Light Metals 2012 *Edited by: Carlos E. Suarez*
TMS (The Minerals, Metals & Materials Society), 2012

STUDIES ON METAL FLOW FROM KHONDALITE TO BAUXITE TO ALUMINA AND REJECTS FROM AN ALUMINA REFINERY, INDIA

B. K. Mohapatra[1], B.K. Mishra[1], C.R. Mishra[2]
[1]Institute of Minerals and Materials Technology (IMMT), Bhubaneswar-751013, Orissa, India
[2]Institute of Advanced Technology and Environmental Studies (IATES),
Plot No. 80A & 83A, Lewis Road, Bhubaneswar-751002, Orissa, India

Keywords: Khondalite, Bauxite, Alumina, Undigested Sand, Red Mud, Metal distribution

Abstract

Alumina is produced from khondalite hosted bauxite of Indian origin in the Alumina refinery employing the time tested Bayer's Process. In the process, about 40% of unwanted elements are rejected as undigested sand and red mud. During the whole cycle; major, minor, trace and RE elements in these litho-units get redistributed, either depleted or enriched. Khondalite, the source rock of bauxite, is rich in silica, moderate in alumina and iron with minor titanium. Bauxite becomes rich in alumina and iron with subordinate titanium and negligible silica. After alumina recovery from bauxite, most of the valuable metals including REE get accumulated in the refinery rejects. The studies, while establishing the extent of various metals dispersed in khondalite, bauxite, undigested sand, red mud and alumina also reveals the state of enrichment of valuable metals in undigested sand and suggests possible means to recover some of them.

Introduction

Alumina is produced from khondalite hosted bauxite of Indian origin in the Alumina refinery employing the time tested Bayer's Process. When 2.4 MT of bauxite per annum is processed in a refinery it produces approximately 0.8 MT of alumina. During the process of alumina production, about 60% of unwanted gangue is rejected as 'undigested sand' and 'Red Mud'. Out of total quantity of 70 Million tonne per year (Mtpy) of red mud in the world, India accounts for about 2Mtpy. This industrial waste material poses tremendous environmental and disposal problems. With the anticipated expansion of alumina Industry in the country, Indian alumina plants have to dispose off over 5Mt of this red mud every year.

Aluminium oxide, from which aluminium metal is extracted, constitutes only 38 to 60% of bauxite ore. The rest is made up of Fe_2O_3, SiO_2, TiO_2 and scores of other metallic oxides. After dissolution of alumina in caustic soda, these impurities remain in suspended form which is separated out after being washed and then pumped as slurry to the nearby pond. During Bayer's process, all alumina content from the bauxite is not recovered and appreciable volume goes into red mud along with other valuable metals. Though some studies have been undertaken to recover metal values from red mud, they are not considered economically viable. Moreover, availability of high-grade ores of hematite, ilmenite, rutile, bauxite, monazite, vanadiferrous iron ore etc. in India, restricts attempts to recover any metal value from the red mud. Nevertheless, some metal values may be recovered in view of their scarcity and significance.

This paper while enlisting the extent of various metal values dispersed in khondalite (host rock of bauxite), bauxite (the feed to the alumina refinery), undigested sand and red mud (the rejects) and aluminium oxide (the product) discusses the dispersion pattern of metal values in all these litho-units, enrichment of valuable metals in undigested sand and suggests possible means to recover some of them.

Materials and Methods

The host khondalite, the feed bauxite, the rejects and the product samples were collected from one of the typical refineries of India. The recovery of aluminium oxide from bauxite through "Bayer's process" involves different stages. The main stages of Bayer's process are digestion of crushed bauxite → de-sanding → de-silication → settling → filtration → washing → pumping to red mud pond. The red mud samples from different stages were also collected for the study. The five samples collected are from Digested Mud (DM), De-silicated Mud (DSM), Settler Mud (SM), Washer Mud (WM) and Mud to Pond (MTP). In order to characterize and establish the mineralogical variation from bulk feed to mud to pond sequentially, each sample was studied by means of XRD (Philips) and SEM (Jeol). To ascertain the metal concentration (major and minor) from host rock to feed bauxite and the product to rejects, the samples were analyzed by XRF (Philips), and ICP-MS techniques (trace and REE).

