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Effect of different ratio and sintering temperature for the immobilization of simulated zinc sludge in aluminium rich ceramics

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Abstract: High amount of heavy metal waste produced from the industrial activities has become an important global issue for several decades due to its hazardous effect on the human and environment. It is difficult to be eliminated as it is harmful to the human and environment due to their non-biodegradable nature. Zinc, Zn is a typical heavy metal which is not biodegradable and could be accumulated in nature, causing various diseases and disorders when exceeding specific limits. Hence, the aimed of this study is to investigate the formation of new compound between the aluminium oxide, Al_2O_3 and zinc oxide, ZnO that lead to the immobilization of this compound in fired clay brick. Al_2O_3 is known to have the second highest amount of clay precursor in brick manufacturing. The Zn is chosen because it is one of the most hazardous metals which is not biodegradable, that are commonly found in industrial waste. The immobilization happen when heavy metal been sintering together at high temperature with another compound to form a new combination of product that less hazardous. A research has been conducted and able to stabilize the heavy metals with well conducted thermal treatment schemes. The experiment showed that the stabilization of Zn only appear when it is exposed at high temperature; 1000°C and high ratio; 1:1 resulting in high formation of $ZnAl_2O_4$. The incorporation of heavy metal into clay precursor through thermal treatment process seen to help in clay dependency and heavy metal pollution. Thus, this findings may be a beneficial guide to immobilize hazardous metal in crystal form and to be used as the raw material in the development of ceramic products since it is not harmful to environment and humankind to be used as building material.

Keywords: heavy metal, immobilization, phase transformation, thermal treatment process, x-ray diffraction.

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1. Introduction

The increasing in industrial waste number is really at the worried state. Industrial waste is the waste produced by industrial activity during manufacturing process. Most of the industrial waste contains heavy metal [1]. Heavy metal pollution has occurred since the pre-modern period when the mining and related human activities have been the main cause of contamination. After the industrial revolution, the heavy metals have been produced in a very large scale from the production activities. In 2000s', heavy metal pollution has been reduced through e-waste treatment, incineration and dumping sites including crushing, separating and open field burning which cause air, soil and water pollution in the middle of 20th century. Its high concentration and density may harm and kill living organisms [2].

Several researchers had attempts various experiments to incorporate waste in the production of brick for example limestone powder dust (LPW) and wood sawdust (WSW) [3], process waste tea (PWT) [4], and incinerator fly ash slag (MWSI) [5] and had proven that by adding incorporate waste it will be environmentally advantageous and increase the brick properties performance. Therefore, by adding industrial waste in clay brick might give the same or even better result as another additional component in the properties of the fired clay brick. From the research by Vicenzi et al, (2005) [6]. Aluminium-rich, Al-rich sludge was use as raw material for ceramic products. Based on the stabilization effect caused, the type of sludge rich in aluminium content and its heated and dried forms is expected to serve as a potential ceramic resource that can stabilize hazardous metals.

Heavy metal pollution that cause by metal ions persist in the environment due to their non-biodegradable nature is currently a major environmental problem. Heavy metal only can be transformed into less toxic species because it cannot be breakdown by chemical or biological processes. One of the step to reduce heavy metal is by using bioremediation in order to increase metal stability [7]. Besides, phytoremediation can be used to remove heavy metal by using green plant. There are a few types of phytoremediation such as phytoexraction, phytodegradation, rhizofiltration and phytostabilization [7]. Other than that, deep-well injection also could be a safe method if sites are chosen carefully. Lastly, heavy metal pollution can be reduced by surface impoundments which could store wastes indefinitely with secure double liners. One of the research is to put a certain percentage of heavy metal into fired clay brick. There are many types of heavy metal had been incorporated into fired clay brick. The main problem of heavy metal in the fired clay brick is the immobilization and transformation of heavy metal in fired clay brick. It is because the amount of heavy metal in brick after firing been reduced almost 97% and the content of heavy metal (in ppm) mostly in fired clay brick is acceptable by the USEPA standard (United State Environmental Protection Agency).

