion **8** of the vacuum chamber was a 6 x 8 inch diameter glass port, through which the sprays were observed and photographed. When seen in normal light, the liquid spray appears to be a continuous shower of droplets, however, when photographed with a high-speed motion picture camera, it was seen that the spray was discontinuous. Still photographs with a strobe light and high-speed motion pictures provided the basis for understanding of what was, in fact, occurring. As an entrained gas bubble leaves the nozzle, it forms a bulge in the stream of metal directly below the opening. The gas in the bulge rapidly expands, and breaks from the stream, a discrete segment resembling a spherical cap, which subsequently expands in uneven sheets; this action taking place within several nozzle-opening diameters. Beneath this point, the sheet continues to expand and to break into droplets along its outer edge and at the thinner areas. Finally, the rest of the sheet disintegrates into small radii droplets. These experiments have indicated that the axial velocity of the spray droplets, u, may be represented by the following equation:

$$u=2\sqrt{\frac{P_1-P_2}{e}+gz}$$

where P_1 and P_2 are the liquid and vacuum chamber pressures respectively, e is liquid density, g, the gravitational constant, and z, the liquid head. It may, therefore, be seen that axial velocity of the spray droplets is primarily a function of the potential energy of the liquid entering the vacuum chamber and is only slightly affected

by the expanding gas phase. Consequently, the useful work from the expansion of the gas bubbles is substantially expended in the breaking and radial spread of the stream and the creation of new surface.

Analysis of the results of the above experiments as well as experiments with other molten metals and liquids have demonstrated that the following parameters are important in the control and size distribution of spray droplets.

Rate of gas injection. The peak in the size distribution of particles decreases as the injection rate is increased. Thus, smaller particles, i.e., larger surface area particles, may be obtained by increasing the injection rate for a constant bubble size. However, as gas injection rate is increased, the efficiency (i.e., the percent of maximum energy for the expansion of gas bubbles that is transferred to the liquid in the form of creating new surface and in radial kinetic energy) of spray formation decreases. Of course, if the principal objective were increased surface and if economics of gas utilization were of little concern, then very high gas injection rates could be employed to obtain very finely distributed particles. However, once a spray is established, an increase gas injection rate results in a disproportionately smaller increase in product surface area, since the gas tends to be utilized in the acceleration of the droplets in the axial direction as well. Aside from efficiency, the rate of injection should always be below that at which the bubbles join together to form a continuous stream of gas.

Size of injected bubbles (with respect to stream inlet orifice). In general, a decrease in bubble size will shift the product distribution to smaller, and hence large surface area particles. For maximum efficiency, there is, however, an optimum range of bubble sizes, depending on the surface tension of the molten material. If the bubbles are too small, they will expend their energy in overcoming surface tension forces, rather than in the disintegration of the sheets of liquid. On the other hand, too large bubbles tend to burst prematurely, prior to transferring their energy to radial expansion. It was noticed in the above experiments, that when the bubbles were exceedingly large, this premature bursting produced slugs of solidified metal which had not been disintegrated. In all instances, the bubble diameter should be substantially smaller than the effective diameter of the nozzle opening so as to provide a liquid matrix with a discontinuous gas phase entrained therein.

Ratio of pressures, P_1/P_2. Decreasing the pressure in the chamber in relation to the pressure of the liquid in the reservoir tank will result in a decrease in mean particle size, i.e., larger surface area. It may, therefore, be seen that very low chamber pressures, P_2, are not necessary for good spray formation. Thus, in the experiments in which P_1 was atmospheric pressure, chamber pressures of 0.5, 5 and 50 torr provided respectively, maximum expansion work of 95%, 87% and 67% of the value for a theoretical zero chamber pressure. Thus, in most cases, little is to be gained in striving for chamber pressures less than several torr. It may also be seen that when oxidation of the droplets in the chamber is not a critical factor, (as in the case of resinous materials), it may be more economical to increase the pressure of the liquid in the reservoir, P_1, rather than extensively reducing the pressure in the chamber, P_2.

The enhanced disintegration of the liquid, by utilization of the instant process, is effected by causing the liquid (with its entrained bubbles) in the vessel in which it is contained to be suddenly accelerated, as it passes through a nozzle opening of substantially reduced cross-sectional area and into the zone of reduced pressure. It is therefore preferred, that the ratio of the cross-sectional area of the vessel 1, to the cross-sectional area of the nozzle opening be at least 2:1. With this acceleration of the liquid, there is a corresponding sudden drop in pressure from that at the base of the vessel to that of the reduced pressure zone. Entrained gas bubbles are rapidly taken from the high pressure region to the low pressure region where bubble expansion results in the radial spread of the liquid stream and an ensuing spray of fine, large surface area droplets.

To achieve an efficient utilization of gas bubbles for such expansion, the gas should be injected at a point wherein the liquid is at a pressure considerably higher than that of the reduced pressure zone. Thus, the gas may be injected at any point within the containment vessel above the nozzle, where the pressure within the bubble will be approximately that of the surrounding liquid. However, it is most efficiently injected at a point sufficiently proximate to the orifice (as depicted) so as to cause a substantial portion of the bubbles to be entrained within the increment of liquid entering the reduced pressure zone.