The trace and rare earth elements (REE) were analysed at NGRI, Hyderabad, India using Inductively Coupled Plasma-Mass Spectrometer (ICP-MS). The ICP-MS used was a plasma quad PQ-1 (Fisons Instruments, U.K) controlled by an IBM PC-XT microcomputer and associated software. The ion-detection system and the data acquisition system consists of a Channeltron Electro Multiplier (CEM) and a multi channel analiser (Tracor Northern). To analyse the trace and REE the standard acid dissolution procedure was adopted for sample preparation as prescribed by Balaram et al.[1]. The sample solutions were prepared by HF-HNO3 acid digestion with Indium (100mg/ml) as internal standard. To ensure precision of the data a set of international ore standards of different chemical compositions were analysed along with the sample

Results and Discussion

Mineral distribution in host rock, feed, rejects and products

Detailed mineralogy of bauxite from several parts of world has already been reported[2-4]. Khondalite, the host rock of bauxite, is a quartzo-feldspathic, garnetiferous (almandine) schist/gneissose rock with or without mica /sillimanite (**Table-1**).

Table 1: Mineralogical variation between host rock and feed bauxite (as brought out through XRD)

Broad Litho type	Nature of Sample	Major Phase	Minor Phase
Bauxite	Pink	Gibbsite	Kaolinite, Muscovite
	Yellow	Gibbsite, Goethite	Muscovite
	Brown	Gibbsite, Goethite	Hematite, Sillimanite
	Black	Hematite, Gibbsite	-
	Red	Goethite, Hematite, Gibbsite	-
	Feed bauxite From Alumina refinary	Gibbsite, Hematite Goethite	Kaolinite, Sillimanite, Zircon
Host Rocks	Lithomarge/ clay	Kaolinite, Gibbsite	Goethite, Illite
	Partly altered Khondalite	Gibbsite, Kaolinite, Illite	Goethite, Microcline
	Khondalite	Gibbsite, Sillimanite	Almandine, Kaolinite

Table 2A: Mineralogical variation between classified plants and sample from alumina refinery rejects (as brought out through XRD)

Size fraction, in micron	Major Phase	Minor Phase
-1000+500	Gibbsite, Hematite	Goethite
-500+300	Hematite, Goethite	Gibbsite, Muscovite
-300+100	Hematite	Rutile, Goethite
-100+45	Hematite, Goethite	Quartz
-45	Hematite, Goethite	Quartz, Gibbsite

Table 2B: Mineralogical variation between red mud samples collected at different stages from alumina refinery (as brought out through XRD)

Type of Red Mud	Major Phase	Minor Phase
DG Mud	Rutile, Goethite, Hematite	Gibbsite, Sodalite
DSL Mud	Rutile, Goethite, Hematite, Gibbsite	Bohemite, Sodalite
SM	Hematite, Goethite, Rutile	Sodalite, Gibbsite, Bohemite
WM	Hematite, Goethite, Rutile	Goethite, Gibbsite, Sodalite
MTP	Hematite, Goethite	Sodalite, Gibbsite, Rutile, Bohemite

DM – Digested Mud; DSM - Desilicated Mud; SM - Settler Mud; WM- Washer Mud; MTP- Mud to Pond.

Minor mineralogical variation between khondalite, partly altered khondalite and lithomarge is recorded (Table-1). Bauxite, the feed to the alumina refinery, may occur in varied colours such as: yellowish, pinkish, grayish, brownish and show vesicular to massive texture and appears mostly in non-pisolitic form. X-Ray diffraction analysis indicated the bauxite to be composed predominantly of gibbsite with minor hematite (**Table-1**). The additional minerals found through Infrared spectroscopy are allophane, boehmite with subordinate diaspore and goethite. Other minor minerals recognised under reflected light microscope and scanning electron microscopes are rutile, ilmenite and zircon along with rare sillimanite, mica and kaolinite booklets. The rejects are broadly of two types, namely undigested sand and red mud. Megascopically, the undigested sand looks black to brick red in colour.

