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Effect of Temperature and Ratio for the Immobilization of Zinc Oxide in Aluminium Rich Ceramics

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Abstract: *Zinc is one of the dangerous elements that is frequently present in municipal solid waste incinerator ash. When exceeding specific limits, there are various diseases and disorders caused by accumulation of zinc in nature as their characteristics is not biodegradable. By using zinc oxide to imitate zinc sludge, this study reveals the immobilization mechanisms that occur when zinc and ceramic precursors are used as waste-to-resources strategies. From the result, immobilization occur better at higher temperature at 10000C where the formation of Al2O4Zn is higher. Meanwhile, ratio 1:1 of ZnO and Al2O3 shows a better result of formation Al2O4Zn. Hence, it is possible to use the sintering process with precursors to turn hazardous wastes into secure products where the heavy metals can form phases that can demonstrate adequate stability due to irreversible phase transformation.*

Keywords: zinc, waste-to-resources, sintering, precursor, immobilize

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1. Introduction

Due to the increase of the industrial waste, an alternative had been made to reduce the waste in a proper way by incorporated them into fired clay bricks. This is because, even though the industrial waste had been treated, there are still probability of the treated waste will contain toxic elements. Utilization of waste into brick seems to reduce negative effects of their disposal to the environmentally friendly way (Munoz Velasco et al., 2014). Many attempts have been made to incorporate waste in the production of bricks using organic or inorganic waste. Practical solution had been observed to the pollution problem by recycling the wastes and incorporating them into building materials. Utilizing wastes in clay brick can have some beneficial effects on its qualities (Inecar, 2000).

Any metallic element with a relatively high density that is hazardous or poisonous even at low concentrations is referred to as a heavy metal. Yet, being a heavy metal has more to do with chemical properties than density. Heavy metal includes lead (Pb), zinc (Zn), arsenic (Ar), cadmium (Cd), mercury (Hg), silver (Ag), chromium (Cr), copper (Cu), iron (Fe) represents the most common heavy metal contaminant. Unlike organic contaminants, these metals cannot be degradable to harmless products, such as carbon dioxide, and the cleanup usually requires their removal. Since heavy metals are carcinogenic and mutagenic substances, their presence poses a serious threat to all living things (Alaboudi et al., 2018).

All those heavy metals are causing a major environmental pollution known as heavy metal pollution in developing world. These metals come from a wide range of sources. One of them is industrial wastewater. This waste increases rapidly from daily activities of production by industries to meet the needs of the growing population of human (Kadir&Sarani,2012). Zinc has been relatively non-toxic especially if taken orally (Nolan, 2003). However, excessive zinc intake can lead to system dysfunction and hinder growth and reproduction (Inecar, 2000).

There are some ways to reduce the waste such as by using phytoremediation treatment, dumping and thermal treatment. A developing, cost-effective remediation technique called phytoremediation utilizes plants to extract metals from contaminated soil (Khalid et al., 2017). Meanwhile, since long-term impacts and external costs are not considered, land filling is the most affordable method of disposing of municipal solid waste (MSW). There are methods in research that aim to detoxify harmful residue or render them dormant so that they could be reused or at the very least deposited without risk. These methods are based on immobilization with cement, wet chemical treatment, or thermal treatment.

A promising method for the treatment of sludge that contains metals is the conversion of zinc sludge into usable ceramics, as this reduces the leachability of harmful metals by immobilizing them in a stable matrix (Xu et al., 2008). Hazardous metal ions are bound into the glass or crystalline phases of ceramic products to help them functional (Machado et al., 2011). With carefully controlled crystallization and devitrification of a glass matrix, the method often produces ceramic goods. (Rawlings et al., 2006). In a prior work, it was shown that ceramic matrices made of aluminate and silica-aluminate could immobilize up to 99% of lead and be chemically resistant (Lu et al., 2013). It has also been stated that hematite can be manufactured using metal-containing trash to effectively stabilize dangerous metals (Pinakidou et al., 2007). Thus, for the inexpensive ceramic precursor that used to immobilize dangerous lead in sludge, hematite was employed.

