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Characterization of iron ore with Al_2O_3 as oxygen carrier in Chemical Looping Combustion (CLC)

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Abstract. Chemical looping combustion (CLC) is one of the carbon capturing technologies, in which oxygen from oxygen carrier reacts with fuel inside fuel reactor to produce CO_2 and H_2O. Fe-based oxygen carrier is widely used in CLC due to the low cost and less susceptible to carbon formation. This research focuses on synthesizing and characterizing iron ore with alumina in order to analyse the suitability of Malaysia iron ore as an oxygen carrier for CLC application. Iron ore with alumina was prepared using ball milling at various milling time which are 1 hour, 6 hours, 8 hours and 10 hours. The phase transformation, morphology and elemental composition of obtained samples were characterized using X-Ray Diffraction (XRD), scanning electron microscopy (SEM), and Energy Dispersive X-Ray (XRD), respectively. In CLC application, high reactivity of CLC can be obtained with the small particle size of oxygen carrier. This research succeeded in producing Fe_2O_3/ Al_2O_3 using ball milling with particle size that less than 10µm with crystallite size 17nm at 10 hours, which favourable to be used as oxygen carrier.

1. Introduction

Greenhouse gases that released to the environment cause the global warming and subsequent rise in atmospheric temperature. Many methods were used to control the emissions of carbon dioxide (CO_2) such as increase the efficiency of energy conversion, to use renewable energy and to capture the emissions of CO_2. However, capturing and storing the CO_2 (CCS) is the best method to reduce CO_2 emissions since combustion of fossil fuels will release the CO_2 into the air [1].
Chemical looping combustion was introduced as one of the carbon capture Storage (CCS) method that required low energy consumption and capable to increase efficiency of power plant comparing to conventional method [2]. CLC consists of two reactors, which are fuel reactor and air reactor. In CLC application, oxygen carriers play a very significant role in the overall process since oxygen carrier provides an oxygen for the combustion process. The oxygen carrier will react with the fuel in the fuel reactor in order to produce pure CO_2 and H_2O as a product. Then, this pure CO_2 can be stored in a liquid form instead release to the air. The reduced oxygen carrier will be flow into air reactor to oxidise with the air prior flow into fuel reactor again. Therefore, efficiency of CLC greatly depends on the performance of oxygen carriers. A good oxygen carrier should have good fluidization properties, high oxygen carrying capacity, high melting points, low cost, and environmental friendly [3].
The common metal based that was used as oxygen carriers are Fe-based, Cu-based, Ni-based, Mn-based and Co-based. From these five metals based, Fe-based, Cu-based and Mn-based are widely been applied in CLC system [4]. However, Cu-based has low melting point, meanwhile Ni-based shows a

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thermodynamic limitation, costly, and needed special safety measure to handle this material due to its hazardousness [5]. Fe-based is low cost, less susceptible to carbon formation, non hazardous, so the total operational cost would be low in CLC. However, particle agglomeration becomes major concern after several redox reactions when using Fe-based oxygen carriers [6]. Therefore, supported materials such as Al_2O_3, $MgAl_2O_4$, SiO_2, TiO_2 and YSZ were used with Fe-based in order to increase the durability of Fe-based oxygen carriers [7]. Iron ore was used continuously as Fe-based oxygen carrier in CLC since it shows good fluidization properties and minimum attrition [8].

Many methods were used to synthesize oxygen carrier such as mechanical mixing, dissolution method, freeze granulation method and sol-gel method [9-11]. However, mechanical mixing is a facile method that can produce small particle size of oxygen carrier. Song et al. [12] stated that, oxygen carrier with smaller particle size are more stable at high temperature and have superior mechanical properties. Hence, it can increase the efficiency of CLC.

Therefore, this research focuses on synthesizing and characterizing iron ore with Al_2O_3 using ball milling method and investigates the suitability of obtained iron ore from iron mining site in Malaysia as an oxygen carrier. In this research various ball milling time were varied in order to determine formation of Fe_2O_3 with Al_2O_3 since no extensive studies were found on synthesizing Malaysia iron ore with Al_2O_3 as an oxygen carrier for CLC application.

