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Possibilities of Exploitation of Bauxite Residue from Alumina Production

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1. Introduction

The world aluminium production was 31.9 million tons in 2005 (Hetherington et al., 2007) and the demand is still growing. Aluminium is obtained from aluminium ore called bauxite in compliance with the locality where it was found near Les Baux in southern France in 1821. Bauxite can be found on all the continents, except for Antarctica, while the most abundant deposits are situated in tropical and subtropical zones, which provide optimal climatic conditions to its formation (Bardossy, 1990).

From ecological point of view the most significant world problem in aluminium production is creation of considerable amount of insoluble bauxite residue (BR). It is waste that, due to way of its production, is determined as red mud (RM) or brown mud (BM). 1 t of produced aluminium gets 2 t of waste and it is estimated that nowadays, during the bauxite processing, 120 million t of BR is produced worldwide (Red Mud Project, 2011).

Even if there is all-out effort in the world to reuse the waste from the aluminium production (Paramguru, 2005; Snars & Gilkes 2009; Liu et al., 2011; Maddocks et al., 2004), its majority part is dumped and due to its dangerous properties (high pH value, strong alkalinity, increased content of radioactive substances etc.) it is a significant environmental burden now and it will be a load also in the future. All world aluminium producers try to solve above mentioned problem more or less successfully. In this chapter, we propose a brief overview of the most significant possibilities of waste mud exploitation together with assessment of possible influence on the environment based on ecotoxicity tests.

2. Waste from alumina production

Many aluminium works are not located near to bauxite deposits. Consequently, they have to import bauxite or they buy pure aluminium oxide and the waste dumps stay at the localities of deposit or factories producing Al_2O_3 . Chemical composition of bauxite differs significantly in dependence on the locality of its deposit. Aluminium, as its most important component, occurs in bauxite in the form of hydrated aluminium oxide, whose content varies, while only ores with Al_2O_3 content more than 65 % are economically interesting. In dependence on chemical composition, physical properties of bauxite also vary, e. g. colour

(from yellow-white to grey, from pink to dark-red or brown) or structure (earthy, clay or compact ore).

Bauxite refining for Al_2O_3 production can be realized via several procedures (alkaline, acid, thermic, or high-pressured). However, the most spread procedure is the one of Austrian chemist K. J. Bayer, first patented in 1888 (Sintering process) and second in 1892 (Bayer process). Comparison of both processes is shown in Figure 1. (Klauber et al., 2009). The Bayer process resides in bauxite leaching by NaOH at increased pressure and temperature, in clarification of dissolved sodium aluminate and consequently in precipitation and calcination (Hind et al., 1999). Aluminium oxide, produced by the Bayer process, is relatively clean. It contains just several hundredths of per cent of impurities (oxides of iron and silicon). Waste is called red mud due to its intensive red colour. It is a suspension of very fine solid particles (more than 90 vol. % is lesser than $75 \mu\text{m}$) and solid concentration $\sim 400\text{g.l}^{-1}$.

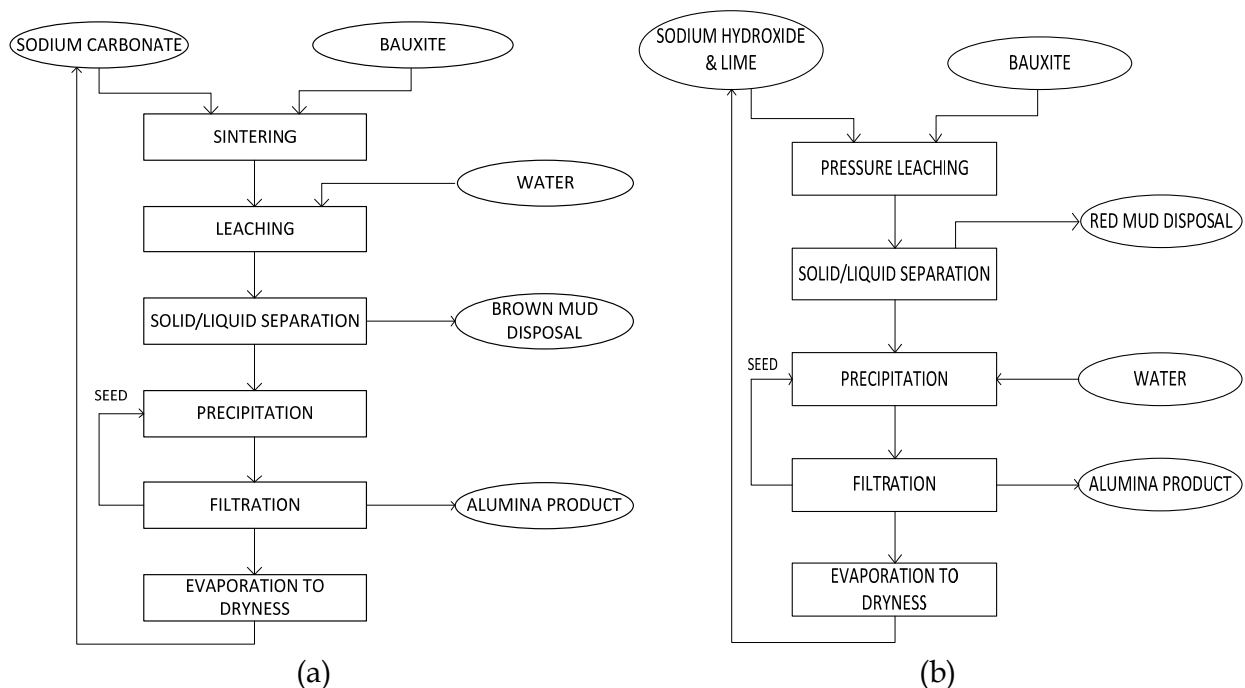


Fig. 1. Alumina production flowsheets of Sintering (a) and Bayer process (b) (Klauber et al., 2009).