In this study, the phase transformation of simulated zinc sludge in aluminium rich ceramic been studied to investigate the formation of new compound between the Al_2O_3 and ZnO that lead to the immobilization of this compound in fired clay brick. Alumina or Al_2O_3 is chosen as the main raw material. Al_2O_3 is known to have the highest amount in brick manufacturing compared to Fe_2O_3 and MgO. When aluminium is coated with inert Al_2O_3, it becomes non-toxic and water soluble. Therefore, it became chemically inert where it unable to react rapidly with additional water molecules or atmospheric oxygen as the structure has changed. It has high heat resistance and corrosion resistance making it more feasible to handle with. Besides, the particle of Al_2O_3 is strongly believed to be efficient in removing heavy metal waste due to their high specific surface area and surface reactivity. Other material used in this study is ZnO. The Zn is chosen because it is one of the most hazardous metals which is not biodegradable, that are commonly found in industrial waste. Zn could accumulate in nature that can caused various diseases and disorders when the specific limits have been exceeded. Various researchers attempt in immobilize metals by adding amendments but failed to recognize the mechanism of occurred reactions and to investigate the transformation of compounds within system. However, a research has been conducted and able to stabilize the heavy metals with well conducted thermal treatment schemes. Thus, the product was proposed to be used as the raw material in the development of ceramic products.

2. Materials and Methods

In order to determine the phase transformation between zinc sludge and aluminium oxide, a series of process were performed. The method involve in this study includes preparation of sample materials, mixing of sample and analysis of sample by using x-ray diffraction machine. The following diagram, Figure 1 shows the summarization of the major experimental procedure in a flow chart type.

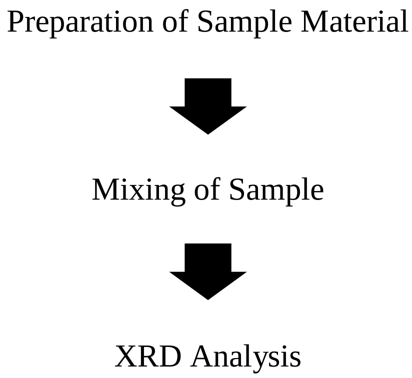


Figure 1: Flow Chart of Experimental Procedure

2.1 Preparation of Sample Materials

Al_2O_3 and ZnO were used in this study as a raw material. Al_2O_3 represent as clay while ZnO act as the heavy metal to be incorporated in the sample. In this study, both raw materials are bought from Saintifik Bersatu Sdn. Bhd. Both samples were in powder form. Al_2O_3 is first weighed and sintered in laboratory furnace at Material Physic Laboratory at 650°C for 3 hours to activate the reactive site of Al_2O_3 to γ- Al_2O_3. Its high specific surface area and high catalytic activity will help in increasing reactive site for ZnO to react with Al_2O_3.

2.2 Mixing of Sample

The Al_2O_3 and ZnO is mixed in conical flask with total weight of 50g. The molar ratios of ZnO and Al_2O_3 were 1:1 and 1:9 with the ratio of ZnO was constant. Each ratio was added with 250ml of deionized water. Then the mixture is placed on a bench top shaker at Food Microbiology Laboratory located at Block J for 18 hours in order for them to perfectly homogenize. After that, the slurry sample is filtered by using filter paper and been dried for 2 days. The sample is then sintered at three different target temperature; 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°C. Then, the samples were ground using an agate mortar and pestle for XRD analysis.

2.3 X-ray Diffraction Analysis

The powder XRD patterns were recorded on a Bruker D8Advance X-ray powder diffractometer equipped with Cu Kα radiation and a LynxEye detector. The diffractometer was operated at 40 kV and 40 mA, and the 2θ scan range was from 10°C to 90°C.