2. Methodology

In this research, ball milling method was used to produce Fe_2O_3/Al_2O_3 powder. Iron ore that found in Malaysia was heated up at temperature 950 ℃ prior to the milling process. This process is to maximise the oxygen content in the powder that capable to carry more oxygen in CLC application[8]. The mixture of iron ore and alumina are inserted into the milling jar and milled with the speed of 300 rpm at various milled time which are 1 hour, 6 hours, 8 hours and 10 hours. Then, the obtained powder was characterized. The morphology and elemental composition of obtained samples were characterized using scanning electron microscopy (SEM), and Energy Dispersive X-Ray (XRD), respectively.

The crystalline phase of Fe_2O_3/Al_2O_3 powder was characterized using X-Ray diffraction (SHIMADZU 6000) at a scan speed 3°/min using CuKα radiation. The crystallite size was calculated by using Scherer equation. The Scherer equation is given below [13],

$$D = z\frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

where D is crystallite size (in nm), λ is the radiation wavelength (for CuKα radiation, λ 1.5418Å), θ is the diffraction peak angle and β is the broadening of the line (“half width”) measured at half its maximum intensity (in radians). The morphology and elements of the Fe_2O_3/Al_2O_3 powder were observed using Scanning Electron Microscopy, SEM and Energy dispersive X-Ray, EDX respectively (JEOL).

3. Powder characterizations

3.1. Phase of obtained powder

The phase of obtained powder was analyzed using XRD analysis. Figure 1 shows XRD results of Fe_2O_3/Al_2O_3 when milled at 1 hour, 6 hours, 8 hours and 10 hours. The presence of Fe_2O_3 and Al_2O_3 can be observed in figure 1 corresponding to JCPDS no 39-1346 and JCPDS no 01-070-3319, respectively. However, the XRD reflection intensity decreases with increasing of milling time. The reduction of this intensity indicates the formation of amorphous structure that caused by the plastic deformation and disintegration of Fe_2O_3 and Al_2O_3. The XRD patterns shows only Fe_2O_3 and Al_2O_3 reflections, indicating that both compositions did not undergo significant reaction and physico-chemical changes [14]. Phourghahramani. P et al. stated that the X-Ray amorphization increases with increasing of milling time even with the particle size reduction and high BET surface area [14].

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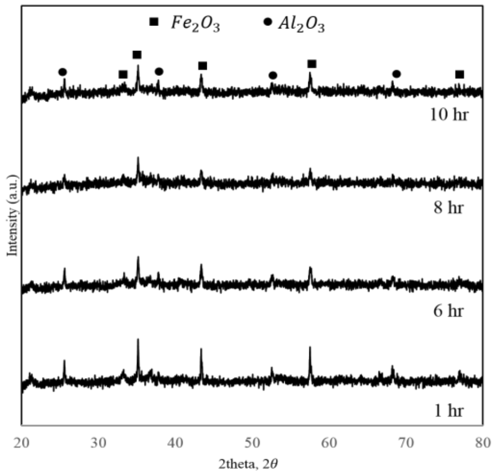

Figure 1. XRD result of Fe2O3 /Al2O3 when milling at 1 hour, 6 hours, 8 hours and 10 hours.

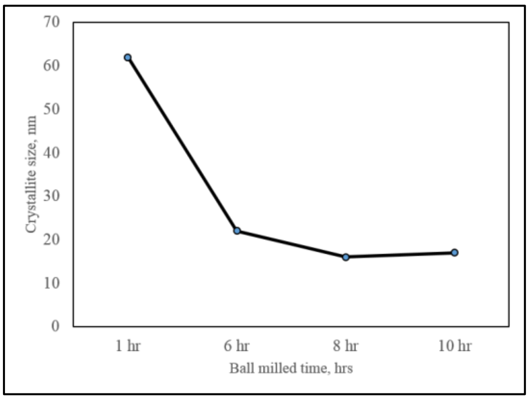

Figure 2. Average crystallite size of Fe2O3 /Al2O3 when milling at 1 hour, 6 hours, 8 hours and 10 hours.

3.2. Crystallite size
Figure 2 shows the average crystallite size of Fe_2O_3/ Al_2O_3 powder when milling at various milling time. The average crystallite size starts to decrease from 62 nm to 17 nm when milling time increases from 1 hour to 10 hours. The decrease in crystallite size is resulted from broken crystals even at the nanoscales due to the impact and abrasion forces generated inside milling jar [14].