In the present study, a synthetic brick had been made by using material (ZnO) representing zinc sludge was thermally heated with aluminum rich ceramics at different Zn/Al molar ratio. The zinc sludge was studied to immobilize inside the brick and new compound formed in the brick. The formation of crystalline species inside the synthetic brick was identified by using X-ray diffraction. The main aim of this study is to identify crystallization of zinc oxide (ZnO) and Aluminum oxide ($Al_2 O_3$) with different temperature and ratio.

3. Methodology

3.1 Preparation of material

Zinc oxide (ZnO) was chosen as material to represent heavy metal in sludge. Aluminum oxide (Al_2O_3) represent for component in the clay. ZnO and Al_2O_3 bought from Scientific Bersatu Sdn Bhd. Al_2O_3 were calcined in air at 300°C for 3h to activated Al_2O_3 into γ Al_2O_3 to increase reactive site of Al_2O_3. Its high specific surface area and high catalytic activity will help in increasing reactive site for ZnO to react with γ Al_2O_3.

3.2 Mixing of sample

The γ Al_2O_3 with ZnO were mixed in conical flask with bench top shaker in water slurry with γ Al_2O_3/ZnO molar ratios of 1:1 and 1:9 for at 50g of weight added with 250 ml of deionized water for each ratio for 18 hours in order for it to perfectly homogenized. The ratio ZnO is constant, while the ratios of Al_2O_3 are variable. The slurry samples were then filtered using

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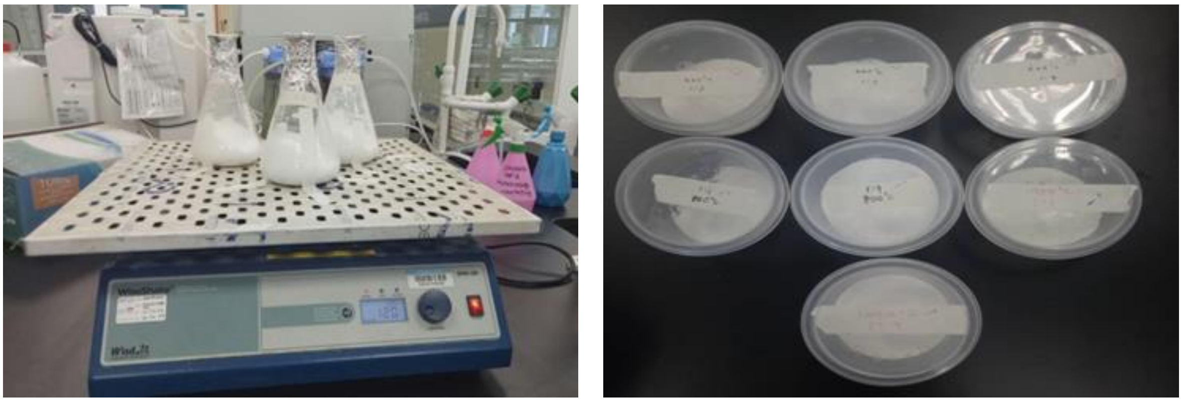
filter paper and then dried in air for two days. The samples were sintered at different target temperatures which are 600°C, 800°C and 1000°C. After the completion of firing process, the sample had not been removed until they become cool to 30^0C. After sintering, the samples are grounded by using agate mortar for XRD analysis. Table 1 shows the summarization of each sample in the study.

Table 1: Summarization of Samples

Compound	Ratio	Total Weight (g)	Sintering Temperature (^{0}C)
ZnO : Al_2O_3	1:1	50	600
ZnO : Al_2O_3	1:1		800
ZnO : Al_2O_3	1:1		1000
ZnO : Al_2O_3	1:9		600
ZnO : Al_2O_3	1:9		800
ZnO : Al_2O_3	1:9		1000

3.3 X-ray diffraction

The powder for XRD patterns were recorded on Bruker D8 advanced x-ray powder diffractometer equipped with Cu kα radiation and LynxEye detector. The diffractometer was opened at 40 kV and 40 mA and the 2θ scan range was from 10° to 90°, with a size of 0.02° and scan speed of 0.3 s per step.