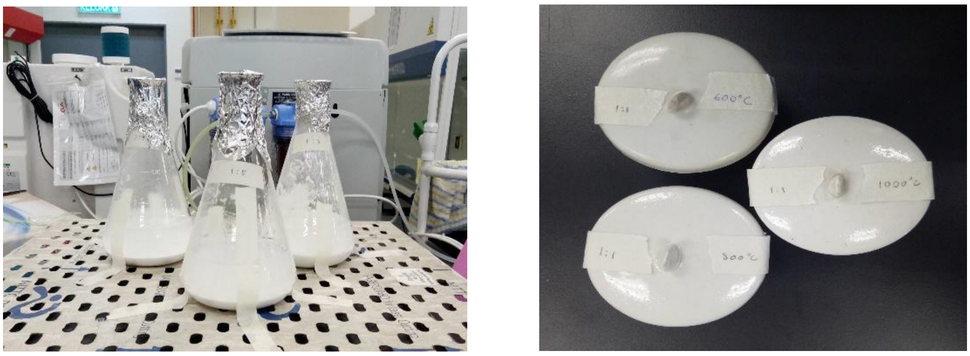


Figure 2: Picture taken during the experimental process

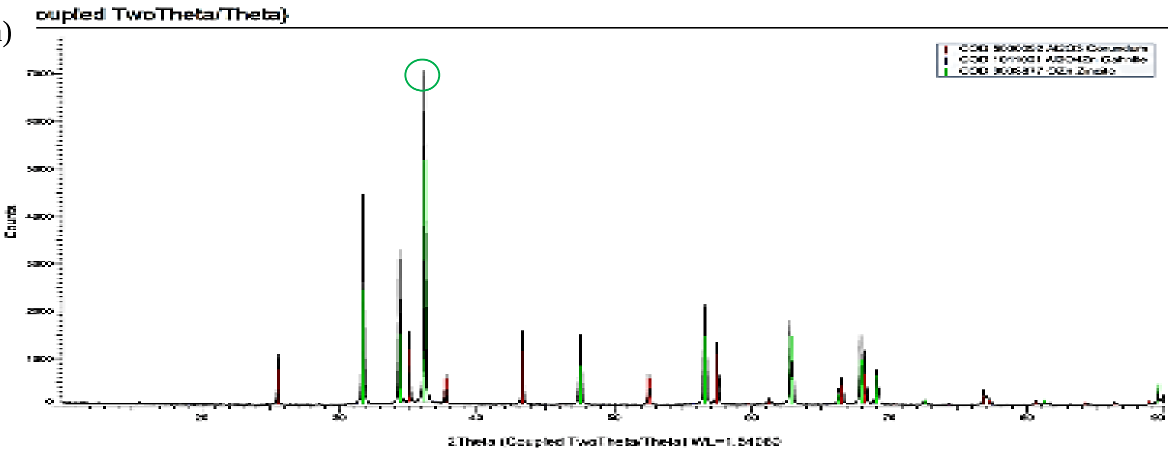
3. Results and Discussion

3.1 Zn transformation at different sintering temperature

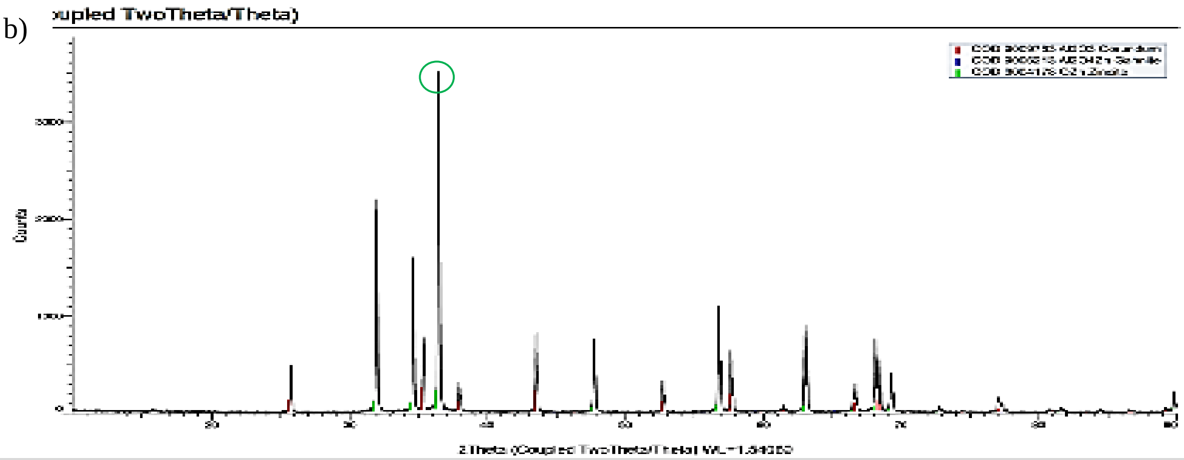
Figure 3 collates the XRD patterns and bar graph of sintered ZnO and Al_2O_3 at three different temperature; 600°C, 800°C and 1000°C at 1:1 ratio. In this study, $ZnAl_2O_4$ is seen to be formed after 3 hour of sintering process. Therefore, the incorporation of ZnO in $ZnAl_2O_4$ with alumina precursor can be describe as follows:

$$Zn_{(s)} + \gamma\text{-}Al_2O_{3(s)} \rightarrow ZnAl_2O_{4(s)} \qquad \text{Eq 1}$$

