ABSTRACT

Systems and associated methods for carbothermally producing aluminum are provided, the systems generally including a reactor having a depth such that when the reactor contains molten liquid hydrostatic pressure of the molten liquid is at least about 0.5 atm as measured proximal the bottom of the reactor. A plurality of horizontally disposed electrodes, which may be offset from one another in a vertical and/or horizontal direction, may also be used in accordance with the system to provide selective heating gradients within the molten liquid.
Figure 3

Effect of increased pressure on aluminum alloy production.

Moles of alloy produced vs. pressure (atm).

- 2400 (K)
- 2450 (K)
- 2500 (K)
SYSTEMS AND METHODS FOR CARBOETHERMICALLY PRODUCING ALUMINUM

FIELD OF THE INVENTION

[0001] The present invention relates to systems and methods for carbothermically producing aluminum, specifically using increased pressure.

BACKGROUND OF THE INVENTION

[0002] The direct carbothermic reduction of alumina has been described, for example, in U.S. Pat. No. 2,974,032 to Grunert et al., as having an overall reaction of:

$$\text{Al}_2\text{O}_3+3\text{C} \rightarrow 2\text{Al}+3\text{CO}$$

(1)

This reaction (1) takes place, or can be made to take place, in two steps:

$$2\text{Al}_2\text{O}_3+9\text{C} \rightarrow 2\text{Al}_3\text{C}_3+6\text{CO}$$

(2)

$$\text{Al}_3\text{C}_3+6\text{Al}_2\text{O}_3 \rightarrow 6\text{Al}+3\text{CO}$$

(3)

[0003] Reaction (2) takes place at temperatures below 2000°C and generally between 1900°C and 2000°C. Reaction (3) takes place at appreciably higher temperatures (e.g., above 2600°C). In addition to the species stated in reactions (2) and (3), volatile species including gaseous Al and gaseous aluminum suboxide, that is AlO, may be formed, such as via one or more the following reactions:

$$\text{Al}_2\text{O}_3+2\text{C} \rightarrow 2\text{AlO}+2\text{CO}$$

(4)

$$\text{Al}_2\text{O}_3+3\text{C} \rightarrow 2\text{Al}+3\text{CO}$$

(5)

It is highly desirable to suppress these vapor forming reactions (4), (5) so as to restrict the amount of aluminum lost to vapor during the production process.

[0004] Attempts have been made to suppress these vapor forming reactions. For example, U.S. Pat. No. 3,971,653 to Cochran ("Cochran"), which is incorporated herein by reference in its entirety, discloses that increased vapor pressures may be used to suppress the aluminum vapor forming reactions. Cochran does not disclose methods of suppressing vapor reactions outside of this scope.

SUMMARY OF THE INVENTION

[0005] In view of the foregoing, a broad objective of the present invention is to provide systems and methods for carbothermically producing aluminum with restricted loss of aluminum to vapor off-gas.

[0006] In addressing one or more of the above objectives, the present inventor has recognized that a sufficiently deep reactor may be used, wherein the hydrostatic pressure due to the depth of the molten liquid within the reactor acts to suppress the aluminum vapor producing reactions. The present inventors have further recognized that such a deep reactor facilitates the use of a plurality of horizontally and/or vertically spaced heating electrodes, thereby enabling different heating rates within the reactor.

[0007] In one aspect of the present invention, a system for carbothermically producing aluminum is provided, the system comprising a reactor having a bottom, sidewalls and a top defining a reaction chamber, a plurality of heating electrodes extending from the sidewalls, and a vapor recovery unit fluidly interconnected to the reaction chamber. The sidewalls of the reactor are of a sufficient height such that when the reactor is operated and contains molten liquid, the total pressure within the reaction chamber, as measured proximal the bottom of the reactor, is at least about 1.50 atm. Thus, the hydrostatic pressure due to the molten liquid should be at least 0.5 atm, such as at least about 0.75 atm or even at least about 1 atm, thereby producing total pressures within the reaction chamber of at least 1.5 atm, such at least about 1.75 atm, or even at least about 2 atm, as measured proximal the bottom of the reactor. These increased pressures suppress aluminum vapor forming reactions, thereby increasing aluminum production efficiency.