Bauxite of higher silicon content is not suitable for the Bayer process, so in this case aluminium oxide has to be produced by so-called sintering process. For instance, in aluminium work in Žiar nad Hronom in Slovakia, 4.548 million t of Al_2O_3 were produced and 8.5 million t of waste mud emerged from 1957 to 1997¹ (Fig. 2.). Waste from the sintering process is called brown mud. In comparison with red mud it is more solid, because it contains higher portion of solid component, but it has lower content of Fe_2O_3 and Na_2O .

Similarly to the change of chemical and mineralogical composition of bauxite in dependence on its deposit, the composition of waste mud changes, too. Predominate component of waste mud are oxides of iron in the form of crystalline hematite (Fe_2O_3) that is the source of red colour of bauxite, or goethite $\text{FeO}(\text{OH})$. Aluminium oxide in the form of boehmite ($\gamma\text{-AlOOH}$) has its important place, and oxides of Ti, Na, Si, Ca, K and other metals,

¹In this year was alumina production terminated in Slovakia and ready alumina began to import.



Fig. 2. Bauxite residue (brown and red mud) dump in aluminium work in Slovakia before (left, 2006) and after (right, 2011) revitalization.

predominantly in the form of quartz, sodalite, gypsum, calcite, gibbsite, rutile, represent other minority components. In Tab. 1. chemical composition of waste mud from several world aluminium oxide producers is shown. All the data, except for Slovakia, is related to waste mud originated by the Bayer process (red mud).

Origin of Al ₂ O ₃ producent	Fe ₂ O ₃	Al ₂ O ₃	TiO ₂	Na ₂ O	SiO ₂	CaO	CO ₂	LoI*
France	19.4	27.3	1.9	5.2	10.2	8.6	-	16.5
Greece	41.9	14.8	5.8	10.0	10.0	-	-	-
Turkey	37.5	19.3	5.1	9.8	16.4	2,3	2.1	7,4
Guinea	56	21.1	2,3	0.4	3,7	0.2	0,8	15.7
Egypt	31	20	21	6	8	4	-	-
Russia	41,6	14.2	3,8	3.3	9,2	12.0	6,6	11.9
India	31.9	20.1	21.2	6.5	8.5	3.0	-	8.5
USA	41.6	16.4	5.5	6.8	1.3	6.7	-	-
Hungary	33-48	16-18	4-6	8-12	9-15	0.5-3.5	-	-
Brazil	43.7	16.8	4.0	9.8	-	-	-	-
Slovakia	45	15	6.0	7.5	13	2	-	6
Slovakia**	32	12.5	3.3	4.0	11.5	24	-	11
China	27.9	22.0	2.3	10.5	20.9	6.23	-	10.0

Note: *LoI - Lost of Incineration, **data related to brown mud

Table 1. Chemical composition of bauxite residue (BR) by origin of Al₂O₃ producer (in wt. %). Modified from article written by Kurdowski & Sorrentino (1997).

From ecological viewpoint it does not matter whether it is red or brown mud, because they both are dangerous waste and environmental load. The danger resides mostly in high alkali content, where pH values in fresh mud are higher than 13. There is a risk of percolating into groundwater, in case of rain storm there is a danger of dam crash (e. g. in Hungarian aluminium work Ajka in 2010), during the dry period of the year it is fly of dust particles and air pollution by aggressive aerosol and blocking large surface by dangerous waste.

Except for quoted predominant components, lesser amount of other metals, e. g. Na, K, Cr, V, Ni, Ba, Cu, Mn, Pb, Zn, Ga, Tl etc. can be found in waste mud.

Specific surface of waste mud reaches relatively wide values in the extent of 10 - 21 m².g⁻¹ and density of 2.51 - 2.7 g.cm⁻³. 1,4 % from original waste mud is dissolved in water, but 12,3 % is dissolved in hydrochloric acid (Kurdowski & Sorrentino, 1997).

Apart from above mentioned inorganic compounds in waste mud, the presence of many organic substances was described in literature (Hind et al., 1999) – such as polyhydroxyacids, alcohols and phenols, humic and fulvic acids, saccharides, sodium salts of acetic acid and oxalic acid etc. The presence of above mentioned organic substances is related to decay products of vegetation that grew up on the sludge beds and concerning enormous amount of red mud it is insignificant.

Concerning anticipated use of waste mud from Al₂O₃ production, especially as a part of construction materials, intensive research of its radioactivity was carried out, too. In several works (Hind et al., 1999; Somlai et al., 2008; Idaho State University, 2011) there was found out that total radioactivity of bauxite and of waste mud as by-product of bauxite processing is several fold higher than background. Therefore it is necessary to examine radioactivity level in products, in which waste mud from Al₂O₃ production was used as additive, while its part by weight would not exceed 15 %. In Tab. 2 there are average, minimal and maximal values of radioactivity for ²²⁶Ra, ²³²Th and ⁴⁰K of Hungarian authors (Somlai et al., 2008), who present that activity concentrations of original minerals can considerably change in the samples of bauxite and red mud during their chemical treatment, while no significant differences were marked in samples taken from the surface or from inside of the material.