In view of its appearance like sand sized grains, the sample is sometimes termed as 'plant sand'. X-ray diffraction analysis brought out the predominance of hematite, goethite and rutile in this sand. The presence of minor gibbsite was noticed in its coarser fraction (>500micron) (**Table-2A**) and quartz in finer fraction (<100 m). When this sample is subjected to physical beneficiation, tabling in particular, the concentrate is found to contain additional minerals like ilmenite and zircon. Red mud is brick red in color, very fine and soils the finger. The size analysis of red mud indicates that its average size is around 3 micron, the d_{50} passing size is around 8 micron and about 35% by weight of solids contain below 5-micron particles. However, under SEM isolated grains > 20μ (boehmite and rutile / ilmenite) are recorded. The chief constituents of red mud in order of abundance are hematite, gibbsite, goethite, sodalite, boehmite and rutile. The red mud at different stages (DM, DSM, SM, WM, and MTP) looks identical and has more or less similar mineralogical composition. However, a minor variation in their volume percentage at different stages is brought out by XRD (**Table-2B**). Gibbsite dominates in feed bauxite and mud to pond samples. Presence of ilmenite, zircon, sphene in some samples is recorded only under electron microscope.

The refinery product aluminium oxide looks perfectly white and contains only gibbsite.

Metal Distribution in feed, rejects and products

The geochemistry of weathering sequence in bauxite profile has been reported by some workers[5-9]. The major, trace and rare earth metal distribution in host khondalite, bauxite fed to alumina refinary, undigested sand, red mud and aluminium oxide (the product) are shown in tables (3to 6). After the alumina is recovered by Bayer's process in the refinery, all other metals originally present in the bauxite are partly disposed into undigested sand and finally into red mud.

Major metals

The host rock khondalite is enriched in two elements such as alumina (29.37%) and silica (54.89) with subordinate iron (3.46%). During the process of bauxitisation, the alumina content gradually increases from host rock to partly altered khondalite followed by lithomarge. Bauxite derived from host khondalite rocks contains two major metals, namely aluminium (av.: 44% Al_2O_3) and iron (av.: 20% Fe_2O_3). Because of high iron content it appears red to yellow coloured and termed as ferruginous type.

When the alumina content is high, the bauxite sometimes appears pink or buff in colour. In the plant sand, the first reject from the refinery is found to be very rich in iron (av.:70% Fe_2O_3) and titanium (av.: 14% TiO2) metals. Some amount of alumina (av.: 8% Al_2O_3) also get released in to plant sand. The bulk red mud contains mainly four metal oxides like ferric oxide, aluminium oxide, titanium dioxide and silicon oxide in varying proportions. High Fe content in the red mud (Fe_2O_3: 51 to 58%) is obvious, because after alumina is digested in to solution the leached residue gets enriched in iron (**Table-3**). The product alumina contains 99.99% of Al_2O_3 and trace of iron, silica and titanium (each 0.01%).

Table 3: Major metal variation in feed, rejects and product

Major Component, in wt%	Host rock	Feed Material	Reject materials		Product
	Khondalite	Bauxite	Plant Sand	Red Mud	Alumina Hydrate
Al_2O_3	29.37	44.23	8.35	15.16	99.99
Fe_2O_3	3.46	20.16	70.89	53.98	0.01
SiO_2	54.89	2.66	1.52	12.13	0.01
TiO_2	1.15	1.99	14.23	3.88	0.01

Minor metals

The minor metals in bauxite of some significance include titanium (TiO_2: 2.2 to 3.00%), phosphorus (P_2O_5: 0.10 to 0.19%), manganese (MnO_2: 0.12 to 0.18) and sodium (Na_2O: 0.15 to 0.37). Potassium, calcium and magnesium are of rare occurrence (max. up to 0.1%). During treatment of bauxite all these elements go into red mud. Minor amount of calcium, phosphorus and manganese is recorded in red mud samples. Although calcium and phosphorus contents are negligible in first three stages of disposal [DM, DSM & SM], the value increases at washer mud (CaO: 1.56%; P_2O_5: 0.32%) and mud to pond (CaO: 2.3%; P_2O_5: 0.4%) sample (**Table- 4A & B**).

