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(57) Abstract: The present invention relates to a method and an apparatus for forming flakes, especially metal flakes. The method comprises producing a heated stream of molten material, feeding the stream in a substantially vertically downward direction, receiving the downwardly directed stream and forming flakes therefrom, and effecting a change in the temperature of the stream subsequent to the production thereof whereby flakes of a desired thickness are obtained. The present invention is applicable to any metal which melts when heated and is capable of being formed into flakes. Examples of metals are Al, Cu, Mo, V, Ag, Cr, Zr, Nb, Ni, Fe, Co, Ti, Au, Pd, W, Hf, Rh, Ir, Pt, Cd or alloys thereof, such as chromium-nickel, iron-nickel, iron-chromium and nickel-cobalt, wherein Cu, Ag, Ti, or Al, or alloys thereof are preferred and Al, or Ag, or alloys thereof are most preferred.
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Apparatus and Method for Producing flakes

The present invention relates to a method and an apparatus for forming flakes, especially metal flakes. The present invention is applicable to any metal which melts when heated and is capable of being formed into flakes. Examples of metals are Al, Cu, Mo, V, Ag, Cr, Zr, Nb, Ni, Fe, Co, Ti, Au, Pd, W, Hf, Rh, Ir, Pt, Cd or alloys thereof, such as chromium-nickel, iron-nickel, iron-chromium and nickel-cobalt, wherein Cu, Ag, Ti, or Al, or alloys thereof are preferred and Al, or Ag, or alloys thereof are most preferred.

EP-A-289240 discloses an apparatus for forming flakes of material from a heated stream of molten material. The apparatus comprises means for feeding the stream in a downwards direction into a rotating cup, the cup being arranged with its open mouth facing upwardly such that molten material within the cup is caused to flow over the upper edges of the cup and flow outwards in a radial direction due to centrifugal force. The apparatus also includes a pair of spaced apart substantially parallel plates arranged about the cup such that the material leaving the cup by centrifugal force passes through a gap defined between the plates. The plates are mounted within a vacuum chamber arranged such that a vacuum is applied to the space between the plates to draw air from outside the chamber between the plates in a radial direction to prevent the molten material from touching the sides of the plates and to cool material until it reaches a solid state pulling the material in a radial direction thereby keeping the material in the form of a flat film and breaking it into small platelets.

According to the present invention there is provided an apparatus for forming flakes, especially metal flakes comprising means for producing a heated stream of molten material, means for feeding the stream in a substantially vertically downward direction, means for receiving the downwardly directed stream and for forming flakes therefrom, and means for effecting a change in the temperature of the stream subsequent to the production thereof whereby flakes of a desired thickness are obtained.

The present invention is applicable to any material which melts when heated and is capable of being formed into flakes. Such materials can be divided into metals and non-metallic materials. Examples of metals are Al, Cu, Mo, V, Ag, Cr, Zr, Nb, Ni, Fe, Co, Ti, Au, Pd, W, Hf, Rh, Ir, Pt, Cd or alloys thereof, such as chromium-nickel, iron-nickel, iron-chromium and nickel-cobalt, wherein Cu, Ag, Ti, or Al, or alloys thereof are preferred and Al, or Ag, or alloys thereof are most preferred.
The thickness of the flakes is in the range of 10 to 300 nm, especially 30 to 200 nm.

According to the present invention the term "aluminum" comprises aluminum and alloys of aluminum. Alloys of aluminum are, for example, described in G. Wassermann in Ullmanns Enzyklopädie der Industriellen Chemie, 4. Auflage, Verlag Chemie, Weinheim, Band 7, S. 281 to 292. Especially suitable are the corrosion stable aluminum alloys described on page 10 to 12 of WO00/12634, which comprise besides aluminum silicon, magnesium, manganese, copper, zinc, nickel, vanadium, lead, antimony, tin, cadmium, bismuth, titanium, chromium and/or iron in amounts of less than 20 % by weight, preferably less than 10 % by weight.

Highly lustrous gold colored flakes contain, for example, 1 to 49 % by weight aluminum and optionally 0.1 to 6 % by weight silicon besides copper.