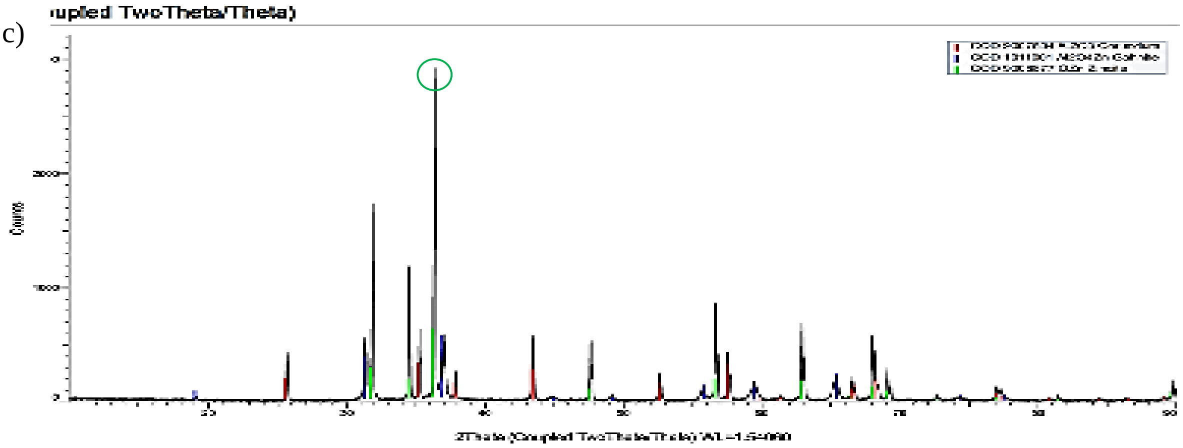
a)



b)



c)



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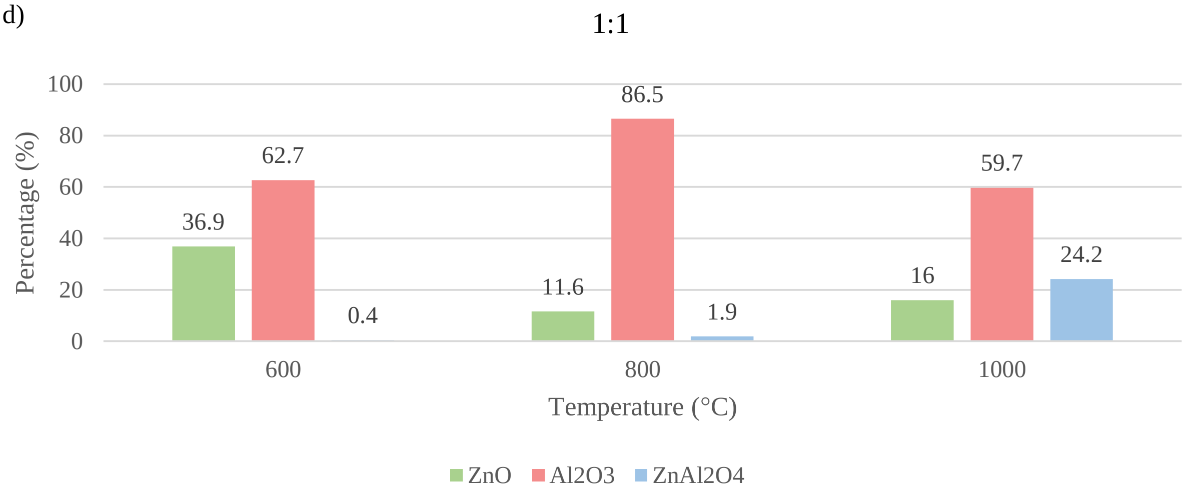

Figure 3: Analysis of reaction between ZnO and Al_2O_3 at 1:1 ratio forming $ZnAl_2O_4$ after 3hr of sintering at different temperature (a) 600°C, (b) 800°C (c) 1000°C and (d) bar chart of percentage versus temperature.