[0008] As noted, the deep reactor facilitates the use of a plurality of horizontally and/or vertically spaced heating electrodes. In one aspect of the present invention, the reactor comprises a plurality of horizontally and vertically spaced heating electrodes. A first set of heating electrodes may be disposed above a second set of electrodes, wherein the first set of electrodes may be operable to heat the molten liquid at a first heating rate and the second set of electrodes may be operable to heat the molten liquid at a second heating rate. For example, the first set of electrodes may be operable to heat the molten liquid to temperatures that facilitate the reduction of aluminum oxide to aluminum carbide material (e.g., temperatures of from about 1900°C to temperature of about 2000°C C.); and the second set of electrode may be operable to heat the molten liquid to temperatures that facilitate the reduction of aluminum carbide to aluminum metal (e.g., temperatures of at least about 2600°C C.). In combination with the deep reactor, restricted aluminum vapor production may be witnessed with the deeply submerged heating electrodes.

[0009] The inventive carbothermic aluminum production system may also include other components. For example, a submerged or submersible feed tube may be utilized to supply feed materials proximal the bottom of the reactor. Supplying feed materials proximal the bottom of the reactor further restricts the aluminum vapor forming reactions as such feed materials will be subjected to increased hydrostatic pressure upon entry to the reactor. In one approach, the feed tube may be substantially horizontally oriented and fixedly interconnected to a sidewall of the reactor for feeding of the feed materials. In this regard, the feed tube may extend through a sidewall of the reactor via a port located proximal the bottom of the reactor. In another approach, the feed tube may be substantially vertically oriented and adapted for vertical movement. In this approach, the feed tube may extend through the top of the reactor and toward a bottom portion of the reactor.

[0010] The inventive carbothermic aluminum production system may include a deep baffle system to facilitate suppressed vapor reactions. For example, the reactor may be a multi-zone reactor, such as described in U.S. Pat. No. 6,440,193 to Johansen et al. and U.S. Pat. No. 6,805,723 to Aune et al., each of which is incorporated herein by reference in its entirety. In this regard, a baffle separating one zone from another may extend from the top of the reactor toward the bottom of the reactor, terminating at a distal and proximal the bottom of the reactor. Thus, molten liquids flowing between two zones are forced proximal the bottom of the reactor, thereby suppressing aluminum vapor producing reactions via the increased pressure. A plurality of the above-noted heating electrodes may be located proximal the distal end of the baffle such that molten liquid flowing from one zone to another may be immediately subjected to a different heating rate to facilitate the production of aluminum in the increased pressure regions of the reactor.
As may be appreciated, various ones of the above-noted aspects, approaches and/or embodiments may be combined to yield various inventive carbothermic production systems and methods. These and other aspects, advantages, and novel features of the invention are set forth in part in the description that follows and will become apparent to those skilled in the art upon examination of the following description and figures, or may be learned by practicing the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1a is a schematic side-view of one embodiment of a carbothermic aluminum production system of the present invention.

FIG. 1b is a schematic side-view of the carbothermic production system of FIG. 1a with the vertical aluminum electrodes in a retracted position.

FIG. 2 is a schematic side-view of one embodiment of a carbothermic aluminum production system of the present invention.

FIG. 3 is a graph illustrating the effect of increased hydrostatic pressure relative to aluminum production.

FIG. 4 is a graph illustrating the effect of increased hydrostatic pressure relative to gas component concentration.

DETAILED DESCRIPTION OF THE INVENTION

Reference will now be made in detail to the accompanying drawings, which at least assist in illustrating various pertinent embodiments of the present invention.