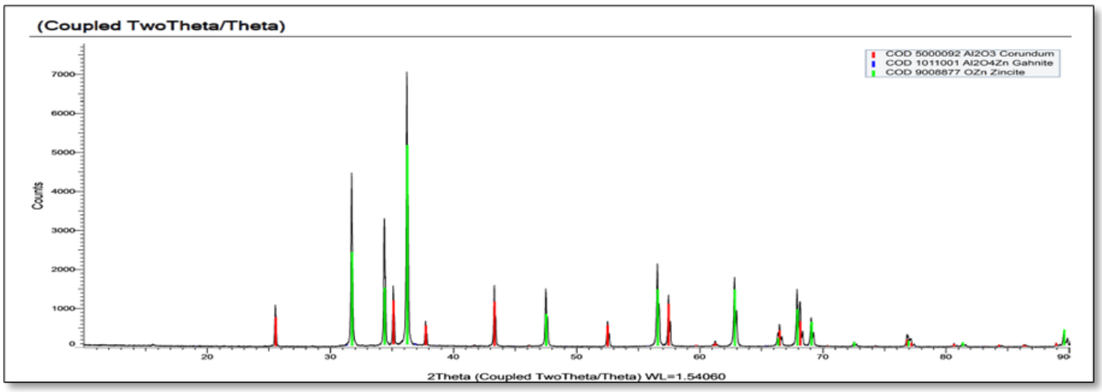


a b

Figure 1: a) Mixing Process of Zno & Al_2O_3 b) Final Samples after Sintering

4. Result and Discussion

Figure 2 shows the XRD analysis of the sintered samples of 1:1 ratio of ZnO/Al_2O_3 with the range of 600 °C, 800°C and 1000 °C.



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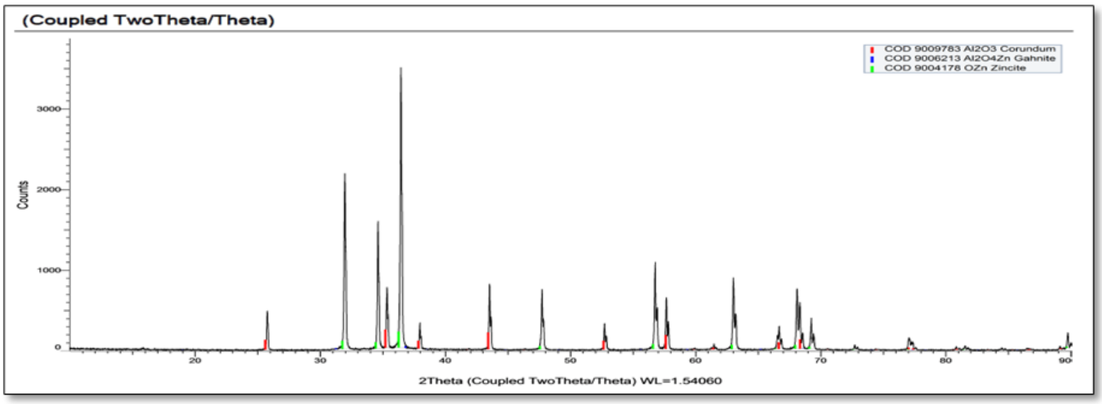

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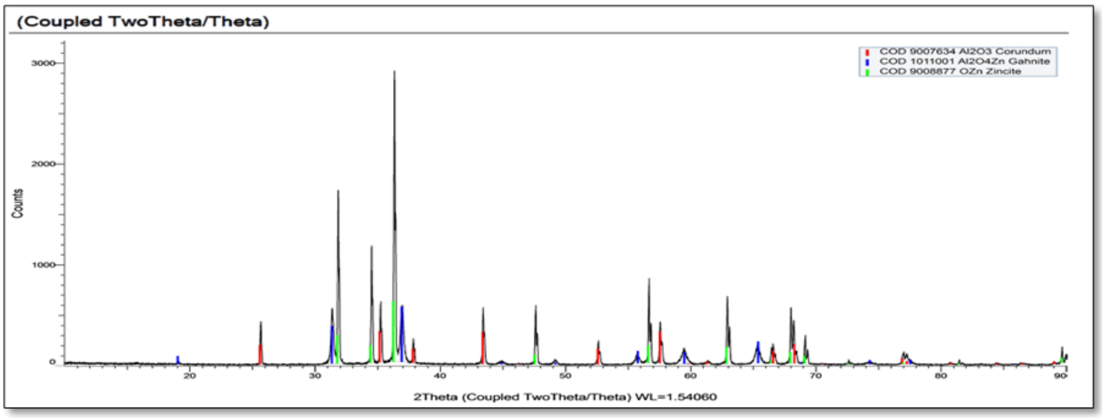


c

Figure 2: a) XRD analysis for 600°C b) XRD analysis for 800°C c) XRD analysis for 1000°C