Sample	Values	Activity concentration (Bq.kg ⁻¹)		
		²²⁶ Ra	²³² Th	⁴⁰ K
Bauxite	average	419	256	47
	min - max	132-791	118-472	10-82
Red mud	average	347	283	48
	min - max	225-568	219-392	4.9-101
Natural background (soil)*	average	48	45	420

Note: *data acquired from Idaho State University (2011)

Table 2. Radioactivity level of bauxite and red mud (Somlai et al., 2008).

As it follows from Tab. II in case of ²³²Th and ²²⁶Ra the measured values reach 5 - 7-fold of the background, while in case of ⁴⁰K they stay at lower value. Similar results were presented also by other authors undertaking radiological research (Akinci, A. & Artir, R., 2008). Waste mud depositing is connected with high fees for the depositing, because the deposit site must be constantly monitored for the ecological threat of possible groundwater contamination.

3. Exploitation possibilities

It is evident from the studied literature (Kurdowski & Sorrentino, 1997; Andrejcek & Soucy, 2004; Liu, Y. et al., 2011; Paramguru et al., 2005; Bhatnagar et al., 2011; Klauber et al., 2009) that exploitation possibilities of red mud for next processing are considerable, though just a few of

described techniques were put into practice, mainly because of economic reasons. These areas can be divided into several basic categories that include thermic, hydrometallurgical, mechanical, magnetic and chemical separation processes summarized in Tab. 3.

Areas of treatment	Processes	Product
metallurgical industry	drying, reduction	production of steel and other metals (Fe, Al, Ti, Ga, V etc.) melting agent
construction industry	drying, sintering, annealing	production of Al ₂ O ₃ and cements
	addition	production of concrete fillers
	drying, pressing, firing	bricks production
glass and ceramic industry	drying, granulation	aggregates of special concretes
	addition	glass manufacturing
chemical industry	drying, pelletisation, annealing	ceramic manufacturing, ceramic glazes
	chemical treatment	catalyst
agriculture	drying and chemical treatment	production of adsorbents, pigments, resin contents, filler for plastics
	addition to soil	improvement of soil physicochemical properties
water supply	neutralization, adsorption	remediation of soil, revegetation
	adsorption	removing of moisture and other undesirable components, treatment of liquid wastes
other	coagulation	coagulant
	drying and chemical treatment	filtration material
	adsorption	neutralization of acid mine drainage

Table 3. Overview of BR application areas from the alumina production.

3.1 Metallurgical industry

Beside material recovery of waste mud, the possibilities of sodium returning into the production process of aluminium oxide were studied on the base of above stated chemical and mineralogical composition (Lamerant, 2000). Processes for the recovery of other metals or of their oxides included in waste mud, e. g. Fe (Li, 2001; Piga et al., 1995; Qiu et al., 1996; Thakur & Sant, 1983), Al₂O₃ (Ercag & Apak, 1997), resp. TiO₂ (Agatzini-Leonardou et al., 2008), individually and in combination are equally important. Chinese authors describe nearly non-waste technology of red mud processing (Liu W. et al., 2009). They studied deep-drawing quality of iron by direct reduction in the sintering process and subsequent magnetic separation under different conditions (temperature and time of sintering, carbon -

red mud ratio and content of additives) and, in the next step, use of aluminosilicate residues in building industry for bricks production, where the problem of soluble sodium salts was solved by their calcification to insoluble aluminosilicates.

The possibility of use of minority components in waste mud still remains interesting. However, in spite of the existence of several patents the economics of the whole technology is decisive. The possibilities of recovery of rare-earth metals (Sc, U, Ga, Tl, V, Ce, Y etc.) were described (Smirnov & Molchanova, 1997; Ochsenkühn-Petropoulou et al., 2002; Gheorghita & Sirbu, 2009), which, in most cases, reside in cementation of waste mud, its extraction in acid medium and finally by the use of ion-exchange methods. Relatively high scandium content in bauxite residue processed by Aluminium of Greece allowed extraction of the Sc by nitric acid leaching followed by ion exchange separation (Ochsenkühn-Petropoulou et al., 1995; 1996). Obviously no impact was observed on the amount of residue to be stored by extraction of only minor component from bauxite residue.

Extraction of alumina and sodium oxide from red mud was investigated (Zhong et al., 2009) by a mild hydro-chemical process and the optimum conditions of Al_2O_3 extraction were verified by experiments as leaching in 45% NaOH solution with CaO-to-red mud mass ratio of 0.25 and liquid-to-solid ratio of 0.9, under 0.8 MPa at 200 °C for 3.5 h. Subsequent process of extracting Na_2O from the residue of Al_2O_3 extraction was carried out in 7% NaOH solution with liquid-to-solid ratio of 3.8 under 0.9 MPa at 170 °C for 2 h. Overall, 87.8% of Al_2O_3 and 96.4% of Na_2O were extracted from red mud. The final residues with less than 1% Na_2O could be utilized as feedstock in construction materials.