To explore the influence of Zn/Al ratio on the incorporation of zinc, the mixture of ZnO and γ- Al_2O_3 with correlative ratio; 1:1 were sintered at three different temperature, 600°C, 800°C and 1000°C for 3hr. As shown in Figure 3, Al_2O_3 and ZnO tends to have fluctuate amount on all three temperature. At temperature 600°C, the percentage of Al2O3 is 62.7%. The number is increasing at temperature 800°C which is 86.5% while at the highest temperature, 1000°C, the number of Al_2O_3 back to the lowest percentage which is 59.7%. The highest amount of ZnO that is still present in the experiment is 36.9% at temperature 600°C while the lowest amount can be seen at temperature 800°C which is 11.6%. At temperature 1000°C, the percentage of ZnO is 16%. Only small amount of $ZnAl_2O_4$ is formed at low temperature, 600°C which is 0.4%. The formation of $ZnAl_2O_4$ showed a slight increase at temperature 800°C which is 1.9% and it is continue to raised up dramatically to 24.2% at 1000°C.

The increasing percentage of Al_2O_3 from 600°C to 800°C is because Al_2O_3 started to react with ZnO forming $ZnAl_2O_4$ resulting in the decreasing amount of ZnO from 36.9% to 11.6%. As the Al_2O_3 start to stable, the percentage is decreasing from 86.5% to 59.7% when it reach 1000°C. At this temperature, Al_2O_3 started to react effectively with ZnO as it is been used more compared to 800°C temperature and forming higher amount of $ZnAl_2O_4$. The increasing amount of product is due to the effect from high sintering temperature inducing in increasing of kinetic energy for the reaction between both reactant to happen. So when the reactant is heated, some of the absorbed energy is stored within the particles while some of the energy increases the motion of the particles. Hence, when the heat increase, the kinetic energy increase as well. Increasing in kinetic energy leading to higher amount of product formation.

3.2 Zn transformation at different Al/Zn ratio

Meanwhile, Figure 4 below shows the results of XRD patterns and bar graph of sintered ZnO and Al_2O_3 at different ratio; 1:1 and 1:9 with a temperature of 1000^0C.