Based on Figure 2, the peak shows that there is more than one angle expressed from the catalyst to form the compound. The higher the formation of Al_2O_4Zn, the less harmful of the compound can be reduce. The higher the Al_2O_4Zn being form, the easier the immobilization of compound can be increased. From this analysis, the formation of Al_2O_4Zn is indicate as predominant product phase after the sintering process. Therefore, the potential reaction before ZnO and Al_2O_3 are shown below:

$$ZnO + \gamma\, Al_2O_3 \rightarrow Al_2O_4Zn$$

Figure 3 show the percentage of different temperature for the crystallization of the compound in the sample which are ZnO, Al_2O_3 and Al_2O_4Zn of ratio 1:1 at 600 °C, 800°C and 1000°C.

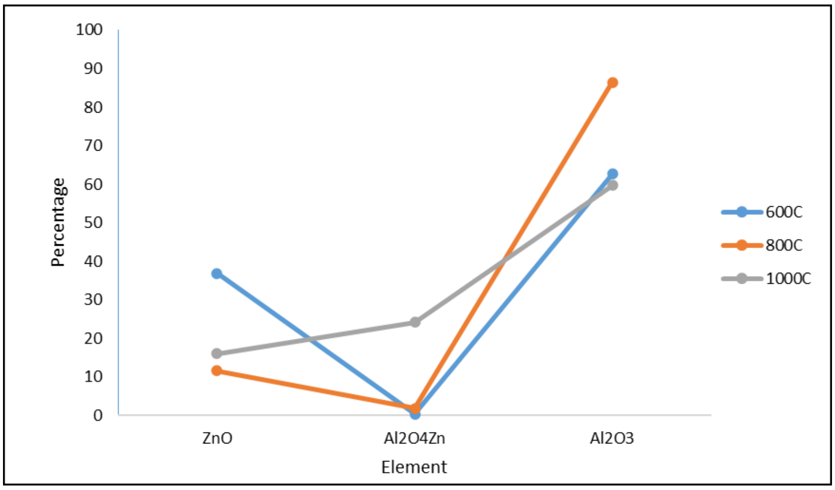
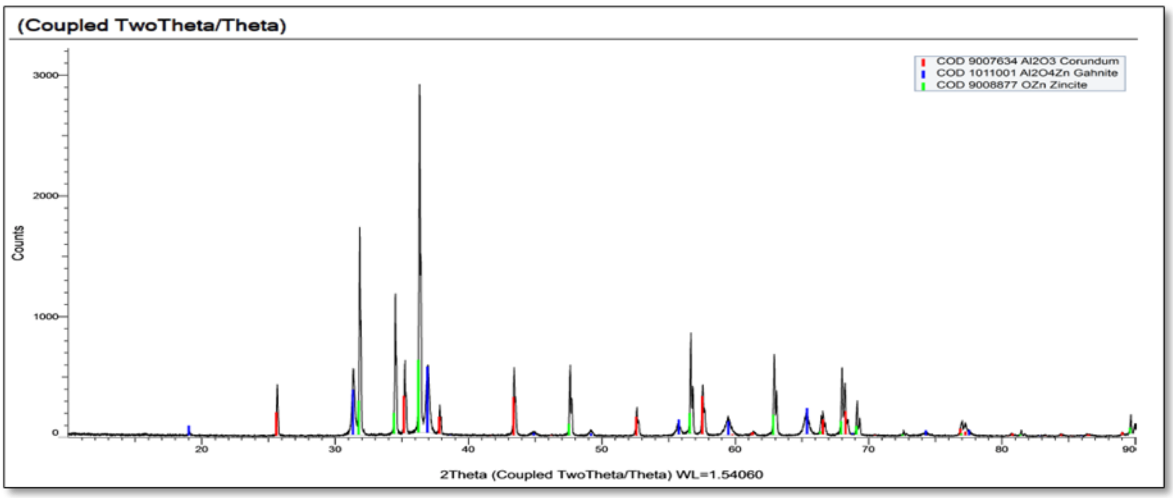