3.3. Morphology of the Fe_2O_3/Al_2O_3
Figure 3 shows microstructure of Fe_2O_3/ Al_2O_3 when milled at 1, 6, 8 and 10 hours. It can be observed that particle size at 1 hour exhibit the aggregation of large irregular particle. Particle size start to decrease when milling time was increased from 1hour to 10hours. This is due to the effect of impact force and shear friction that generate cracks continuously and decrease the particle into small particle size [15]. However, no observable effect on the particle morphology when increases the milling time from 1hour to 10hours. Therefore, increases the milling time capable to produce Fe_2O_3 and Al_2O_3 at small particle size, hence high BET surface area. Song et al. [12] stated that, smaller particle of oxygen carriers are more stable at high temperature, and have superior mechanical properties which can increase CLC performance.

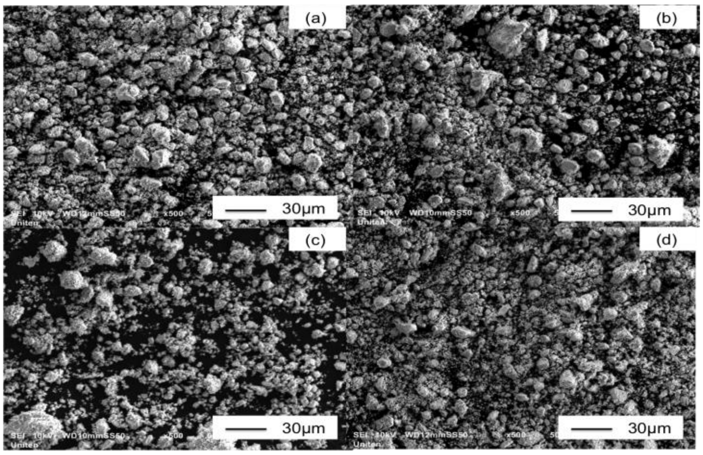

Figure 3. SEM images of Fe_2O_3 /Al_2O_3 when milling at different time a) 1hr b) 6hrs c) 8hrs d) 10hrs.

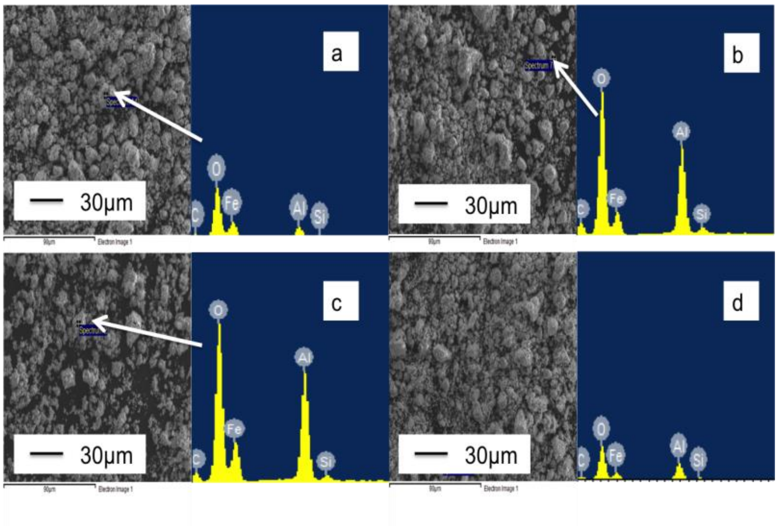

Figure 4. Elemental composition of Fe_2O_3 /Al_2O_3 when milling at different time a) 1hr b) 6hrs c) 8hrs d) 10hrs.

3.4. Elements of the Fe_2O_3/Al_2O_3

Figure 4 shows the elemental composition of Fe_2O_3/ Al_2O_3 when milling at different time. Increases milling time does not affect the elemental composition of the powder. The presence of Fe, Al and O can be seen at all these milling times. In addition, the presence of Si and C can be found in these powders which is in agreement with Wong et al. [16]. Si would not affect the performance of CLC [17]. Therefore, it can be negligible. Presence of C was due to the contamination from milling process and carbon tape during sample preparation prior analysis. Hence, this Fe_2O_3 powder is fit to be tested in thermogravimetric analysis (TGA) for simulating the CLC application.

