1-1-2013

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Recommended Citation
Huda, Nazmul; Rhamdhani, M Akbar; Brooks, G A.; Monaghan, B J.; and Prentice, L, "Aluminium production route through carbosulfidation of alumina utilising H2S" (2013). Faculty of Engineering and Information Sciences - Papers: Part A. 524.  
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Aluminium production route through carbosulfidation of alumina utilising H2S

Abstract
Indirect carbothermal reduction of alumina for the production of aluminum utilizes different reducing agents to convert alumina into intermediate aluminum compounds. In the present study, the carbosulfidation route for aluminum production utilizing H2S(g) as the reductant and sulfur source has been investigated, in particular the formation of AlS3 in the first step of the process. The results of the thermodynamic analysis predicted that conversion of Al2O3(S) to AlS3(I) significantly increases above 1400°C at 1 atmosphere pressure. Experimental investigations were carried out at temperatures of 1100 to 1500°C using dilute H2S(g) gas in argon. The reaction products were analyzed using scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS), X-ray diffraction (XRD), inductively-coupled plasma absorption emission spectroscopy (ICP-AES) and chemical filtration. The X-ray diffraction results confirmed the presence of AlS3(S). Percentage of conversion from Al2O3 to AlS3 was found to be over 80% at 1500°C.

Keywords
route, h2s, production, utilising, aluminium, alumina, carbosulfidation

Disciplines
Engineering | Science and Technology Studies

Publication Details

This conference paper is available at Research Online: https://ro.uow.edu.au/eispapers/524
Aluminium Production Route through Carbosulfidation of Alumina utilising H$_2$S

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Keywords: Aluminum, carbosulfidation, H$_2$S

Indirect carbothermal reduction of alumina for the production of aluminum utilizes different reducing agents to convert alumina into intermediate aluminum compounds. In the present study, the carbosulfidation route for aluminum production utilizing H$_2$S(g) as the reductant and sulfur source has been investigated, in particular the formation of Al$_2$S$_3$ in the first step of the process. The results of the thermodynamic analysis predicted that conversion of Al$_2$O$_3$(s) to Al$_2$S$_3$(l) significantly increases above 1400°C at 1 atmosphere pressure. Experimental investigations were carried out at temperatures of 1100 to 1500°C using dilute H$_2$S(g) gas in argon. The reaction products were analyzed using scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS), X-ray diffraction (XRD), inductively-coupled plasma absorption emission spectroscopy (ICP-AES) and chemical filtration. The X-ray diffraction results confirmed the presence of Al$_2$S$_3$(s). Percentage of conversion from Al$_2$O$_3$ to Al$_2$S$_3$ was found to be over 80% at 1500°C.

Equilibrium Calculations of Al$_2$O$_3$-C-H$_2$S Reaction Systems

The equilibrium calculations were carried out using FactSage 6.1 thermodynamic package. The equilibrium calculations for Al$_2$O$_3$-C-H$_2$S system were carried at temperatures 1000°C to 2000°C at different pressures. For all equilibrium calculations, 3 moles of C and 3 moles of H$_2$S were considered for 1 mole of Al$_2$O$_3$. Figure 1 shows equilibrium calculation of Al$_2$O$_3$+3C+3H$_2$S for temperature range of 1000 to 2000°C at 1 atm pressure. Figure 1(b) show that significant amounts of gases are produced with majority of H$_2$(g) and CO(g) at higher temperatures. Al$_2$S$_3$ is predicted to be the main intermediate aluminum compound when H$_2$S is reacted with Al$_2$O$_3$ and C at 1000 to 2000°C at 1 atmospheric pressure. Formation of Al$_2$S$_3$ is predicted to be very low at 1100 to 1300°C at 1 atm pressure (0.1012 mol Al$_2$S$_3$/ mol Al$_2$O$_3$) and predicted to increase with increasing temperature to 1800°C. Formation of CO is predicted to be lower at 1100°C (0.035 mol/mol Al$_2$O$_3$) and significantly increases with increasing temperature (2.6 mol/mol Al$_2$O$_3$ at 1800°C). Along with CO and other gases significant amount of H$_2$(g) gas is also predicted to form at 1100°C (1.37 mol/ mol Al$_2$O$_3$). This content of H$_2$(g) was predicted to increase to 2.62 mol/mol Al$_2$O$_3$ when temperature is at 1800°C.
Experimental results

Experimental investigation on carboxulfidation of Al₂O₃(s) by using C(s) and dilute H₂S(g) (5% H₂S – 95% Ar) at different temperatures (1100 to 1600 °C) and reaction duration were carried out using a horizontal tube resistance-furnace (Nabertherm RHTV 200-600). A schematic diagram of the experimental setup is shown in Figure 2.

Figure 3 shows the comparison of XRD pattern of the samples after experiments at 1400°C for three different times (3, 6 and 9 hours). Al₂O₃ and Al₂S₃ peaks are marked by “1” and “2”, respectively. As shown in Figure 3, significant aluminum sulfide (Al₂S₃) was detected after 6 and 9 hours of reaction. This is indicated by the higher and sharper Al₂S₃ peaks at 6 and 9 hours compared to those from at 3 hours. Al₂O₃ peaks are still present, indicated that some Al₂O₃ remains and unreacted in the samples. However, it can also be seen clearly that there is a gradual decrease of the intensity with increasing reaction time.
Figure 3: X-ray diffraction pattern of the samples after 3, 6, and 9 hours experiments at 1400 °C. (1 = corundum (Al₂O₃), 2 = aluminum sulfide (Al₂S₃) and 3 = Graphite (C))

The percentage of conversion from Al₂O₃ to Al₂S₃ was determined by chemical dissolution and filtration. As pure Al₂S₃ completely dissolves in hydrochloric acid (HCl), a portion of the experimental samples were dissolved in HCl (36% w/w aqueous solution) and the solution was then filtered out. The amount of mass that dissolves in HCl represents the formed Al₂S₃ while the residues are the unreacted Al₂O₃ and C. From the filtration results, the percent of conversion (\( \eta \)) of Al₂O₃ to Al₂S₃ was calculated using following equation:

\[
\eta = \frac{\text{amount of sample dissolved}}{\text{amount of initial Al}_2\text{O}_3} \times 100\%
\]

The details of calculated conversion from selected experiments are shown in Table I. The highest conversion was found for experiment at 1500°C and 9 hours duration. The conversion showed an increasing trend with respect to time and temperature.

Table I: The conversion of Al₂O₃ to Al₂S₃ from selected samples at 1400°C and 1500°C

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Duration (hours)</th>
<th>Weight of Sample (g)</th>
<th>% of Conversion (( \eta ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1400</td>
<td>6</td>
<td>0.2012</td>
<td>75.4</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>0.2051</td>
<td>77</td>
</tr>
<tr>
<td>1500</td>
<td>6</td>
<td>0.2186</td>
<td>78.9</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>0.2060</td>
<td>81.6</td>
</tr>
</tbody>
</table>

In summary, the results, from XRD, SEM, EDS, ICP and conversion calculation, indicate that it is possible to form high amount of Al₂S₃ from Al₂O₃ using C and H₂S gas in the range of conditions studied. The results also suggest that the conversion to Al₂S₃ increases with increasing temperature and duration of experiments.