Figure 3: Percentage of Formation ZnO, Al_2O_3 and Al_2O_4Zn at Different Temperature of Ratio 1:1

From Figure 3, the percentage of each compound, ZnO, Al_2O_3 and Al_2O_4Zn in different temperature of 1:1 ratio had been obtained from XRD analysis. At temperature from 600°C to 1000°C, average percentage formation of ZnO is decreasing but the lowest percentage of ZnO formation at temperature 800°C. Meanwhile, at temperature from 600°C to 1000°C, the percentage formation of Al_2O_4Zn increasing by 0.4%, 1.9% and 24.2%. The formation of Al_2O_3 increasing at temperature 600°C to 800°C, while decreasing at temperature 1000°C.

The difference in sintering temperature of samples exposed in the experiment was caused by kinetic energy. Kinetic energy of a compound is the energy that it possesses due to its motion. When the sintering temperature increases, it contributes to higher kinetic energy to maximize the formation and therefore it increases the energy to react. Due to higher energy, it leads to the collision of molecules in compound and provide more free space to make the molecules speed up and thereby gain energy (Aramide, 2015; Xu et al., 2008). Therefore, the purpose for more formation Al_2O_4Zn are to help in immobilization in the sample. This had been proved that the higher sintering temperature, the higher formation of Al_2O_4Zn in the Figure 3.

Figure 4 show that analysis of sintered sample of 1:1 and 1:9 ratio of ZnO/ Al_2O_3 at same temperature which is 1000°C.



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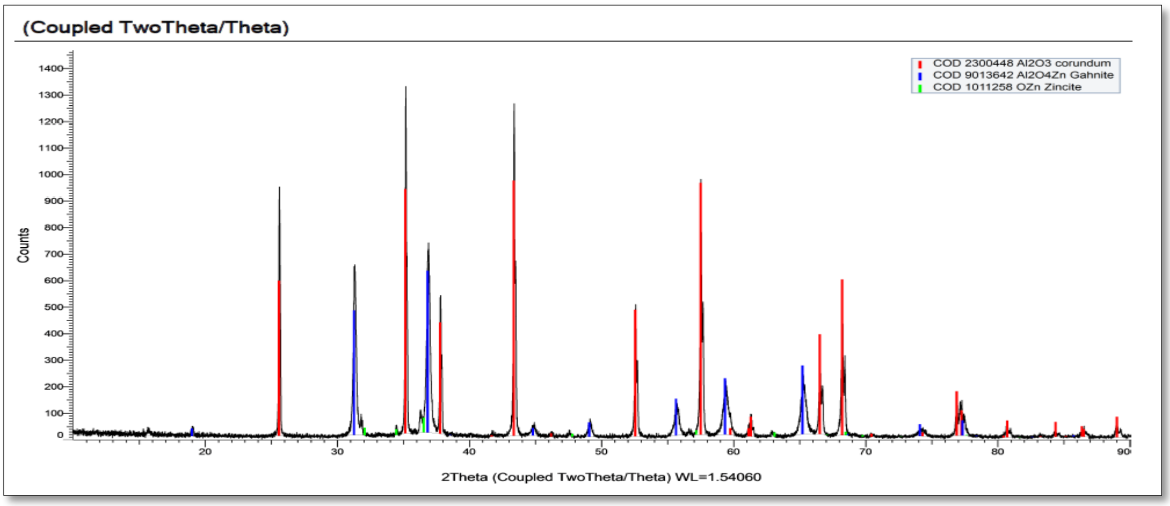

b

Figure 4: XRD analysis of ZnO, Al_2O_3, and Al_2O_4Zn for a different ratio of ZnO and Al_2O_3
a) Ratio for 1:1 b) Ratio for 1:9

By referring to the XRD analysis, the peak for ratio 1:1 and 1:9 is same with XRD analysis for the previous temperature. It can be seen on the Figure 4 that the peak appear in the analysis are same because the formation of the compound proven to be same compound which are ZnO, Al_2O_3 and Al_2O_4Zn. Even though the formation of compound appears to be the same but the percentages for each compound are different.