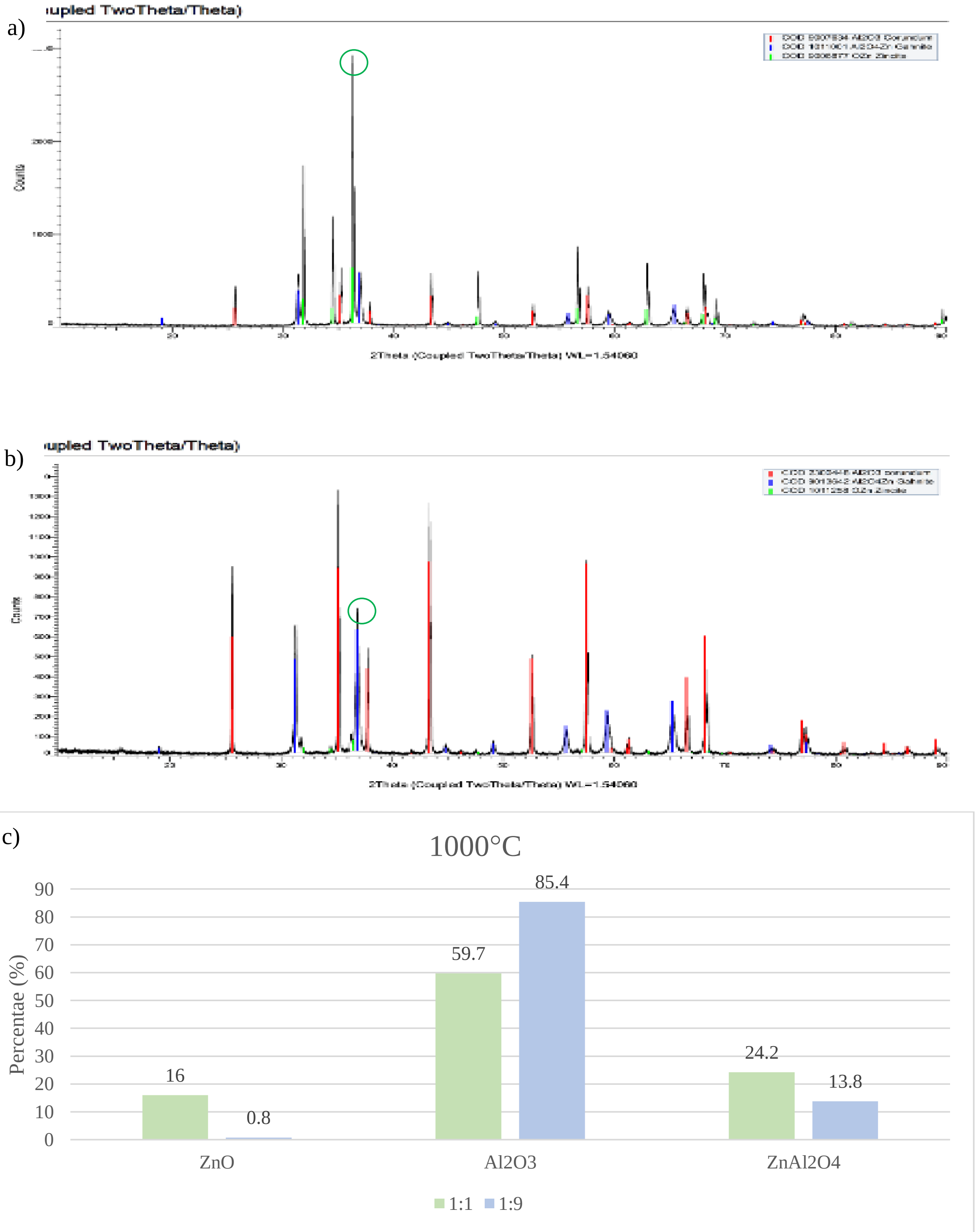

Figure 4: Analysis of reaction between ZnO and Al_2O_3 at different ratio (a) 1:1 and (b) 1:9 and (c) bar chart of percentage versus ratio forming $ZnAl_2O_4$ after 3hr of 1000°C sintering temperature.

Besides temperature, the molar ratio also plays an important role for the reaction to occur effectively. Result showed that, if ZnO and Al_2O_3 is sintered at 1:1 ratio, the amount of $ZnAl_2O_4$ obtained is higher which is 24.2% compared to 1:9 ratio, only 13.8%. At 1:1 ratio, the amount of Al_2O_3 is 59.7%, ZnO is 16% and $ZnAl_2O_4$ is 24.2%. For 1:9 ratio the amount of Al_2O_3 increased up to 85.4% that showing 30.7% differences between the 1:1 and 1:9 ratios. The number of ZnO and $ZnAl_2O_4$ is decreasing respectively in 1:9 ratio. For ZnO the amount is only 0.8% while the $ZnAl_2O_4$ is 13.8%. The differences between the ZnO and $ZnAl_2O_4$ is 15.2% and 10.4%.

The limitation in one of the reactant amount leading to the stops of the reaction abruptly in 1:9 ratio. In this study, ZnO plays an important role for the reaction to happen as it is the limiting reactant. The lower the ratio of the reactant, the lower the product will be formed. Limiting reactant is important as it will limit the amount of $ZnAl_2O_4$ can be formed. The reaction between ZnO and Al_2O_3 will eventually stops when ZnO is fully consumed. Thus, affecting the amount of $ZnAl_2O_4$ formed.

4. Conclusion

In conclusion, both experiment showed that, the formation of $ZnAl_2O_4$ only appear when it is exposed at high temperature and high ratio. The technological advancement has greatly improved and ease human life at the expense of hazardous material that is vulnerable and risky for human life. In addition, 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. From XRD data, it shows that it has been immobilize. Thus, it is not harmful to environment and humankind to be used as building material.

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