Table 4A: Major metal variation (in wt %) in red mud generated at different stages

Code	Al_2O_3	Fe_2O_3	SiO_2	TiO_2
Bulk	15.20	52.70	12.10	4.40
DM	14.80	51.10	12.71	5.20
DSM	15.90	57.65	14.50	4.10
SM	15.83	56.95	14.30	3.60
WM	14.53	51.80	10.70	3.20
MTP	14.73	52.39	8.44	3.30

Table 4B: Minor metal variation (in wt %) in red mud generated at different stages

Code	CaO	V_2O_5	P_2O_5	MnO
Bulk	0.94	0.16	0.23	0.15
DM	0.80	0.21	0.17	0.17
DSM	0.03	0.17	0.13	0.15
SM	0.03	0.15	0.13	0.14
WM	1.56	0.14	0.32	0.18
MTP	2.30	0.16	0.40	0.13

DM – Digested Mud; DSM - Desilicated Mud; SM - Settler Mud;WM- Washer Mud; MTP- Mud to Pond.

Higher calcium is due to addition of lime in the settler as flocculants and the minor amount of phosphorus (P_2O_5, 0.13 to 0.40%) may be from undigested sphene. Manganese content remains more or less similar (MnO: av. 0.1%) in the entire set of samples.

Trace metals

Distribution of trace elements in Indian bauxite is recorded by Banerjee[10]. In the present report the trace metal distribution pattern in the feed and rejects shows following interesting results:

i. The values of Sc, Ni, Sr and Cr increase sequentially from bauxite to plant sand and then to red mud (**Table 5**).
ii. Some metal values like Ga, Cs and Rb deplete from bauxite to plant sand and then increases in red mud. Appreciable Ga (35 ppm) goes with aluminium hydrate.
iii. Concentration of many metals like Co, Y, Hf, Ta, Cu, Zr, V, Zn, Nb, Ba, Pb, Th, and U show a rise from bauxite to plant sand and then fall in red mud.
iv. Significant rise in Zr, (2074 ppm), Cr (914 ppm), Ba (765 ppm) Ga (137 ppm) and Hf (84 ppm) values in finer fraction (-100μ) of plant sand is recorded. Higher Zr value is supported by concentration of zircon grains in this fraction.
v. The trace metals distributed in red mud samples at different disposal points (**Table 6**) also show some variation. While a few elements like Sc, V and Cr show increasing trend from DGM to MTP, many other elements (Ni, Y, Zr, Nb, Cs, Ba, Ta, Pb, Th and U) show a gradual depletion. However, elements like Cu, Zn, Co, Ga, Rb, Sr, Bi and Hf do not show any trend.

Table 5: Trace element variation in feed, rejects and products

Element, in ppm	Host Khondalite	Feed bauxite	Plant Sand	Red Mud	Alumina Hydrate
Sc	76.94	25.29	57.19	57.59	0.25
Co	13.55	16.18	98.98	24.21	0.01
Ni	68.73	30.39	46.75	53.01	1.65
Ga	64.42	79.15	74.69	91.31	35.26
Rb	2.63	14.33	0.68	5.75	0.07
Sr	12.48	20.45	36.53	43.57	3.87
Y	14.29	8.2	15.58	13.29	0.03
Cs	0.07	0.41	0.05	0.44	0.01
Hf	2.54	8.74	29.83	25.16	0.02
Ta	2.48	2.8	13.6	3.61	0.34
V	400.63	292.75	738.25	517.34	2.16
Cr	265.84	318.44	706.49	739.1	5.14
Cu	116.88	42.72	226.17	105.52	2.06
Zn	100.87	66.96	328.09	101.57	4.15
Zr	61.01	170.54	749.76	279.28	1.04
Nb	38.55	41.7	261.75	54.8	0.35
Ba	174.18	233.94	350.82	287.25	4.01
Pb	40.69	43.83	161.93	71.77	0
Th	41.89	117.63	295.52	175.8	0.08
U	2.80	1.86	6.31	2.82	0.1