Of the all amount of bauxite residue applications, only 15 % relate to metallurgical industry (Klauber et al., 2009) and of this amount about 30 % is related to steel making and slag additives. Bauxite residue is used as a source of aluminium, silicon and calcium to modify the properties of the slag to improve separation, setting and other qualities.

3.2 Construction industry

The most widespread use of red or brown mud processing is in construction industry (production of building components – bricks, concrete blocks, ceramic materials etc., concrete aggregate, special cement etc.). Brown mud obtained by sintering calcination method contains suitable reactive components, e. g. $\beta\text{-}2 \text{CaO}\cdot\text{SiO}_2$, so it can be used (unlike red mud) for direct bricks production (Liu et al., 2009), where it is pressed into blocks and is calcined at high temperature, while compression strength of produced bricks reaches value of 1.9 MPa (Amritphale & Patel, 1987). In the second method red mud is mixed with binders and consequently it is hydraulically hardened or calcined at low temperature. Bricks from suitable mixture of clay, red mud and fly ash, calcined at 1000-1100 °C, reach high compression strength (40 to 70 MPa) (Dass & Malhotra, 1990) and they are suitable material for multi-storey buildings construction – due to their low moisture absorption, suitable density, exceptional fire resistance and characteristic colour and texture (Dimas et al., 2009).

Due to suitable Fe oxides-Al oxides ratio red mud is used in Portland cement production (Satish, 1997; Tsakiridis et al., 2004) or as additive in special cements production (Singh, et al., 1996). In both cases the addition of red mud into cement is limited especially due to strong alkalinity and it does not usually exceed 15 wt. %. According to the Japanese patent cement cinder was produced by mixing of calcium materials with red mud and

subsequently by compressing and calcining in tunnel furnace at the temperature of 1300 - 1450 °C (Ogura, 1978). Natural red or red-brown colour of waste mud imitates the colour of brick and it is used for creation of interesting architectonic effects, for colouring concrete compounds, while colour adjustment of red mud can be done by pH adjusting, mixing with other oxides or by mild calcining (Satish, 1997). Replacing one-third of the content of silicate fractions in concrete compounds by red mud results in the fact that hardened concrete gains greater compression strength than concrete obtained by the use of quartz sand (Buraev & Kushnir, 1986). Hardening of concrete compound by utilization of red mud in amounts of 1 to 15 % under higher pressures (Kohno et al., 1998) helps to improve some final product properties, e.g. compression strength, water resistance, fire resistance etc. Several possibilities of the use of such concrete compounds were described, e.g. a filling for construction materials, materials used in dump construction, production of concrete blocks filling the dam structures or filling of building constructions (Klein, 1998; Kane, 1979; Di San Filippo, 1980; Browner, 1995).

3.3 Glass and ceramic industry

Red mud belongs to the group of so-called pozzolanic materials, which, after mixing with lime in the presence of water, harden and they form stable and durable compounds. The additions of red mud into clay compounds were described in the production of ceramic glass (Wagh & Douse, 1991; Sglavo et al, 2000a; Yalcin & Sevinc, 2000; Singh et al., 1996; Pratt & Christoverson, 1982). The content of red mud did not affect the sample porosity, but more deflocculated system originated, in which critical moisture content was increased. By augmenting red mud content to 20 %, after calcination at the temperatures of 950 and 1050 °C, growth of density and flexural strength were noticed in final ceramic products. It was brought about by bigger amount of glass share at bigger red mud content, which moreover by its natural colour replaces pigments addition to obtaining red-brown tones (Sglavo et al, 2000b).

3.4 Chemical industry

In organic synthesis red mud can be used as hydrogenation catalyst after its activation e.g. in hydrogenation of naphthalene to tetraline (Pratt & Christoverson, 1981). After several modifications (e.g. sulphidization) it is used as catalyst in petrochemical industry (Eamsiri, 1992; Álvarez et al., 1999), in dehydrochlorination by tetrachlorethylene (Ordoñez, 2001), as catalytic converter (exhaust fumes), more specifically in methane catalytic incineration (Paredes, 2004), removing sulphur from waste gas (Khalafalla & Haas, 1972), selective catalytic reduction of nitrogen dioxide (Lamonier, 1995), removing hydrogenchloride, carbon monoxide and dioxins (Hosoda et al., 1995) and recently also in ammonia decomposition (Ng et al., 2007). Other possibilities of catalytic use of red mud are described in literary review (Sushil & Batra, 2008).

Considering the size of specific surface of waste mud, its use as cheap adsorptive agent gives us a wide scale of possibilities in different areas. When preparing adsorbents from waste mud, it is necessary to realize firstly neutralization, washing, drying or, if necessary, another activation. It is important to note that, when there is variety of waste mud composition in dependence on processing technology (content of metals, radioactive elements, as well as organic substances – see chapter 2), it is required to perform leaching

tests to manage the risk of possible contamination, e. g. in water treatment technology. Applications of red mud as adsorbent were presented (Wang et al., 2008) in adsorption of water solutions anions (PO_4^{3-} , F^- , NO_3^-), trace elements cations – metals and metalloids (Cu^{2+} , Pb^{2+} , Cd^{2+} , Zn^{2+} , Ni^{2+} , Cr^{6+} , As^{3+} and As^{5+}), pigments (rhodamine B, Congo red, methylene blue), organic substances (phenols, chlorinated phenols), but also in adsorption of toxic gases in emissions cleaning (H_2S , SO_2 , NO_x). Titanium extraction by diluted sulphuric acid under atmospheric conditions was carried out without any previous treating of red mud, while following interactions of leaching process were monitored: acid concentration, temperature and ratio of solid and liquid part on which efficiency of titanium separation depended (recovery/yield of 64,5 % was achieved) (Smirnov & Molchanova, 1997). By the adsorption on red mud, there were described: separation of sulphuric compounds (cyklohexantiol) from oil (Singh et al., 1993), separation of phosphoric compounds (Mohanty et al., 2004) or removing bacteria and virus (*Escherichia coli*, *Salmonella adelaide* and poliovirus-1) by waste water filtration through sand filter mixed with red mud (Ho et al., 1991).