FIG. 1 illustrates one carbothermic production system useful in accordance with the present invention. The system 1 includes a batch reactor 10 having a bottom 12, a top 14 and sidewalls 16 defining a reaction chamber 18. The system 1 includes a plurality of substantially horizontal heating electrodes 20 and may include one or more substantially vertical heating electrodes 22. The system also includes a vapor recovery unit 30 for recovering off-gases produced in the reactor 10. The system 1 may include feed tube 40.

The depth of the reactor 10 is such that when the reactor 10 is operated, the molten liquid (e.g., molten Al₂O₃, Al₂C₃ and mixtures thereof) creates a significant hydrostatic pressure within the reactor as measured proximal the bottom 12 of the reactor. More particularly, the reactor 10 has a depth D such that when the reactor 10 contains molten liquid 60, the hydrostatic pressure created by the molten liquid is at least about 0.5 atm, such as at least about 0.75 atm, or even at least about 1.0 atm. Thus, the total pressure at the bottom of the reactor (hydrostatic pressure+atmospheric pressure) is at least about 1.5 atm, such as at least about 1.75 atm, or even at least about 2 atm. These increased pressures decrease the thermodynamic favorability of the vapor forming reactions, thereby restricting vapor formation with increased aluminum production.

The depth D of the reactor 10 is dependent on the approximate density of the molten liquid within the reactor. For example, the slag produced during carbothermic aluminum production processes generally has a density of about 3.5 g/cm³. To achieve a hydrostatic pressure of about 0.5 atm proximal the bottom 12 of the reactor 10 due to the slag, the reactor 10 should have a depth of at least about 1.48 meters. To achieve a hydrostatic pressure of about 0.75 atm proximal the bottom 12 of the reactor 10 due to the slag, the reactor 10 should have a depth of at least about 2.2 meters. To achieve a hydrostatic pressure of about 1 atm proximal the bottom 12 of the reactor 10 due to the slag, the reactor 10 should have a depth of at least about 2.95 meters.

Despite the foregoing, some aluminum-containing gases will be produced during the carbothermic production operations. In this regard, the system 1 generally includes a vapor recovery unit 30 to recover aluminum in the gases. However, with the present invention, the load on the vapor recovery unit 30 may be substantially decreased, thereby increasing the efficiency of carbothermic aluminum production processes. Suitable vapor recovery units are described in, for instance, U.S. Pat. No. 6,530,970 to Lindstad, which is incorporated herein by reference in its entirety.

As noted, the reactor 10 includes a plurality of substantially horizontal heating electrodes 20. The electrodes 20 may be spaced from one another in a vertical and/or a horizontal direction. The heating electrodes 20 may be consumable graphite electrodes or non-consumable inert electrodes. Each of the electrodes 20 is individually supplied with electric current. By using a plurality of electrodes 20 in the sidewall 16 of the reactor 10 and a variable current supply for each of the electrodes 20, a selective temperature profile may be achieved within the reaction chamber. Due to the depth of the reactor 10, the heating electrodes 20 may be spaced at various depths within the reactor 10 to facilitate selective heating of molten liquid. For example, a first set of heating electrodes 20a may be disposed above a second set of electrodes 20b, wherein the first set of electrodes 20a may be operable to heat the molten liquid at a first heating rate and the second set of electrodes may be operable to heat the molten liquid at a second heating rate. The first set of electrodes 20a may be operable to heat the molten liquid to temperatures that facilitate the reduction of aluminum oxide to aluminum carbide (e.g., temperatures of from about 1900° C. to temperature of about 2000° C.), and the second set of electrodes 20b may be operable to heat the molten liquid to temperatures that facilitate the reduction of aluminum carbide to aluminum metal (e.g., temperatures of at least about 2060° C.). Thus, a selective temperature gradient may be achieved within the reactor and molten liquid, thereby increasing the efficiency of carbothermic processes. Moreover, higher heating rates may be achieved proximal the bottom 12 of the reactor 10, where the hydrostatic pressure is high, thereby suppressing aluminum vapor producing reactions that may accompany the reduction of aluminum carbide to aluminum. Any number of heating electrodes 20 may be used to achieve the desired heating rates and heating gradients.