The length of the nozzle should be short, (for purposes of this invention, the term "nozzle" is considered to be a device for converting the pressure and potential energy of a fluid into kinetic energy and will therefore also include a knife edge orifice), so that the passage of liquid with its entrained bubbles will be sufficiently rapid, thereby preventing the bubbles from expanding to a substantial extent prior to the time the liquid has left the nozzle. Consequently, substantially all the gaseous expansion will be available for spray formation. The maximum nozzle length will depend on a number of factors. Thus, the diminished expansion caused by a relatively long nozzle could be overcome to some extent by increasing the ambient pressure of the liquid and/or by increasing the force by which the liquid is forced through the nozzle. These latter expedients serve to increase the velocity of liquid through the nozzle and to increase the initial stored energy in the bubbles. Thus, to prevent such substantial expansion of the bubbles within the nozzle, with its consequent loss of available energy, it is considered desirable to limit the residence time of the incremental portion of liquid within the nozzle. This time limit is basically a function of the pressure of the liquid at the point of gas injection (and hence the pressure inside the bubbles prior to any expansion). It is, therefore, preferred that the liquid be urged through the nozzle with sufficient force so that the residence time of the liquid within the nozzle is defined by the equation:

$$t < 0.1\sqrt{P_1}$$

where

t=residence time in seconds, and

P_1=pressure in atmopheres, of the liquid, at the point of gas injection.

To achieve such desirable short residence times, the liquid is preferably accelerated through the nozzle orifice by the application of a force in addition to, and acting in concert with the atmospheric force. Thus, in the Figure, the head of liquid (gravity) and the atmosphere both aid in forcing the liquid through the nozzle. However, the instant method need not rely on gravitational (i.e. downward flow) or atmospheric forces. Thus, for example, a piston device (not shown) may be employed to force the liquid into a region of reduced pressure.

Since the disintegration of the liquid stream is not dependent on saturating or dissolving a gas within the liquid, any "non-deleterious" gas may be injected for purposes of spray formation. Thus, in addition to argon, helium or other gases classified as "inert," any gas e.g., nitrogen, may be employed which does not react detrimentally with the liquid being treated. Thus, in many instances (i.e. resinous materials or glasses), inexpensive air injection may be employed.

We claim:

1. A method for the disintegration of a liquid material into particulate form and the production of powders thereby, which comprises:

passing the liquid from the vessel in which it is contained, through a nozzle and into a zone maintained

at a pressure substantially lower than the pressure of the liquid within said vessel, the cross-sectional area of said nozzle being less than 0.5 the cross-sectional area of said containment vessel;

injecting a non-deleterious gas into said liquid prior to its passage through said nozzle, at a point wherein the liquid is at a pressure substantially higher than that of said lower pressure zone, said gas being injected at a rate by which spaced apart bubbles are formed, said point of injection being sufficiently proximate said nozzle so as to cause a substantial portion of said bubbles to be entrained within the increment of liquid entering said nozzle, said bubbles being of a diameter substantially smaller than the effective diameter of the nozzle opening and

causing the passage of liquid through said nozzle to be sufficiently rapid so that those bubbles, entrained within the increment of liquid entering said nozzle, are prevented from expanding to a substantial extent prior to the time the liquid has left the nozzle, whereby the resultant expansion of said bubbles in said zone of lower pressure supplies a major portion of the energy for the disintegration of the liquid material into fine droplets, and quenching said droplets in a fluid medium to produce solid particles.

2. The method of claim **1**, in which said liquid is urged through said nozzle, by the application of a force acting in addiiton to and in concert with the force exerted by the atmosphere ambient to the liquid in said containment vessel.

3. The method of claim **2**, wherein said liquid flows through said nozzle in a downward direction.

4. The method of claim **3**, wherein said liquid material in said containment vessel is substantially devoid of dissolved gases.

5. The method of claim **4**, wherein said mean particle size may be decreased by decreasing the diameter of said bubbles.

6. The method of claim **3**, wherein said liquid material is a metal.

7. The method of claim **6**, wherein said gas is substantially insoluble in, and non-reactive with said metal.

8. The method of claim **7**, wherein said gas is selected from the group consisting of "inert" gases and nitrogen.

9. A method for the disintegration of a liquid material into particulate form and the coating of a substrate thereby, which comprises:

passing the liquid from the vessel in which it is contained, through a nozzle and into a zone maintained at a pressure substantially lower than the pressure of the liquid within said vessel, the cross-sectional area of said nozzle being less than 0.5 the cross-sectional area of said containment vessel;

injecting a non-deleterious gas into said liquid prior to its passage through said nozzle, at a point wherein the liquid is at a pressure substantially higher than that of said lower pressure zone, said gas being injected at a rate by which spaced apart bubbles are formed, said point of injection being sufficiently proximate said nozzle so as to cause a substantial portion of said bubbles to be entrained within the increment of liquid entering said nozzle, said bubbles being of a diameter substantially smaller than the effective diameter of the nozzle opening; and

causing the passage of liquid through said nozzle to be sufficiently rapid so that those bubbles, entrained within the increment of liquid entering said nozzle, are prevented from expanding to a substantial extent prior to the time the liquid has left the nozzle, whereby the resultant expansion of said bubbles in said zone of lower pressure supplies a major portion of the energy for the disintegration of the liquid material into fine droplets, and quenching said droplets on a substrate to produce a casting of sheet material thereon.

10. The method of claim **9**, wherein said liquid is urged through said nozzle, by the application of a force acting in adidtion to and in concert with the force exerted by the atmosphere ambient to the liquid in said containment vessel.

11. The method of claim **10**, wherein said liquid flows through said nozzle in a downward direction.

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ROBERT F. WHITE, Primary Examiner

J. R. HALL, Assistant Examiner

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