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Review

Utilization of hazardous waste of black dross aluminum: processing and application-a review

Maya Lukita[1], Zaenal Abidin[2*], Etty Riani[3], Ahyar Ismail[4]

[1] Graduate School of Natural Resources and Environmental Management Science, IPB University, Bogor, Indonesia
[2] Department of Chemistry, IPB University, Bogor, Indonesia
[3] Department of Aquatic Resources Management, IPB University, Bogor, Indonesia
[4] Department of Resource and Environmental Economics, IPB University, Bogor, Indonesia

*corresponding author: abidinz.ipb@gmail.com

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Abstract

Aluminum black dross is produced by the secondary smelting process of aluminum. Aluminum black dross is classified as hazardous waste because it is reactive with water and produces substances and gases that are harmful to humans and the environment. Generally, aluminum black dross is managed by landfill method, but because it is produced in large amounts every year, the aluminum black dross needs to be utilized to reduce the impact on the environment. Aluminum black dross consists of large amounts of metal oxide and salts. The amount of metal oxide content in aluminum black dross can be used as raw material. This paper review types of processes for utilizing black dross aluminum as raw material in value-added products. aluminum black dross can be used as alumina, adsorbent, zeolite, composites, geopolymers, refractories, and fillers. By utilizing aluminum black dross waste into various products that have economic value, besides being able to protect the environment, it can also reduce environmental resource use.

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Introduction

The aluminum primary industry generates aluminum waste 30% to 60% of the raw material. Aluminum waste is classified into two types, namely skimming with an aluminum metal content of more than 45% and dross with an aluminum metal content of less than 45%. Dross is divided into two types, namely white dross generated from primary aluminum industrial with aluminum metal content 20% to 45% and black dross generated from aluminum secondary industry with aluminum metal content 10% to 20% (Tsakiridis, 2012).

Aluminum skimming and white dross have been reused for the recovery of aluminum metal because they still have high levels of aluminum metal, while the aluminum black dross has a low metal content, generally, it is not reused for aluminum recovery. Aluminum black dross has a composition of 20% to 50% metal oxide and 40% to 55% salt flux (Tsakiridis, 2012). Metal oxides are formed from metals that come into contact with air in the aluminum smelting process, while salt flux comes from the addition of salt flux in the aluminum secondary industry smelting process. Total aluminum contained in aluminum black dross is distributed as metal aluminum, aluminum nitride (AlN), corundum (Al_2O_3), and spinel ($MgAl_2O_4$) (López-Delgado et al., 2020).

Aluminum black dross from the aluminum secondary industry is hazardous waste according to European Waste Catalog and Hazardous Waste List with code 10 03 09*. Aluminum black dross is a hazardous waste because it is reactive with water and moisture which can produce toxic and flammable gases (Mahinroosta et al., 2019; Nguyen and Lee, 2019a; López-Delgado et al., 2020). During the aluminum black dross leaching process, most of the compounds dissolved and released ammonia and hydrogen gases. Hydrogen gas is produced from the reaction of aluminum metal with water, while ammonia gas is formed from AlN which reacts with moisture in the soil and with geothermal heat which leads to exothermic reactions releasing gases that are harmful to health (Ramaswamy et al., 2019b).

More than one million tonnes of aluminum black dross are produced per year by aluminum producers worldwide. This is causing considerable disposal costs and is a serious environmental problem. The management of aluminum black dross waste is mostly carried out by landfill methods, which raises other problems because it requires a large area of land for disposal and has a leaching effect that can cause water, soil and air pollution (Tsakiridis, 2012). Therefore, it is very important to make efficient use of all types of dross, both white dross, black dross and salt flux (Gil and Korili, 2016; Mahinroosta and Allahverdi, 2018; Chobtham and Kongkarat, 2020).

Utilization of Aluminium Black Dross

There are several processes of utilizing aluminum black dross. This paper classifies into three types, namely wet, dry and without process. Generally, the wet process is carried out by using an acid or base leaching process. The dry process is carried out by heating the sample to a temperature above 1000 ºC; generally, in this process, the addition of precursors is carried out. Combination processes can be carried out by combining wet and dry processes. The direct use of aluminum black dross is done without processing. Aluminum black dross before use, treated using hot water to remove salt flux, and activated by ball milling. A summary of the study utilization of aluminum black dross can be seen in Figure 1.