To prepare gypsum waste mud is elutriated by sulphuric acid or by mixture of H_2SO_4 , HNO_3 , KNO_3 , soil microorganisms (*Deuteromyces*) and water in weight ratio of 10:3:14:72, with pH value < 1, still mixed for 100 - 200 hours. Created solid phase, after separation by filtration and drying, provides gypsum and potassium aluminium sulphate is obtained from filtrate after crystallisation. After crystallisation it is possible to thicken mother liquor by evaporation. Red pigment is gained from originated solid amorphous phase with majority share of hematite after drying, followed by calcination at the temperature of 600 °C and treating particles size by ultrafine milling under 5 µm (Kušnierová et al., 2001).

3.5 Agriculture and soil remediation

Extensive laboratory experiments were carried out in Australia (Summers et al., 2001). They dealt with red mud utilization as additive into soil to improve its properties during 10 years, when reduced loss of nutrients from soil into water was found out and agriculture production was increased (van Beers et al., 2009). Contaminated soil remediation by agents based on red mud was described. In the remediation there is adsorption of toxic substances and the structure and texture of soil improve. A series of applications was characterized in processing of acid mine drainage at sludge beds, which are old environmental loads and where application of alkaline red mud decreases soil acidity and, due to its excellent sorption properties, it binds a lot of toxic metals, e. g. Cd, As, Cu, Pb, Zn, Cr etc. (Gray et al., 2006; Bertocchi et al., 2006). Except for pH value increase, addition of red mud into soil shifts chemisorption of metal ions to Fe-oxide form. Consequently acidic extractability of these metals decreases and their fixation in red mud is ensured (Lombi et al., 2002). Synergistic effect of red mud additives to compost compounds, which provide for organic carbon increase in created soil, was studied, too (Brunori et al., 2005). In this soil, there was monitored sequential extractability of several fractions (poorly adsorbed, reducible, oxidizable and the rest) by using different extractants (CH_3COOH , $\text{NH}_2\text{OH}\cdot\text{HCl}$, $\text{H}_2\text{O}_2 + \text{CH}_3\text{COONH}_4$ and compound of HCl, HClO_4 and HF). After addition of red mud into soils contaminated by heavy metals, decreasing of metal mobility was confirmed (the most significant leachability reduction was in case of Mn, Zn and Ni). This makes reusing of red mud in big quantities very promising (Maddocks et al., 2004).

Absorption ability of red mud to remove pollutants has to be supported by activation of fresh mud from refineries. Different activation methods and their effect on physiochemical properties are summarized in review (Liu, Y. et al., 2011).

3.6 Other utilization

Bauxite waste mud can be used in mining industry in gold extraction by cyanide method (Browner, (1995), where it is a very efficient modifier of pH value that has to be kept high in the process, while gold, possibly adsorbed on red mud, together with gold in ore, is separated by gravitational sedimentation. Recently a new method of production of shielding material against X-ray radiation using red mud and barium compounds has been developed (Amritphale et al., (2007). In comparison with traditional lead protective screen, needed thickness to retain the same energy was significantly lower, while other mineralogical (X-ray diffraction) and mechanical properties (pressure force and impact force) of new shielding material were studied, too.

Effective passivation of steel surface by using of red mud as anticorrosive agent (Collazo et al., 2005) before subsequent modifications (grinding, dipping, degreasing, painting etc.) was described. It was found out that, after immersing steel into decanting suspension of red mud and continuously mixed during 24 hours, depassivation runs at lower value of Cl^-/OH^- ratio and at lower pH value than untreated samples. The results of X-ray diffraction analysis confirmed that red mud does not create continuous layer on passivized steel surfaces, but it creates discrete distribution of individual elements formed mainly by oxy-hydroxides of Fe and Al. The development of electrode potentials on steel surfaces using electrochemical impedance spectroscopy was studied. It was studied in dependence on bating length in red mud bath, where, in comparison with untreated samples, significant potential of current density was reached (Collazo et al., 2007).

Hamdy and Williams (2001) studied bacterial amelioration of bauxite waste mud. They describe isolation of 150 bacterial cultures (the most significant representatives were *Bacillus*, *Lactobacillus*, *Leuconostoc*, *Micrococcus*, *Staphylococcus*, *Pseudomonas*, *Flavobacterium* and *Enterobacter*). After addition of nutrients and possible chemical modification the microorganisms were able to grow until the concentration of 10^9 CFU.g^{-1} of waste mud, while organic acids produced by metabolism reduced pH value from 13 to 7. Aluminium recovery using *Penicillium simplicissimum* (Ward & Koch, 1996), calcium and iron using *Bacillus polymyxa* (Anand et al., 1996) were characterized by biological leaching. By bacterial metabolism, which runs by mechanism of oxidoreduction reactions using not only carbon but also sulphur and metals, it comes up to the metal nucleation at specific places of the cell surface of bacteria. Consequently, under appropriate conditions (especially pH control, aeration and nutrients addition), large amounts of bacterial clusters can catalyse secondary minerals formation (2005), which can be separated from original mixture.