Figure 5 show the percentage of different ratio of 1:1 and 1:9 for the formation of compound in the sample which are ZnO, Al_2O_3 and Al_2O_4Zn at temperature 1000 °C.

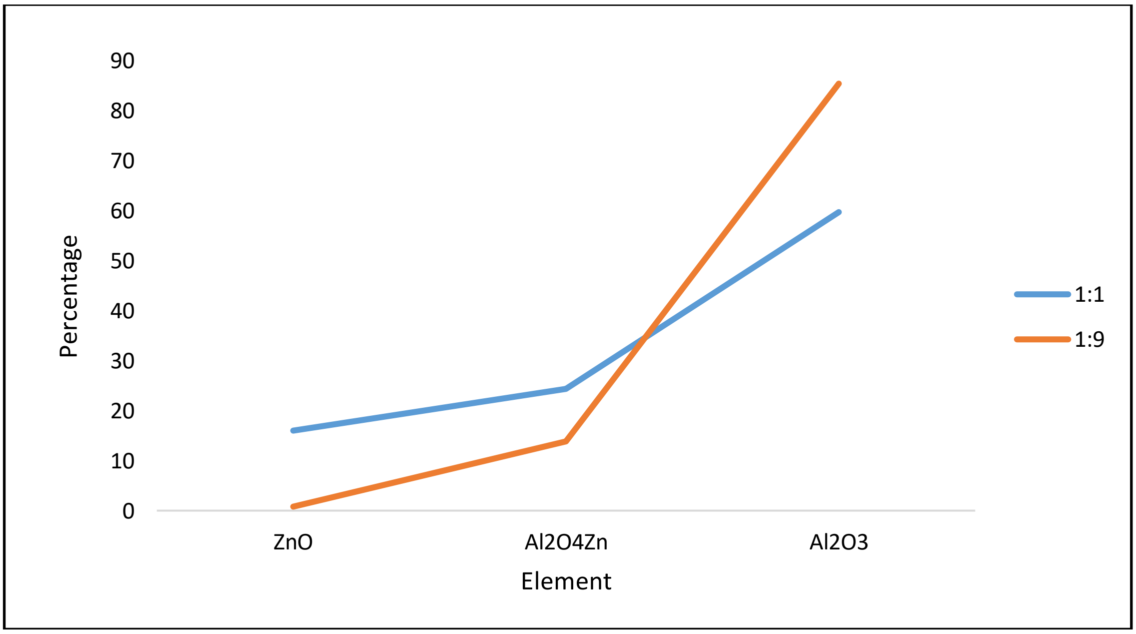

Figure 5: Percentage of formation ZnO, Al_2O_3 and Al_2O_4Zn at different ratios at temperature 1000°C

Based on the Figure 5, it seems that each compound which are ZnO, Al_2O_3 and Al_2O_4Zn increasing in both ratios. The ZnO in 1:1 ratio gives the percentage result which is 16% while in 1:9 ratio ZnO appear to be only 0.8%. Al_2O_4Zn give percentage about 24.3% in 1:1 ratio, while in 1:9 ratio the percentage is 13.8%. Therefore, in ratio 1:1 and 1:9 Al_2O_3 has highest percentage in the compounds which are 59.7% and 85.4% respectively.

In the figure above, the ZnO is decreasing in formation of the Al_2O_4Zn. Therefore, the compound acts as the limiting reactant. As the amount of the ZnO in the reaction decreases, the reaction will become slower. When the ZnO completely reacts, the whole reaction will stop. In addition, large amount of ZnO is needed to maximize the formation of Al_2O_4Zn.Based on this study, the 1:9 ratio is not recommended to immobilized ZnO in the Al_2O_3 as the amount of ZnO too neglible because it departs from our purpose which is to reduce the amount of heavy metal and to incorporate it inside the clay precursor.

5. Conclusion

In conclusion, the incorporation of heavy metal into clay precursors has been proven that this method can be one of the alternative ways to reduce the amount of heavy metal waste being untreated and dumped into the land or sea. These findings perceive to be a beneficial guide to immobilize hazardous metal in crystal form. The zinc content from wastewater treatment sludge can be successfully incorporated into alumina via thermal treatment process since the amount showed a decline after being treated in high temperature. Thus, it is not harmful to environment and humankind to be used as building material as result shows that it has been immobilize from the XRD analysis.

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