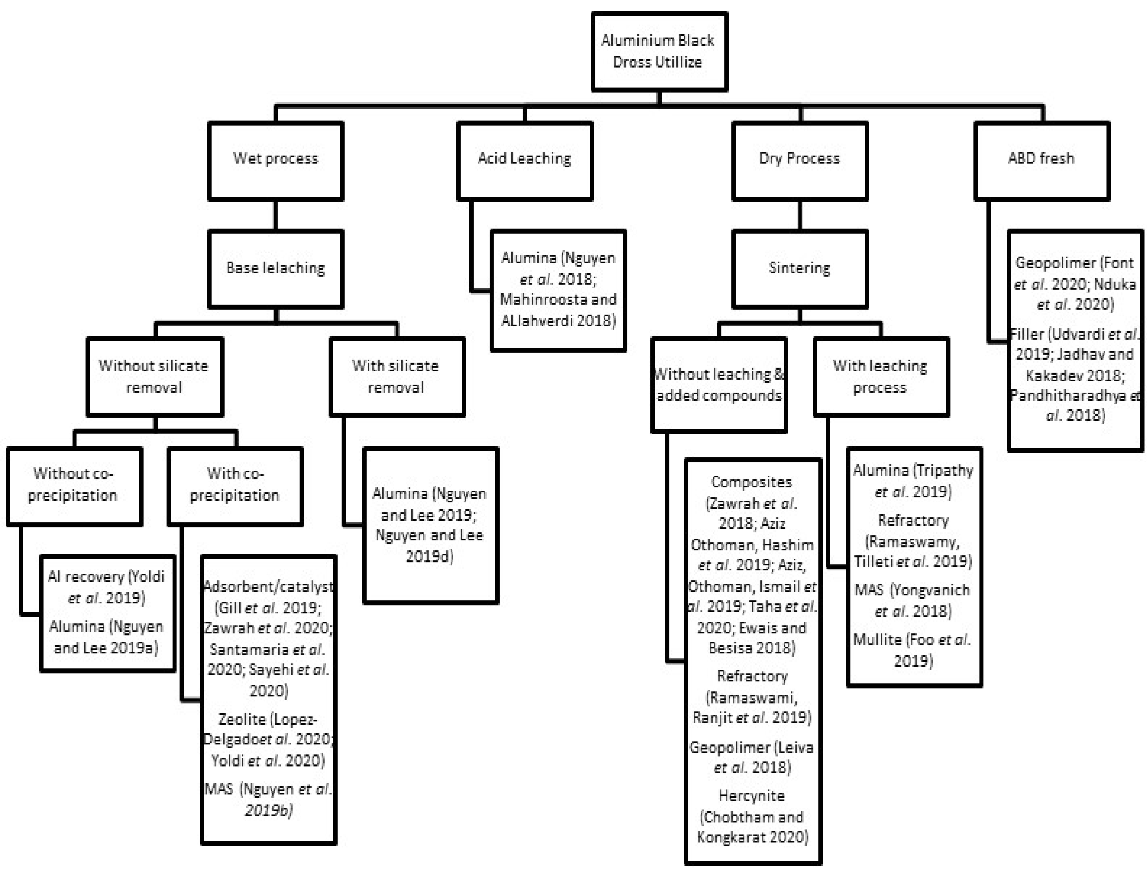

Figure 1. Summary of studies aluminum black dross utilization.

Recovery of aluminum metal

Recovery of aluminum metal on aluminum black dross can use reflux and electrostatic methods (Cao et al., 2019; Yoldi et al., 2019). Extraction of aluminum metal on aluminum black dross by reflux using NaOH solution and calcination. Ammonia and all other volatile compounds present in aluminum black dross can be removed (Yoldi et al., 2019). The chemical composition of aluminum black dross before and after the extraction process can be seen in Table 1.