4. Studies of environmental compatibility of waste mud

4.1 Leachability

The most important dangerous property of waste mud from Al_2O_3 production is high pH value because of the presence of large amount of NaOH in the leaching process that is used redundantly and it causes causticity or corrosivity of this waste. For this reason raw waste

mud is usually neutralized before its placement to sludge bed. Most often it is neutralized by sea water or by other technologies.

Toxic effect of dangerous waste depends on biological accessibility of toxic agents, which is related to solubility in water medium, i. e. leachability. Recently several research of leachability of waste mud from Al_2O_3 production has been performed. Using extraction test TCLP (Toxicity Characteristic Leaching Procedure Test) at pH value of 3,0 Indian authors (Singh & Singh, 2002) studied metals extraction from red mud (mainly hexavalent Cr and Fe), where very low leachability was found out. The possibilities of Cr^{6+} leachability decrease from red mud were studied because of red mud use as a component of building materials and its stabilization by Portland cement in ratio of 3:1 was proposed. Metals leachability from Bauxsol™ was studied, too. Bauxsol™ presents red mud neutralized by sea water and in practice it is used as adsorption material. Detected leachability under different conditions did not exceed limiting values for heavy metals content in waste water (McConchie et al., 1996).

Besides red mud, leachability of heavy metals from different mixtures with red mud was observed. Red mud is added into these mixtures especially as effective adsorbent, e. g. in different soils, composts or in building materials. Noteworthy Italian kinetic study concerns leachability of trace elements using sequential extraction with red mud added to contaminated mining soil, where leachability of some metals, present in large amounts, e. g. Mn, Zn a Ni (Brunori et al., 2005a), was significantly decreased. In another study of Italian authors (Brunori et al., 2005b) leachability of differently modified red mud (neutralization, washing) was observed according to extraction test by deionised water in ratio of L/S = 5 and eightfold repetition of extraction in given time periods – 2, 8, 24, 48, 72, 102, 168 and 384 hours. Values of red mud leachability without washing and after washing by water are in Table 4.

Parameters	Unit	Unwashed treated red mud	Washed treated red mud	Threshold value*
Arsenic	$\mu g.l^{-1}$	45 ± 4	24 ± 3	50
Beryllium	$\mu g.l^{-1}$	< 0.5	< 0.5	5
Cadmium	$\mu g.l^{-1}$	1.0 ± 0.1	0.6 ± 0.1	5
Cobalt	$\mu g.l^{-1}$	< 0.5	< 0.5	50
Chromium	$\mu g.l^{-1}$	16 ± 2	6 ± 1	50
Copper	$\mu g.l^{-1}$	51 ± 5	$2,7 \pm 0,5$	100
Nickel	$\mu g.l^{-1}$	11 ± 1	$1,5 \pm 0,3$	100
Lead	$\mu g.l^{-1}$	1.7 ± 0.2	5 ± 1	50
Vanadium	$\mu g.l^{-1}$	555 ± 50	485 ± 70	50
Zinc	$\mu g.l^{-1}$	< 50	< 50	3 000
Chloride	$mg.l^{-1}$	4880 ± 450	128 ± 25	200
Fluoride	$mg.l^{-1}$	32 ± 3	16 ± 3	1.5
Nitrates	$mg.l^{-1}$	2.4 ± 0.2	n.d.	50
Sulphates	$mg.l^{-1}$	1140 ± 100	530 ± 100	250
pH	range	6.4 – 10.5	8.2 – 10.2	5.5 – 12

*For the Italian leaching test

Table 4. Leachability of neutralised red mud without washing and after washing by deionised water (Brunori et al., 2005b).

Measured above the limit values of some anions were probably caused by processing of red mud by seawater. After washing of such processed mud by deionised water, significant decrease of anion content was reached, in case of chlorides there was 30-fold decreasing. Despite washing, content of sulphates and fluorides was still above limits. From monitored metals vanadium is important, because its concentration is 10-fold above the limit (according to Italian legislation twice above the limit). This high concentration is ascribed by authors to high mobility of vanadium in neutral and alkali environment and also to high content of vanadium in the red mud.

According to EPA report (U.S. EPA, 1990) about special waste from minerals treating for Al_2O_3 production, except for selenium and arsenic, all compounds monitored according to EPA rules were at least two orders below the limits. From 18 indicators monitored in elutriate from red mud, As and Se concentrations were three time above screening criteria, what means (in precondition of elutriate diluting by ground water) at ingestion a cancerogeny risk for people. Despite strict screening criteria of EPA for elutriate, the Report (U.S. EPA, 1990) ascertains that toxicity limits were not exceeded in all monitored cases.