ND: Not Detected

Table 6: Trace element variation in red mud generated at different stages

Elements in ppm	DM	DSM	SM	WM	MTP
Sc	48.58	50.42	52.08	66.93	69.93
V	411.61	423.12	425.04	580.67	746.25
Cr	643.33	654.50	759.76	791.55	846.35
Co	33.33	23.95	22.87	19.79	21.14
Ni	60.61	54.84	53.72	49.99	45.89
Cu	90.63	71.31	76.99	184.82	103.87
Zn	113.48	85.19	88.79	133.79	86.58
Ga	99.52	86.71	85.25	91.88	93.18
Rb	5.57	6.08	5.93	5.26	5.90
Sr	49.98	42..28	37.47	40.47	47.67
Y	16.71	14.28	13.66	11.86	9.95
Zr	380.58	286.37	250.88	244.48	234.10
Nb	79.74	58.47	50.93	44.82	40.05
Cs	0.62	0.57	0.46	0.30	0.23
Ba	440.39	389.84	339.78	175.52	90.75
Hf	15.94	82.59	9.83	9.16	8.29
Ta	5.31	4.14	3.39	2.68	2.55
Pb	95.46	83.56	74.29	57.30	48.22
Bi	103.98	109.66	95.64	72.91	38.05
Th	242.37	216.43	184.65	143.65	90.89
U	3.42	3.38	3.21	2.54	1.56

Table 7: Rare earth element variation in feed, rejects and products

Element, in ppm	Host Khondalite	Feed bauxite	Plant Sand	Red Mud	Alumina Hydrate
LREE					
La	73.71	64.81	215.72	112.46	0
Ce	176.52	97.43	498.33	190.82	0.27
Pr	18.76	10.14	53.95	18.01	0.01
Nd	62.17	32.49	184.99	48.06	0.06
Sm	11.92	5.29	31	8.59	0.12
Eu	1.26	1.54	1.66	2.35	0.01
HREE					
Gd	8.45	4.56	18.94	7.16	0.01
Tb	0.75	0.49	1.38	0.73	0
Dy	3.78	2.5	5.82	3.96	0.01
Ho	0.40	0.3	0.65	0.52	0
Er	0.87	0.8	1.53	1.25	0
Tm	0.22	0.12	0.17	0.19	0.01
Yb	1.79	0.93	1.71	1.76	0
Lu	0.21	0.13	0.29	0.23	0

Rare earth metals

Synthesis on the rare earth metal dispersion in the feed and rejects shows following results:

i. Most of the light rare earth and heavy rare earth metal values decrease from bauxite to plant sand and then increase in red mud (**Table 7**). However, Eu, Tm and Yb values gradually fall from feed to plant sand to red mud.

ii. In the finer fraction of undigested sand a rise in cerium value (498 ppm in bulk increases to 638 ppm in <45 μ) is observed. Similarly, most of the HRE element content (expecting Gd and Tb) show enhancement in finer fraction than that of the bulk sample.

iii. The light rare earth and heavy rare earth metals in red mud (excepting Tm) show gradual fall in their values from DM to MTP stage (**Table 8**).