Aluminum black dross is gray before calcination and brown afterwards. The color change is due to the removal of volatile compounds and the oxidation of some elements or molecules in this process. The results show that 19.2% of aluminum as aluminum metal and AlN in an original waste can be extracted and is not left in aluminum black dross residue after extraction. The value of aluminum oxide increases because some aluminum metals are oxidized during extraction (Yoldi et al., 2019).

Table 1. Composition of aluminum black dross before and after Al extraction with NaOH.

Compound	Chemical formula	Waste before Al extraction (wt %)	Waste after Al extraction (wt %)
Deliverable aluminum: metallic aluminum and aluminum nitride	Al + AlN	19.2	0
Corundum	Al_2O_3	21.7	25.1
Magnesium spinel	$MgAl_2O_4$	21.9	21.4
Calcium carbonate	$CaCO_3$	3.7	3.6
Silica	SiO_2	5.7	4.9
Magnetite	Fe_2O_3	1.2	1.2
Sylvite	KCl	0.9	0.3
Titania	TiO_2	0.6	0.6
Total crystalline structures	-	74.8	56.9
Non-crystal and LOI	-	25.2	43.1

Source: Yoldi et al. (2019).

Recovery of aluminum metal by an electrostatic method based on physical properties of components contained in aluminum black dross. Aluminum recovery with multi-stage electrostatic separation method consists of three stages, namely (1) roll-type electrostatic separator, for preliminary separation; (2) pulse charging device, to improve fine-grained charging status; and (3) free-fall separator, to determine the greatest possible recovery of aluminum in secondary concentrations. Electrostatic multi-stage separation can increase aluminum from 8% to 37% (Cao et al., 2019).

Recovery of alumina

Alumina has amphoteric properties, which are soluble in acidic or alkaline conditions. Acid leaching results in some of the metal oxides in the aluminum black dross being dissolved with the alumina. Alkali dissolving is selective for alumina because the oxides of Ca, Fe, and Mg do not dissolve, which will result in an aluminate solution with a small amount of silicate (Nguyen and Lee, 2019b). The salts contained in the aluminum black dross are removed by hot water and then activated by a ball milling process. The speed of the milling process in NaOH leaching greatly influences oxides of Si, Ca, Fe, Mg, and Ti leaching behavior found in aluminum black dross (Nguyen and Lee, 2019a). In HCl leaching, the reported time and speed of ball milling did not have a significant effect on aluminum black dross leaching (Nguyen et al., 2018).

The development of the NaOH leaching method for the synthesis of mesoporous alumina is carried out using a new recycle procedure consisting of leaching, co-precipitation, separation by selective dissolution, re-precipitation, precipitation and calcination. Mesoporous alumina has a purity of more than 98% and a particle size that can be excellent candidates as a catalyst, catalyst support, or adsorbent (Mahinroosta et al., 2019). The aluminate solution leached with NaOH slightly dissolves silica. There are several ways to remove silica that is dissolved in an aluminate solution. Hydrocalumite is reported to remove silicate dissolved in aluminate solution by easy synthesis. Hydrocalumite can selectively adsorb silicates and has a high adsorption capacity (Nguyen and Lee, 2019b). Silicate removal can be carried out by coagulation using Polyacrylamide (PAM). The adsorption of silicates to PAM occurs due to the formation of hydrogen bonds. Optimal conditions can be removed easily most of silicon gel with pressurized filtering (Nguyen and Lee, 2019a). Recovery of alumina with NaOH leaching can be continued with precipitation using H_2O_2. Under optimal conditions, the percentage of precipitation of aluminum hydroxide is 99% (Nguyen and Lee, 2019c).

Alumina can be recovered using the pyrometallurgical method. The combustion process is carried out by mixing aluminum dross with sodium carbonate. Increasing the amount of sodium carbonate during combustion can increase the recovery of alumina, with the sodium carbonate concentration of 10% recovery obtained to be 90%, but after 10% recovery, it becomes stable. The optimal temperature obtained is at a temperature of 800 ºC with an optimal combustion time of 1 hour. Subsequently, the leaching was carried out using NaOH, from the report that only 2% NaOH was required to dissolve 90% alumina with the optimal temperature and leaching time obtained at 70 ºC for 1 hour (Tripathy et al., 2019).