As described by Friesl (Friesl et al., 2003), after red mud adding to sand soils, significant decrease of heavy metals leachability was observed, e.g. Cd by 70 %, Zn by 89 % and Ni by 74 %. Simultaneously authors monitored catching of these metals by plants, what lead to decrease of metal content by 38 - 87 %. However, if there is added red mud into the soil exceeded 5 wt.%, content of some toxic metals (As, Cr and V) is increased too, and changes in bioavailability of the metals are also discussed. For instance, after increasing of total Cu content after red mud adding, probably because of decrease of Cu bioavailability, decreasing of phytotoxicity occurred what is confirmed by higher production of corn biomass. There were also found out concentrations of other monitored metals: arsenic 5 mg.kg⁻¹, chrome 20 mg.kg⁻¹ and vanadium 5 mg.kg⁻¹. After leaching these amounts cause overcoming of limits and therefore adding of red mud into the soil to enhance of soil properties must be controlled (Friesl et al., 2004). Effect of pH was studied to soils enhanced by addition of red mud, lime and beringite and was confirmed leachability decreasing of Cu, Cd and Zn (Lombi et al., 2003). The leachability is low in wide range of soil pH despite reacidification of soil.

4.2 Toxicity tests and ecotoxicological biotests

Ecotoxicologic biotests and chemical analyses are the most important methods of ecotoxicological detection systems for elutriates from waste. These methods can directly assess detrimental effects on environment regardless off composition and character of tested waste. Significance of these methods consists in identification and assessment of risk mainly from the point of view of migration of contaminants from soil into ground water. The biotests are realized at producer-consumer-decomposer level and its combination can improve or degrade the result of extraction methods. Choosing of appropriate leachability method and detection system are the biggest sources of variability in assessment of leachates from waste. At classification of waste into the hazardousness categories, an economic factor plays important role. Therefore the biggest abuse of factors influencing results of tests at choosing of above mentioned systems from waste producers is assumed. For the kind of waste with extremely high pH, different methods of extraction had to be ordained. These methods were optimised according to the experiments.

Apart from leachability tests, important mainly in landfilling and assessment of environment contamination – contamination of ground water, surface water near sludge bed, ecotoxicity tests of sludge are also important. The tests enable to assess appropriate way of using in above mentioned spheres. Ecotoxicity tests of red mud have been described in literature only recently and just for small number of applications.

In the environmental compatibility study of red mud from Italian authors (Brunori et al., 2005b) three kinds of standard ecotoxicologic tests were realized. Red mud was neutralised by sea water and final pH of processed mud was about 7. Acute toxicity was tested by sea luminescent bacteria *Vibrio fischeri* with biotest Microtox™. Concentration from 0.1 to 2.0 % of red mud in sea water was measured. Obtained value of bioluminescence produced by the bacteria did not reach the detection limit of the method (20 %), what refers to negative environmental effect of the suspension. Similar result was measured by the test with *Dunaliella tertiolecta* according to American Society for Testing Materials (ASTM) methodology. No significant differences between blank and water extract of neutralised red mud at above mentioned concentrations were measured by ASTM test of embryonic toxicity in pluteal phase of sea gastropod *Sphaeroechinus granularis* after 72-hour incubation. Only in paper of Pagano (Pagano et al., 2002), toxicity of raw red mud on sea urchin embryos was detected because of high alkalinity (pH 12). In various samples from sludge bed of 4 European aluminium works (Turkey, France, Greece and Italy), larval retardation, malformation, development malfunctions and early embryonic mortality were studied. Significant sperm toxicity and influence to offspring quality was confirmed.

Acute toxic effect of adsorbent Bauxsol™, which is produced from red mud after neutralisation and processing with sea water in Australia (Corp. Virotec Global Solutions of Gold Coast, Queensland, Australia), was studied by earthworms (*Eisenia foetida*) (Maddocks et al., 2005). Tested Bauxsol™ contained more than 6,450 mg of adsorbed metals per 1 kg of adsorbent. The Bauxsol™ was mixed with dung of cows into various ratios. Final concentrations of the Bauxsol™ were 100 %, 90 %, 80 %, 60 %, 40 % and 20 %. Earthworms were exposed to the mixtures for 28 days. In all samples earthworms exhibited good mobility and no mortality. The highest bioaccumulation of metals in earthworms was found out at 20 and 40 % of Bauxsol™ in dung of cows. Despite it, calculated bioaccumulation factors (BAF) were lower than published threshold values of toxicity causing mortality or published values for middle polluted soils. By sequence extractive analysis of mixture 20 % Bauxsol™ in dung of cows it was found out, that more than 95 % of metals were bound in Fe/Mn oxide fraction and after exposing by earthworms, change in composition of metal fraction Cd/Cr and Fe/Mn happened. Mentioned results indicate that metals adsorbed in Bauxsol™ are not accessible for earthworms and extractive methods are usable for studying of toxicological characteristics.

Genotoxic properties of red mud have been studied by Orešcanin (Orešcanin et al., 2003). Tested samples exhibited no cytotoxic and mutagenic activity at two bacterial strains *Salmonella typhimurium* (TA98 a TA100), which were studied at presence and also at absence of metabolic activation. The authors used sequence extractive analysis of red mud. No toxic effect of new coagulant prepared from red mud was confirmed before starting commercial production of the coagulant. Reusing and regeneration of waste mud were described as very promising.