4. Conclusions

According to the obtained result, Fe_2O_3 and Al_2O_3 was successfully synthesized using iron ore with alumina by ball milling mixing method. Fe_2O_3/Al_2O_3 that was milled at 10hrs exhibit smaller particle size which is less than 10μm with crystallite size 17nm compare to the other milling time. Smaller particle size can increase CLC performance, hence produced oxygen carrier that can be used in CLC application. Furthermore, this Fe_2O_3 with Al_2O_3 can be tested using TGA analysis in order to simulate the CLC process for its performance.

5. Acknowledgments
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6. References
[1] Yamasaki A 2003 An overview of CO_2 mitigation options for global warming- emphasizing CO_2 sequestration options *J. Chem Eng of Japan* **36** 361-375
[2] Shijaz H, Attada Y, Patnaikuni VS, Vooradi R, Sarath BA 2017 Analysis of integrated gasification combined cycle power plant incorporating chemical looping combustion for environmental friendly utilization of Indian coal *Eneryg Converse Manag* **151** 414-425
[3] Adánez J, Abad A, Garc a-Labiano F, Gay án P, de Diego LF 2012 Progress in Chemical looping combustion and reforming technologies. A review *Prog Energy Combust Sci* **38** 215-282
[4] Wang B, Yan R, Lee DH, Zheng Y, Zhao H, Zheng C 2011 Characterization and evaluation of Fe_2O_3/Al_2O_3 oxygen carrier prepared by sol-gel combustion synthesis *J Anal Appl Pyrolysis* **91** 105-113
[5] Tang M, Xu L, Fan M 2015 Progress in oxygen carrier development of methane based chemical looping reforming. A review *Applied energy* **151** 143-156
[6] Nandy A, Loha C, Sai G, Sarkar P, Karmakar K 2016 Present status and overview of Chemical Looping Combustion technology *Renew Sust Energ Rev* **59** 597-619
[7] Gao ZF, Wu ZJ, Liu WM 2016 Preparation and chemical looping combustion properties of Fe_2O_3/ Al_2O_3 derived from metallurgy iron-bearing dust. *J Environ Chem Eng* **4** 1653-1663
[8] Haider SK, Azimi G, Duan L, Anthony EJ, Patchigolla, Oakey JE, Leion H, Mattisson T, Lyngfelt A 2016 Enhancing properties of iron ore and manganese ores as oxygen carriers for chemical looping combustion *Appl Energ* **163** 41-50
[9] Zhao HB et al. 2008 $NiO/NiAl_2O_4$ oxygen carrier prepared by sol-gel for chemical looping combustion fuelled by gas *J Fuel Chem Tech* **36** 261-266
[10] Ishida M, Yamamoto M and Obata T 2002 Experimental results of chemical looping combustion with $NiO/NiAl_2O_4$ particle simulation at 1200°C *Energ Converse Manag* **43** 1469-1478
[11] Cho P, Mattinson T, Lyngfelt 2004 Comparison of iron-, nickel-, copper- and manganese based oxygen carriers for chemical looping combustion *Fuel* **83** 1215-1225
[12] Song Q, Xiao R, Deng Z, Zhang H, Shen L, Xiao J 2008 Chemical looping combustion of methane with $CaSO_4$ oxygen carrier in a fixed bed reactor *Energ Converse Manag* **49** 3178-3187
[13] Parra MR and Haque FZ 2014 Aqueous chemical route synthesis and the effect of calcination temperature on the structural and optical properties of ZnO nanoparticles *Journal of Materials Research and Technology* **3** 363-369.
[14] Pourghahramani P, Forssberg E 2006 Comparative study of microstructural characteristic and stored energy of mechanically activated hematite in different grinding environments *Int. J. Miner. Process* **79** 130-139
[15] Wang B, Wei S, Wang Y, Guo L, Xue J, Pan, F, Tang A, Xu B 2018 Effect of milling time on microstructure and properties of Nano-titanium polymer by high energy ball milling *Appl Surf Sci* **434** 1248-1256
[16] Wong YW, Woon HS, Tan CY 2014 Purification and conversion of Malaysian iron ores into industrial grade iron oxide colour pigment *Mater Res Innov* **18** 159-163, 2014.
[17] Linderholm C, Schmitz S, 2016 Chemical-looping combustion of solid fuels in a 100kW dual circulating fluidized bed system using iron ore as oxygen carrier *J Environ Chem Eng* **4** 1029-1039