Synthesis of alumina from aluminum black dross can be carried out by HCl leaching. Particle size has a significant effect on the recovery efficiency of alumina with HCl leaching; a smaller particle size can increase the extraction efficiency because the specific surface area is larger so that it increases the contact area and can speed up leaching time. However, particle size smaller than 38 μm can decrease the efficiency of alumina recovery because other metal oxides compete with aluminum to absorb hydrochloric anions. Increasing initial leaching time increases alumina recovery efficiency then decreases thereafter. That is because the reaction between metal oxides and HCl occurs and reduces the chloride anion available for Al_2O_3. The reaction of HCl with aluminum black dross is exothermic so that the equilibrium concentration of $AlCl_3$ decreases with temperature. The optimal temperature leaching is 85 °C. Recovered alumina shows the gamma phase as the main phase, kappa phase and theta phase as the minor phase (Mahinroosta and Allahverdi, 2018).

Adsorbent and catalyst

Aluminum black dross can be utilized as a hydrotalcite compound by extracting aluminum followed by co-precipitation. Hydrotalcite with a higher absorption capacity than has been reported in the literature can be prepared using cobalt, magnesium, nickel nitrate and Na_2CO_3 as precursors (Gil et al., 2018). Synthesis of ZnTiAl double-layer hydroxide (LDH) was carried out by adding a 3:1 molar ratio of Zn/(Al, Ti) as precursors; it is very effective in removing diclofenac (Santamaría et al., 2020). Synthesis of Mg-Al and Ni-doped LDH nanoparticles was carried out by adding $Mg(NO_3)_2{\cdot}6H_2O$ and $Ni(NO_3)_2.6H_2O$ and $Al(OH)_3$ (Zawrah et al., 2019).

Waste with aluminum and silicon content can be utilized as an adsorbent and catalyst. Aluminum black dross can be used as one of the materials because it is Al_2O_3 based waste with the highest content 42% to 88% (wt/wt). Amorphous silica-alumina synthesis is very similar to the process of amorphous silica synthesis by the addition of alumina precursors. Using compelling, precipitation, sol-gel, and air gel techniques can be used to synthesize silica-rich alumina with amorphous silica with crystalline c-alumina copresence (Sayehi et al., 2020).

Zeolite

Zeolite X, zeolite A, and sodalite can be synthesized from aluminum black dross by a hydrothermal method involving alkaline aluminum extraction with residual waste separation, and hydrothermal treatment at low temperatures and long reaction times (Yoldi et al., 2020). Zeolite synthesis was carried out by hydrothermal method and addition of Na_2SiO_3 (water glass) to increase the Si/Al ratio of the zeolite synthesized (López-Delgado et al., 2020; Yoldi et al., 2020).

Composite

Aluminum black dross containing metallic aluminum and oxide can be used as a composite material with added another compound and sintering. Al_2O_3/Al porous composites can be made with the addition of graphene. The increase in the amount of graphene added and the sintering temperature led to an increase in the mechanical properties and electrical conductivity of the composites (Zawrah et al., 2018). Alumina-spinel hollow composite fiber membrane can be made by combination phase inversion method and sintering. Composites were successfully synthesized from aluminum black dross after washing and calcination at 1100 ºC, causing the formation of Al_2O_3 and $MgAl_2O_4$ (Aziz et al., 2019a; Aziz et al., 2019b). The composite hard outer layer can be made by adding SiC, TiO_2, and sintering. The two composite layers formed after sintering contain phases and have different properties. The two composite layers formed after sintering contain phases and have different properties. The outer layer is solid and hard, while the layer has lower pores and hardness (Taha et al., 2020).