It is also possible for the finished aluminum flakes to be subjected to after-coating or after-treatment, which further increases the stability to light, weathering and chemicals or facilitates handling of the pigment, especially incorporation into various media. The processes described in EP-A-477433, EP-A-826745 or EP-A-1084198, for example, are suitable as after-treatment or after-coating.


In order to be able to use the aluminium flakes (flake-form aluminium) in aqueous compositions, it is necessary for those pigments to be protected against corrosion by water. According to R. Besold, Aluminiumpigmente fürwassrige Beschichtungen - Widerspruch oder Wirklichkeit?, Farbe + Lack 97 (1991) 311 - 314, a large number of procedures, which can be divided into two groups, are known for the stabilisation of aluminium pigments:

- adsorption of corrosion inhibitors on the pigment surface
- dimeric acids: DE-A-3002175, and
- encapsulation of the pigments with a continuous inorganic protective layer:
  - Fe₂O₃: DE-A-3003352,
  - TiO₂: DE-A-3813335,
- or organic protective layer:

Examples of non-metallic materials are meltable thermoplastic polymers, such as polyolefins, polystyrols, polycarbonates, polyethersulfones, polyesters, polyamides etc; waxes and meltable metal hydroxides, or oxides.

The temperature changing means may be arranged to effect a change of temperature in the stream while it is traveling in a vertically downward direction. Alternatively, the temperature changing means may be arranged to effect a change of temperature in the stream prior to it traveling in a vertically downward direction. Heating can, for example, be made by putting along the stream heaters. Heaters are preferred for polymers, in case of which RF has no effect.

In accordance with the present invention the apparatus includes means for applying a high frequency (RF) current to the vertically downwardly traveling stream.

In another embodiment in accordance with the present invention, means are provided for applying an electric current to the vertically downwardly travelling stream.

The present invention is illustrated in more detail on the basis of metals, but is not limited thereto.

If the metal, such as aluminum, is susceptible to oxidation, the method is conducted under an atmosphere of inert gas and the apparatus comprises means for providing the inert gas atmosphere. The vacuum on the external side of the parallel plate is between 1 and 100 mbar. What is important, if aluminum flakes are produced, is the purity of the inert gas, for example, argon and/or helium. It should be higher than 99 %, preferably higher than 99.9 %.
In a further embodiment in accordance with the present invention, the apparatus is alternatively or additionally provided with means for cooling the stream prior to it being fed in a downward direction. The cooling means may include a conduit through which the stream is fed, said conduit being surrounded by a cooling coil or jacket through which an appropriate cooling fluid, such as an inert gas, may be fed. The effect of cooling the stream within the conduit is to solidify an outer region of the stream in the vicinity of the outlet from the conduit. In this way, the volume mass flow of the flow stream is reduced.

Although this variation in the volume of the flow stream is produced by varying the temperature of at least the outer region of the flow stream, it should be appreciated that variation of the volume of the flow stream represents, in general, an alternative or additional method of controlling the thickness of the resultant flakes.

Accordingly, considered in another aspect, the present invention also provides apparatus as defined above in which in addition to or in substitution of the temperature changing means there are provided mass flow control means. Such control means are typically positioned to effect the mass flow prior to the stream being fed in a vertically downward direction.

The present invention further provides a method for forming flakes, especially metal flakes comprising producing a heated stream of molten material, feeding the stream in a substantially vertically downward direction, receiving the downwardly directed stream and forming flakes therefrom, and effecting a change in the temperature of the stream subsequent to the production thereof whereby metal flakes or a desired thickness are obtained.

The present invention also provides a corresponding method in which the mass or volume flow of the stream is controlled, prior to the stream traveling in a vertically downward direction, in order again to produce flakes of a desired thickness.

The apparatus includes a tank for holding molten metal. Extending from the tank is an outlet conduit or bushing which terminates in an outlet orifice. The stream is found in a conduit from material fed from the tank and the internal diameter of the orifice defines the diameter of a stream of liquid metal at the point where it leaves the conduit and descends vertically from the orifice. The stream exiting from the orifice descends vertically downwards towards a spinning device which may be substantially as described in EP-A-0289240. Indeed the
apparatus includes further components for producing the flake from the liquid stream, which may be substantially as shown and described in EP-A-0289240.