As noted, the system 1 may include a feed tube 40 for feeding supply materials to the reactor 10 (e.g., aluminum oxide and pet coke or aluminum carbide containing feed materials). The feed tube 40 may be fixedly positioned relative to a depth of the reactor, or the feed tube 40 may be selectively positionable to facilitate feeding of supply materials at a desired depth. In one approach, the feed tube 40 is positioned or positionable such that its outlet is proximal the bottom of the reactor to further restrict the aluminum vapor forming reactions as such feed materials will be subjected to increased pressure upon entry to the reactor 10. In the illustrated embodiment, the feed tube 40 is substantially vertically oriented and adapted for vertical movement. In this embodiment, the feed tube 40 extends through the top of the reactor 10 toward the bottom 12 of the reactor 10. A motor or other mechanical means may be utilized to raise or lower the feed tube as necessary to facilitate feeding of the feed material at a desired vertical location within the reaction chamber 18. It is preferred that the feed tube 40 forms a seal with the top 14 so that off-gases do not escape at the interface between the top 14 and the feed tube 40.
In another embodiment (not illustrated), the feed tube may be substantially horizontally oriented and fixedly interconnected to a sidewall of the reactor for feeding of the feed materials. In this embodiment, the sidewall may include a port for receiving the feed tube and the feed tube outlet may extend therefrom. The outlet of the feed tube may terminate proximal the bottom of the reactor. In this regard, the port within the sidewall should be located just above the bottom of the reactor.

The system 1 may include one or more vertically oriented electrodes 22 extending through the top 14 of the reactor 10. The electrodes 22 are generally used in addition to the plurality of horizontally disposed electrodes 20. The electrodes 22 may be consumable graphite electrodes or inert electrodes (e.g., see U.S. Pat. No. 6,818,106, which is incorporated herein by reference in its entirety). During certain phases of operation of the reactor 10, the electrodes 22 pass through the bath and are submerged in the molten liquid to supply energy by resistance heating. For instance, in FIG. 1a, the electrodes 22 are submerged in the reactor to assist in the heating the molten liquid 60 to reduce alumina to aluminum carbide. In FIG. 1b, the electrodes 22 have been removed during the aluminum metal production step.

In this step, the horizontal electrodes 20 are generally used to heat the molten liquid 60 to produce an aluminum-containing liquid 62. This process is described in more detail in, for instance, U.S. Patent Application Publication No. 2006/0042413, which is incorporated herein by reference in its entirety.

The reactor 10 may include a top 14. The top 14 is generally utilized to cover the reactor so as to restrict heat and vapor loss. As may be appreciated, the use of the top 14 will also facilitate a slightly increased vapor pressures within the reactor 10, thereby assisting in suppressing the aluminum vapor producing reactions.

Another carbothermic production system useful in accordance with the present invention is illustrated in FIG. 2. The system 100 includes many of the components of the above system 1 of FIG. 1, but uses a different reactor 110. The reactor 110 has a top 114, bottom 112, and sidewalls 116, and further includes a baffle 117 that separates the reactor 110 into first and second zones 113, 115. In this regard, the reactor 110 may be operated as a continuous flow reactor, such as described in U.S. Pat. No. 6,440,193 to Johansen et al., which is incorporated herein by reference in its entirety.

Similar to the batch reactor 10 of FIG. 1, the continuous flow reactor 110 has a depth D that facilitates the creation of a hydrostatic pressure of at least about 0.5 atm as measured proximal the bottom 112 of the reactor 110. The system 100 may also include the plurality of horizontally disposed electrodes 22, which, again, may be horizontally and/or vertically spaced from one another. The baffle 117 separates the first zone 113 from the second zone 115 and defines a passageway P for passage of the molten liquid between the two zones.