Relatively wide-ranging and long term ecotoxicity tests of red mud were realized also in France (Ribera & Saint-Denis, 2002) in connection with depositing of waste from two aluminium works near Marseilles on the sea bed of Lion bay. In decade 1997 – 2007 all ecotoxicity tests were negative - Microtox® by *Vibrio fischeri*, chronic toxicity tests by sea-urchins, gastropods and oysters, Ames test of genotoxicity and acute tests by European seabass *Dicentrarchus labrax* - 152 tests total from 19 sampling places, except for two contact tests by larval phase of sea-urchin, where relatively low number of development anomalies were observed (< 39 %). In the study (Dauvin, 2010), besides ecotoxicologic aspects there is described also effect of deposited red mud on the sea bed relief, on mezzo fauna and macro fauna in connection with dispersion of red mud in the sea environment. There were also studied changes in macro benthos of deep sea communities and risks of consumption of sea animals which were in contact with waste sludge from Al₂O₃ production. Last mentioned study was carried out at risk part of human population (women, children) and no cumulative risk for majority elements (Al, Fe, Cr, Pb, Cu, Mn, V and Zn) was confirmed.

According to EPA report (U.S. EPA, 1982; U.S. EPA, 2008) red mud after neutralisation is not classified as hazardous waste (Wang et al., 2008), because in four tested hazardous properties (corrosivity, reactivity, ignitability and toxicity according to TCLP (Toxicity Characteristic Leaching Procedure Test) it does not fulfil criteria for such classification. Performed ecotoxicity tests indicate that neutralised red mud can be widely reused not only as a building material, raw material for metal production in metallurgic industry or in glass production. Because of large surface, red mud after activation becomes excellent adsorbent and coagulant which can be used in remediation of soil in agriculture, mining industry - neutralisation of Acid Mine Drainage (Paradis et al., 2006), in removal of toxic metals in waste treatment plants, in catalysis in chemical industry etc. At many above mentioned applications of red mud, its benefit effect on environment was confirmed by experiments (e.g. improvement of soil properties after adding of red mud into unproductive (thin) clay or sand soils, or as a catcher of toxic inorganic and organic substances not only from soil, but also from water and air, as a gas cleaner. While heavy metals remain adsorbed and leachate contains safety levels of the metals even in low pH – acid rains). Despite huge amount of mentioned applications of the mud reusing, because of low economic profit rate the mud in many cases ends up at sludge beds.

5. Conclusion

From this literature survey it is obvious that there is extensive effort to reusing of waste mud from aluminium production. Nowadays, patent databases register worldwide more than 1 500 patents related to red mud and its reusing in different fields mentioned in this paper. On the other hand, it is claimed in general that utility of this patent is very low because of economic reasons. A lot of interesting references can be found at web sites directly dedicated to red mud reusing (Red mud project, 2011). Despite the fact that red mud is classified in some countries as hazardous waste because of strong alkalinity (according to OECD classification to Yellow list of waste under code GG 110-SRA), after neutralisation its effect on environment seems to be negligible from the toxicity point of view. A lot of leaching experiments were performed with waste mud with various extractive agents (so-called sequence extractive analysis), mostly to find out toxic metals leachability

(Singh, I. B. & Singh, D. R., 2002; Friesl, et al., 2003). In general limits for waste water were not exceeded in all these experiments. Reusing of waste sludge in building industry to produce final articles requires also performing of acute and chronic ecotoxicity tests by relevant biologic species. Results obtained up to the present (Maddocks et al., 2005; Orešcanin et al., 2003; Brunori et al., 2005b) have not confirmed toxic effects of tested articles or processed red mud on tested organisms (algae, worms, marine gastropods, etc.), meanwhile unprocessed red mud without neutralisation exhibits toxic effect on water organisms (Pagano et al., 2002).

In spite of intensive research of reusing waste mud from Al_2O_3 production, majority of the mud ends up on terrestrial deposits. In seaside areas, where the distance to the sea is not too long, efforts to deposit red mud into the sea after neutralisation by sea water occur (from 84 world producers of Al_2O_3 , deposits of the mud are on the sea bed only in 7 cases) (Agrawal et al., 2004), what results in contradictory reactions of environmentalists. In many countries the problem of sludge bed is solved by neutralisation of alkaline waste water, building of underground sealing wall from bentonite to stop leaching the waste water into surrounding ground water and subsequent recultivation of the sludge bed. One of the major limitations of the successful exploitation of bauxite residue is large transport cost necessary to transfer of waste mud from its disposal sites to the point of application.

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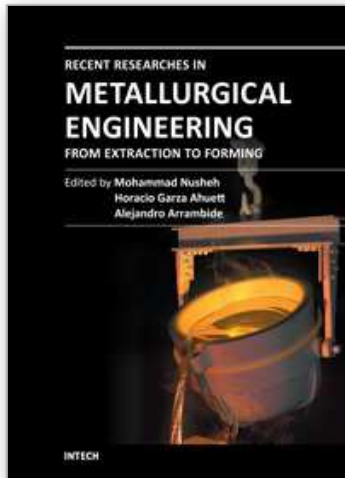
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Metallurgical Engineering is the science and technology of producing, processing and giving proper shape to metals and alloys and other Engineering Materials having desired properties through economically viable process. Metallurgical Engineering has played a crucial role in the development of human civilization beginning with bronze-age some 3000 years ago when tools and weapons were mostly produced from the metals and alloys. This science has matured over millennia and still plays crucial role by supplying materials having suitable properties. As the title, "Recent Researches in Metallurgical Engineering, From Extraction to Forming" implies, this text blends new theories with practices covering a broad field that deals with all sorts of metal-related areas including mineral processing, extractive metallurgy, heat treatment and casting.

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