According to EP-A-0289240 a stream of the molten metal is fed in a downwards direction into a rotating cup which has its open mouth facing upwardly with its rim disposed at a horizontal level between a pair of spaced apart parallel plates which are mounted within a vacuum chamber in such manner that, as an inert gas is drawn from outside the chamber between the plates the molten material is drawn radially outwardly without touching the plates and is cooled with the continuing outward movement causing the material to be broken into flakes.

Instead of the inert gas air can be used, if the metal is not susceptible to oxidation. The inert gas or air can also be partly come from openings set in the rotating cup.

The tank may be replaced by a plasma torch, especially an induction plasma torch. The induction plasma torch is equipped with a powder feeder that operates by entraining the particles in a, upward or downward, stream of gas for transport to the plasma induction torch. In addition, it is also possible to inject the particles as a slurry (e.g. organic solvents) into the plasma reactor. This slurry is atomized at the tip of the injection probe.

The plasma torch is preferably an induction plasma torch. The preferred induction plasma torches for use in the process of the present invention are available from Tekna Plasma Systems, Inc. of Sherbrooke, Quebec, Canada. Boulos et al., US-A-5,200,595, is hereby incorporated by reference for its teachings relative to the construction and operation of plasma induction torches.

In a preferred embodiment of the invention the transport gas is inert, i.e. does not react with the outer surfaces of the particles. Typically, the fluidizing gaseous medium is selected to be compatible with the particles, i.e. do not substantially adversely affect the quality of the particles. Examples of such transport gases are argon, nitrogen, helium, oxygen or mixtures such as dry air or argon/hydrogen and argon/oxygen. Generally, gases such as air, nitrogen, argon, helium and the like, can be used, with air being a gas of choice, where no substantial adverse oxidation reaction of the particles takes place.

The apparatus includes a coil which surrounds the stream around about half its length in a central section of the vertically downward path. This coil is suitable for passing a high frequency (RF) current therethrough. The coil is connected to an RF generator which supplies the desired current level.
The high frequency magnetic field generated by the current in the surrounding coil is absorbed in the stream of molten metal flowing downward transmission. A pyrometer measures the temperature of the molten metal and a suitable closed loop control circuit leads to a stabilization of the molten metal temperature.

Another method of directly heating the molten metal stream involves the passing of an electric current through the stream between an upward connection in the form of an electrode connected to the bushing. Such a connection can be achieved by using a bushing made of an electrically conductive material so that the bushing is itself the electrode or, alternatively, positioning an electrode either immediately in front of the bushing within the tank or immediately after it and in contact with the flow stream.

At the other end of the flow stream, electrical connection to the spinning device is made by means of a slip ring attached to the shaft of the spinning device and including static brushes through which the electrical connection is made. Control of the current is by way of a transformer with suitable voltage and current output. Current variation may be achieved by, for instance, thyristor control and an infrared receptor as described above.

In addition or as an alternative to the above described means for heating the metal stream, the apparatus may be provided with means for controlling the mass flow.

These means are provided at the conduit and involve cooling the glass stream emerging from the tank. The conduit is provided with an oversized aperture and is externally clad with a cooling jacket through which cooling fluid may be fed. The jacket may be a simple coil wrapped round the bushing and fed with water or it may be an external annular ring through which compressed air is passed. As the molten metal passes through the bushing, the bushing is cooled and a layer of molten material is solidified within the bushing orifice. This has the effect of reducing the aperture size and thereby reducing the mass flow. Although there is a loss of heat from the flow stream, this is relatively small because the melt steam material is a poor thermal conductor when solidified.

The change in temperature is linear with mass flow and the flow rate can therefore be controlled by monitoring the outflow temperature with an infrared receptor directed at the flow stream immediately below the bushing. This receptor is connected to suitable electronic circuitry to vary the amount of coolant causing solidification within the bushing. Any heat
losses arising from this control method are compensated for by the temperature control methods described above.

The above described methods used either individually or in combination allow fine control of flow streams being fed into the spinning devices such that flakes may be produced with thicknesses below 250 nanometers and with thickness variations as low as 10 per cent.