The baffle 117 may be fixedly positioned relative to the reactor, or the baffle 117 may be moveable so as to facilitate selection of a suitable passageway P height. The baffle 117 may be positioned such that its terminal end T terminates proximal the bottom 112 of the reactor 110. Thus, as molten liquid flows from the first zone 113 to the second zone 115, the molten liquid will be subjected to an increased pressure, thereby suppressing aluminum vapor forming reactions. One or more of the horizontally disposed electrodes 22 may be located proximal the second zone 115 side of the passageway P, thus facilitating the reduction of aluminum carbide to aluminum at increased pressures.

The system 100 may also include the feed tube 40, which may be fixedly positioned or positionable such that its outlet is proximal the bottom 112 of the reactor 110. The feed tube 40 may be utilized on either side of the baffle 117. For example, if the feed material comprises alumina and/or pet coke, the feed tube 40 is utilized in the first zone 113 of the reactor 110, as illustrated. If the feed material comprises aluminum carbide, such as aluminum carbide recovered from the vapor unit 30, the feed tube 40 would be utilized in the second zone 115 of the reactor 110 (not illustrated). As may be appreciated, the feed tube 40 and baffle 117 may be integrated as a unitary structure wherein a single device is utilized to serve as both a baffle and a feed tube.

**EXAMPLES**

A computerized program (FACTSAGE, Thermo-Crct, Montreal, Canada and GTT-Technologies, Herzogenrath, Germany) was utilized to simulate the effect of increased pressures in the operation of the carbothermic reactor. The results from the simulation are given below in Table 1. FIG. 3 graphically illustrates the effect of increased pressure relative to aluminum production. FIG. 4 graphically illustrates the effect of increased pressure relating to gas phase composition. More aluminum metal and CO gas are produced and less aluminum vapor species, Al(v) and Al2O (s), are produced. The program validated that increased pressures within the reactor would result in increased aluminum metal production with decreased aluminum vapor production.

<table>
<thead>
<tr>
<th>Pressure (atm)</th>
<th>Temperature (K)</th>
<th>Gas CO (%)</th>
<th>Gas Al2O (%)</th>
<th>Alloy Al (%)</th>
<th>Slug (mol)</th>
<th>Carbide (mol)</th>
<th>Graphite (mol)</th>
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<tbody>
<tr>
<td>1</td>
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<td>78.7</td>
<td>14.5</td>
<td>6.8</td>
<td>0.34</td>
<td>0.14</td>
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<tr>
<td>2</td>
<td>2400</td>
<td>2.78</td>
<td>82.6</td>
<td>12.5</td>
<td>4.8</td>
<td>0.016</td>
<td>0.19</td>
</tr>
<tr>
<td>1</td>
<td>2450</td>
<td>3.46</td>
<td>76.5</td>
<td>13.2</td>
<td>10.2</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1.5</td>
<td>2450</td>
<td>3.29</td>
<td>80.4</td>
<td>10.8</td>
<td>8.8</td>
<td>1.2</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>2450</td>
<td>3.21</td>
<td>82.4</td>
<td>11</td>
<td>6.6</td>
<td>1.31</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>2500</td>
<td>3.63</td>
<td>74</td>
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<td>8.7</td>
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</tr>
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<td>9.6</td>
<td>8.6</td>
<td>1.34</td>
<td>0</td>
</tr>
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</table>
While various embodiments of the present invention have been described in detail, it is apparent that modifications and adaptations of those embodiments will occur to those skilled in the art. However, it is to be expressly understood that such modifications and adaptations are within the spirit and scope of the present invention.

What is claimed is:

1. A carbothermic aluminum production system comprising:
   a reactor comprising sidewalls, a top and a bottom defining a reaction chamber;
   a vapor recovery unit fluidly interconnectable to said reaction chamber; and
   a plurality of horizontally disposed electrodes interconnected to said sidewalls of said reactor,
   wherein said sidewalls of said reactor extend from said bottom of said reactor to a height such that when said reactor contains molten liquid the hydrostatic pressure of said molten liquid is at least about 0.5 atm as measured proximal said bottom of said reactor.