Magnesium aluminum titanate (MAT) ceramic composites can be made from aluminum black dross (as a source of Al_2O_3 and MgO) and rutile ore powder (as the source of TiO_2) at 1300 ºC. X-ray diffraction (XRD) analysis results show the presence of $Mg_{0.3}Al_{1.4}Ti_{1.3}O_5$ and $MgAl_8Ti_6O_{25}$; this indicates the formation of magnesium aluminum titanate (Ewais and Besisa, 2018). The possible reactions can be seen in Equations 1, 2, 3, and 4.

$$Al_2O_3 + TiO_2 \rightarrow Al_2TiO_5 \quad (1)$$
$$MgAl_2O_4 + 2TiO_2 \rightarrow MgTi_2O_5 + Al_2O_3 \quad (2)$$
$$MgAl_2O_4 + 3TiO_2 \rightarrow Al_2TiO_5 + MgTi_2O_5 \quad (3)$$
$$3MgAl_2O_4 + 4Al_2O_3 + 13TiO_2 \rightarrow 7Al_2TiO_5 + 3MgTi_2O_5 \quad (4)$$

Geopolymer

Geopolymers can be made from alumina with silica. Aluminum black dross can be used as a source of alumina for geopolymers. Preparation of geopolymers from aluminum black dross and fly ash (as a silica source) in ratio 1:1, which is added to the activating solution. The resulting geopolymer is porous which can absorb noise. Pores in geopolymers are formed from the reaction of aluminum with water and alkalis (Leiva et al., 2019). Application as a substitute portland cement in concrete by mixing fresh concrete with aluminum black waste. The results suggest replacing portland cement with 10% aluminum black waste which can be used to produce concrete with normal strength (Nduka et al., 2020).

Geopolymer Cellular Concrete (GCC) and Alkali Activated Cellular Concrete (AACC) can be made from aluminum black dross with the addition of Fluid Catalytic Cracking Residual Catalyst (FCC) and Blast Furnace Slag (BFS). The aluminum black dross that is loaded into the milling is mixed with the precursor, adding FCC for GCC and BFS for AACC to aid in the aeration process. This method can increase productivity and reduce costs. One of the most influencing parameters on the effectiveness of aeration is aluminum oxidation. Controlled aeration reactions are very important to be able to develop a good void structure. The addition of 2% aluminum black dross produces GCC and AACC with similar physical and mechanical characteristics when adding 0.2% commercial aluminum (Font et al., 2020).

Filler

Aluminum dross can be utilized as an asphalt filler. One of the roles of filler in asphalt is to increase mechanical strength and stiffness. Strength can be increased by increasing cohesion between mineral and bituminous materials. In addition, the shape of the mineral material used in the asphalt mixture will affect the internal friction of the mineral framework and the carrying capacity of the asphalt pavement (Udvardi et al., 2019). Cold bituminous emulsion mixes (CBEM) are widely used as bitumen in India, which have low strength. The quality of CBEM can be improved by the addition of an aluminum black dross. The stability and tensile strength values of the mixture containing aluminum dross were higher than the mixture containing Portland cement. The optimal amount of addition of aluminum dross is 2% (Jadhav and Kakade, 2019).

Aluminum black dross can be utilized for concrete production as a filler or pozzolana. Aluminum black dross has many silicates which are a determining factor in slowing down setting times. Large amounts of water can be absorbed by the aluminum black dross because it has a high surface area. During cement hydration, excess $Ca(OH)_2$ generated from aluminum black dross reduces the pozzolanic reaction in the cement matrix. This is the cause of the slowing effect of setting time. Therefore, aluminum black dross is suitable as a material that can slow down the setting time rate and is suitable for concrete applications for hot weather. Cement replacement with aluminum black dross by 15% gives better results compared to conventional concrete (Panditharadhya et al., 2018).

Magnesium aluminate spinel

Magnesium aluminate spinel ($MgAl_2O_4$) can be synthesized from aluminum black dross by leaching and co-precipitation processes. Aluminum hydroxide solution is obtained by dissolving aluminum black dross with aqua regia. Co-precipitation was carried out by adding MgO with a molar ratio of 2:1 (Al: Mg). During the co-precipitation process, the pH of the solution was maintained at 8.5 with the addition of NH_4OH. The complete spinel conversion can be carried out at 1000 ºC for 5 hours. The reaction equation can be seen in Equations 5, 6, and 7 (Nguyen and Lee, 2019d). Spinel-based ceramic pigments can be synthesized from aluminum black dross and oxide precursors by the solid-state method. Black aluminum dross is used to replace alumina material. As the chromophore, cobalt ion is selected to produce a blue color.