Reference has been made above to the use of apparatus of the present invention for producing metal flakes. However it should be appreciated that the apparatus may be used for producing flakes of any other appropriate material.
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Claims

1. Apparatus for forming flakes, especially metal flakes comprising means for producing a heated stream of molten material, means for feeding the stream in a substantially vertically downward direction, means for receiving the downwardly directed stream and for forming flakes therefrom, and means for effecting a change in the temperature of the stream subsequent to the production thereof whereby flakes of a desired thickness are obtained.

2. Apparatus according to claim 1, wherein the temperature changing means are arranged to effect a change of temperature in the stream while it is travelling in a vertically downward direction.

3. Apparatus according to claim 1, wherein the temperature changing means may be arranged to effect a change of temperature in the stream prior to it travelling in a vertically downward direction.

4. Apparatus according to any of the preceding claims, wherein the apparatus includes means for applying a high frequency (RF) current, or heat to the vertically downward travelling stream.

5. Apparatus according to any of claims 1 to 3, wherein means are provided for applying an electric current to the vertically downward travelling stream.

6. Apparatus according to any of the preceding claims, wherein the apparatus is alternatively or additionally provided with means for cooling the stream prior to it being fed in a downward direction.

7. Apparatus according to claim 6, wherein the cooling means includes a conduit through which the stream is fed, said conduit being surrounded by a cooling coil or jacket through which an appropriate fluid may be fed.

8. Apparatus according to any of the preceding claims wherein the apparatus is provided with mass or volume flow control means in addition or in substitution of the temperature changing means.

9. A method for forming flakes, especially metal flakes comprising producing a heated
stream of molten material, feeding the stream in a substantially vertically downward direction, receiving the downwardly directed stream and forming flakes therefrom, and effecting a change in the temperature of the stream subsequent to the production thereof whereby flakes of a desired thickness are obtained.

10. A method according to claim 9, wherein, in addition to effecting a change in the temperature of the stream, or in substitution therefor, a change is effected in the mass or volume flow of the stream.

11. Flakes, especially metal flakes obtainable by the method according to claims 9, or 10.
### A. CLASSIFICATION OF SUBJECT MATTER

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According to International Patent Classification (IPC) or to both national classification and IPC

### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

- EPO-Internal
- WPI Data, PAJ, COMPENDEX

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

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<td>US 3 780 153 A (PRIVOTT W ET AL) 18 December 1973 (1973-12-18) column 6, line 21 - line 72; figures 1,2,4-6; examples I-IV</td>
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<td>Y</td>
<td>EP 0 289 240 A1 (GLASSFLAKE LTD [GB]) 2 November 1988 (1988-11-02) claims 1,8,9; figure 1</td>
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Further documents are listed in the continuation of Box C

**X** See patent family annex

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**S** document member of the same patent family

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Authorized officer:

Bombeke, Martin
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<tr>
<td>Y</td>
<td>JP 06 002019 A (JAPAN METALS &amp; CHEM CO LTD) 11 January 1994 (1994-01-11)</td>
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<tr>
<td></td>
<td>abstract; figure 1</td>
<td>11</td>
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<tr>
<td>Y</td>
<td>US 4 082 207 A (GARNIER MARCEL A ET AL) 4 April 1978 (1978-04-04)</td>
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<tr>
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<td>GB 2 275 634 A (ATOMIC ENERGY AUTHORITY UK [GB]) 7 September 1994 (1994-09-07)</td>
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<td>X</td>
<td>FR 2 629 573 A1 (AUBERT &amp; DUVAL ACIERIES [FR]; UNIV CLERMONT FERRAND II [FR])</td>
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<tr>
<td></td>
<td>6 October 1989 (1989-10-06) claims 1-5, 8-10; figures 1, 2</td>
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# INTERNATIONAL SEARCH REPORT

Information on patent family members

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<td>US 3780153 A 18-12-1973 NONE</td>
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<tr>
<td>SU 900989 B 30-01-1982 NONE</td>
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<tr>
<td>JP 6002019 A 11-01-1994 NONE</td>
<td></td>
<td></td>
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<tr>
<td>GB 2275634 A 07-09-1994 NONE</td>
<td></td>
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<tr>
<td>US 4925103 A 15-05-1990 NONE</td>
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<td>FR 2629573 A1 06-10-1989 NONE</td>
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