2. The carbothermic aluminum production system of claim 1, wherein said plurality of horizontally disposed electrodes includes a first set of electrodes and a second set of electrodes, said first said set of electrodes being disposed above said second set of electrodes.

3. The carbothermic aluminum production system of claim 2, wherein said first set of electrodes are operable to heat aluminum slag to a first temperature and wherein said second set of electrodes are operable to heat aluminum slag to a second temperature.

4. The carbothermic aluminum production system of claim 1, further comprising:
   a feed tube having a distal end terminating proximal said bottom of said reactor.

5. The carbothermic aluminum production system of claim 4, wherein said feed tube extends through said top of said reactor toward said bottom of said reactor.

6. The carbothermic aluminum production system of claim 4, wherein said feed tube extends through said sidewall of said reactor toward said bottom of said reactor.

7. The carbothermic aluminum production system of claim 6, wherein said feed tube extends through said sidewall via a port located proximal said bottom of said reactor.

8. The carbothermic aluminum production system of claim 1, wherein said sidewalls of said reactor extend from said bottom of said reactor to a height such that when said reactor contains molten liquid the hydrostatic pressure of said molten liquid is at least about 0.75 atm as measured proximal said bottom of said reactor.

9. The carbothermic aluminum production system of claim 1, wherein said sidewalls of said reactor extend from said bottom of said reactor to a height such that when said reactor contains molten liquid the hydrostatic pressure of said molten liquid is at least about 1.0 atm as measured proximal said bottom of said reactor.

10. The carbothermic aluminum production system of claim 1, wherein said reactor further comprises:
    a baffle extending from said top of said reactor toward said bottom of said reactor, thereby separating said reactor into a first zone and second zone, said baffle having a terminal end that terminates proximal said bottom of said reactor, thereby defining a passageway between said first zone and said second zone.

11. The carbothermic aluminum production system of claim 1, wherein the length of said baffle is such that when said reactor contains molten liquid, the hydrostatic pressure of said molten liquid proximal said passageway is at least about 0.5 atm as measured proximal said passageway.

12. The carbothermic aluminum production system of claim 1, wherein the length of said baffle is such that when said reactor contains molten liquid, the hydrostatic pressure of said molten liquid proximal said passageway is at least about 0.75 atm as measured proximal said passageway.

13. The carbothermic aluminum production system of claim 1, wherein the length of said baffle is such that when said reactor contains molten liquid, the hydrostatic pressure of said molten liquid proximal said passageway is at least about 1.0 atm as measured proximal said passageway.

14. A method of carbothermically producing aluminum, the method comprising:
   heating an aluminum slag in a reactor having a plurality of electrodes, said aluminum slag having a hydrostatic pressure of at least about 0.50 atm as measured proximal a bottom portion of said reactor;
   recovering aluminum from said reactor;
   introducing at least one of aluminum carbide, aluminum oxide and a carbonaceous material into said reactor via a feed tube.

15. The method of claim 14, wherein said feed tube extends substantially vertically through said reactor and terminates proximal a bottom portion of said reactor.

16. The method of claim 14, wherein said feed tube enters said reactor through a sidewall proximal a bottom portion of said reactor.

17. The method of claim 16, wherein said feed tube is substantially horizontal.

18. The method of claim 14, wherein said plurality of electrodes are operable to heat said aluminum slag such that a selected temperature gradient is achieved in said aluminum slag.

19. The method of claim 14, wherein said reactor comprises a baffle separating said reactor into a first zone and a second zone, said baffle having a terminal end that terminates proximal said bottom portion of said reactor, thereby defining a passageway between said first zone and said second zone, the method further comprising:
   flowing said aluminum slag from said first zone to said second zone proximal said baffle, wherein the hydrostatic pressure proximal said passageway during said flowing step is at least about 0.5 atm.

20. The method of claim 19, wherein the hydrostatic pressure proximal said passageway during said flowing step is at least about 0.75 atm.

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