$$Mg^{2+} + Al^{3+} + 5\,OH^{-} \rightarrow Mg(OH)_2 + Al(OH)_3 \quad (5)$$

$$\tfrac{1}{2}\,Mg(OH)_2 + Al(OH)_3 \rightarrow \tfrac{1}{2}\,MgO + \tfrac{1}{2}\,Al_2O_3 + 2\,H_2O \quad (6)$$

$$MgO + Al_2O_3 \rightarrow MgAl_2O_4 \quad (7)$$

The spinel phase is formed when fired at 1100 °C, a higher level of purity can be obtained over a longer time. Pigments produce reflections in the blue and blue-green spectrum, as evidenced by UV-vis spectroscopy and colorimetry (Yongvanich et al., 2018).

Refractory

Refractory is a material that can withstand high temperatures and can maintain physical and chemical stability. Generally, refractories are synthesized from silica-alumina geomaterials, but the latest trend is made from ceramic. Aluminum oxide and magnesium aluminate are basic compounds contained in the refractory composition, which are products that have low thermal conductivity and high-temperature shock conditions of more than 1000 ºC. Aluminum black dross can be an alternative material because it has these compounds. Aluminum black dross contains aluminum nitride which is a compound that has a high thermal conductivity which is not ideal for refractories; therefore, it is necessary to remove AlN (Ramaswamy et al., 2019a; Ramaswamy et al., 2019b;). Zirconia addition can be performed on refractory synthesis to increase thermal shock resistance with a 1:1 ratio with 10% polyvinyl alcohol (Ramaswamy et al., 2019a).

Mullite

Mullite-based ceramics can be synthesized from aluminum black dross (Chargui et al., 2018; Foo et al., 2019). Synthesis of mullite from aluminum black dross by adding fly ash and HCl leaching, then compacting and sintering. The effects of sintering temperature, acid leaching, and Al_2O_3/SiO_2 ratio affect the chemical, physical, and thermal expansion properties of the sample. The results showed that the ratio of mixing and washing with acid had a positive effect on the mineralogy, crystallinity, and macromorphology of the samples after sintering. At the sintering temperature of 1500 °C, a ceramic with a high mullite composition with good crystallinity is produced and has excellent thermal expansion properties (Foo et al., 2019).

Synthesis of mullite from black dross aluminum with added kaolin shows that amount of mullite formed increases with high temperature. At 1500 ºC the crystallization of the mixture was almost complete. The morphology of the mullite formed is the bimodal phase, which is the primary and secondary phase. Primary mullite is formed from the gradual collapse of metakaolin from kaolin from 990 °C, which has an elongated crystal form. Meanwhile, secondary mullite which has a granular form is formed from the reaction of alumina from black aluminum dross and excess silica in kaolin (Chargui et al*.,* 2018).

Hercynite

Aluminum black dross as a source of Al_2O_3 can be utilized for the synthesis of hercynite ($FeAl_2O_4$). Synthesis was carried out by heating aluminum black dross at 1200 °C for 1 hour, then compacting it into a substrate using a hydraulic press. The substrate was associated with two types of iron chips containing 0.8% of the aluminum black dross sample. Sintering was carried out at 1550 °C for 6 hours. The results of characterization of samples found that the final product was hercynite which had a dark gray solid phase. Hercynite is formed due to the interaction of Al_2O_3 from aluminum black dross with Fe/FeO (the result of oxidation of iron chips due to excess oxygen in the system) in the iron chip. The carbon content in iron chips is used to influence the formation of FeO, which ultimately affects the formation of hercynite (Chobham and Kongkarat, 2020).

Conclusions

Aluminum black dross is a hazardous waste because it has reactive characteristics. Generally, the processing of aluminum black dross by landfill method, but the amount produced annually is very large, so other processing alternatives are needed. By removing salts, aluminum black dross can be used for recovery of aluminum metal and alumina or into various types of products such as adsorbents, catalysts, zeolites, composite materials, fillers, geopolymers, refractories, magnesium aluminate spinel, mullite, and hercynite. Besides being used to avoid environmental pollution, aluminum black dross can also increase the added value of waste into a product. It is necessary to consider choosing the type of utilization of aluminum black dross which is economical, being a value-added product, and also environmentally friendly.

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