Table 8: Rare earth element variation in red mud generated at different stages

Element, in ppm	DM	DSM	SM	WM	MTP
LREE					
La	168.94	149.93	135.52	66.87	41.05
Ce	252.35	238.86	210.54	157.39	94.96
Pr	27.29	24.29	20.77	10.54	7.15
Nd	73.68	63.27	54.10	30.61	18.64
Sm	13.03	11.54	9.03	6.01	3.35
Eu	3.66	3.20	2.81	1.25	0.82
HREE					
Gd	10.26	9.17	8.46	4.45	3.47
Tb	1.08	0.94	0.78	0.53	0.33
Dy	5.85	4.67	4.23	3.06	2.00
Ho	0.75	0.63	0.50	0.43	0.28
Er	1.65	1.46	1.23	1.12	0.81
Tm	0.29	0.25	0.22	0.17	0.12
Yb	2.26	2.05	1.88	1.60	0.99
Lu	0.30	0.27	0.24	0.24	0.12

DM – Digested Mud; DSM - Desilicated Mud; SM - Settler Mud; WM- Washer Mud; MTP- Mud to Pond.

Recovery of valuable metals from Plant Sand

The authors are in agreement with Rao et al[11] that the pland sand contains lot of valuable metal. For recovery of valuable metals from the plant sand, different conventional physical beneficiation techniques may be employed[12]. The sample was subjected to Tabling and collected the zircon bearing Table Concentrate-I and Ilmenite bearing table concentrate-II and tailings in continuous operation on bench scale. The table concentrate-II was re-tabled to collect the zircon rich concentrate-IIa and ilmenite rich concentrate IIb. The zircon rich concentrates I and IIa were re-tabled and the resulting heavies were subjected to wet high intensity magnetic separation [WHIMS], from which the zircon concentrate was collected as non magnetic product [~ 60% $ZrSiO_2$] and the ilmenite as magnetic product. This magnetic product is mixed with the Ilmenite rich concentrate-IIb and subjected to cell flotation. The flotation concentrate was rich in ilmenite content (~ 40% TiO2).

The table concentrate shows appreciable iron (Fe_2O_3: 67%) and titanium (TiO_2: 26%) along with other metals like Zr, Hf, Nb, Th, Ta, U and Y. All LRE and HRE elements are enriched in table concentrate product. The magnetic product obtained from WHIMS is rich in iron (Fe_2O_3: 76%) with appreciable titanium (TiO_2: 9%) along with minor enhancement in V, Cr, Rb, Sr while the non-magnetic product was enriched in Zr, Hf, Sc, Zn, Y, and

U. All LRE and HRE elements are enriched in non-magnetic fractions.

The flotation concentrate shows highest titanium value while it's tailing gets enriched in iron (Fe_2O_3: 68%). Elements like Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Hf and U values get enriched in the concentrate while value of Sc, V and Cr increases in its tail product. All LRE and most of the HRE elements excepting Gd show higher concentration in flotation concentrate.

Conclusions

Formation of bauxite is due to residual concentration of certain metals like aluminium during chemical weathering of silicate minerals[13,14] from host khondalite. In this bauxitisation process, some metals get depleted and some others get enriched. Due to poor mobility[15], metals like titanium, zircon, hafnium, and niobium are retained in bauxite while a few others like Y, V etc. move with iron and get precipitated in Fe- rich bauxite and laterite.

During then processing of bauxite in Bayer's process, alumina is recovered in the form of sodium aluminate solution while all other minor, trace and rare earth metals get precipitated with iron and discharged in to red mud pond and available therein. Due to intimate association of these metals with iron in the red mud it is difficult to recover some of them through any physical beneficiation techniques. However, some undigested materials, rich in valuable metals are removed as plant sand in the first stage. Following appropriate physical beneficiation process such as tabling, wet high magnetic separation and flotation, the titanium-rich and zirconium-rich minerals, containing several valuable metals in adsorbed state, from plant sand can be recovered. Though trace metals like Ga, Ba, Zn, Sr, Cr, V, Cu etc. are present in the alumna, produced from khondalite hosted bauxite, its superior quality is evidenced from absence any major and minor metals therein.

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