

Performance of nickel-based oxygen carrier produced using renewable fuel aloe vera

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Abstract. Consuming and burning of fuel mainly fossil fuel has gradually increased in this upcoming era due to high-energy demand and causes the global warming. One of the most effective ways to reduce the greenhouse gases is by capturing carbon dioxide (CO₂) during the combustion process. Chemical looping combustion (CLC) is one of the most effective methods to capture the CO₂ without the need of an energy intensive air separation unit. This method uses oxygen carrier to provide O₂ that can react with fuel to form CO₂ and H₂O. This research focuses on synthesizing NiO/NiAl₂O₄ as an oxygen carrier due to its properties that can withstand high temperature during CLC application. The NiO/NiAl₂O₄ powder was synthesized using solution combustion method with plant extract renewable fuel, aloe vera as the fuel. In order to optimize the performance of the particles that can be used in CLC application, various calcination temperatures were varied at 600°C, 800°C, 1050°C and 1300°C. The phase and morphology of obtained powders were characterized using X-ray diffraction (XRD) and Field Emission Microscopy (FESEM) respectively together with the powder elements. In CLC application, high reactivity can be achieved by using smaller particle size of oxygen carrier. This research succeeded in producing nano-structured powder with high crystalline structure at temperature 1050°C which is suitable to be used in CLC application.

1. Introduction

Green house gases (GHGs) are rapidly discharged to the environment throughout these years due to high-energy demands from the industries. The release of GHGs can cause greenhouse effects that bring harm towards the environment. Therefore, many methods were introduced to reduce the emissions of GHGs such as to capture the emissions of CO₂, to increase the efficiency of energy conversion and usage and also to use renewable fuel and nuclear energy [1]. However, capturing and storing CO₂ had gained more interest among researchers due to the inevitable emissions of CO₂ during combustion process [2].

Chemical looping combustion (CLC) was introduced as one of the methods that can capture the CO₂ without requiring high-energy consumptions [3]. In CLC application, oxygen carrier provides oxygen that reacts with fuel to produce CO₂ and H₂O during combustion process. This oxygen carrier will undergo reduction and oxidation process inside the reactors. The regenerability of oxygen carriers is important since it determines the durability of the oxygen carrier [4]. Therefore, efficiency of CLC application highly depends on the performance of oxygen carriers. The oxygen carrier consists of



metal oxide and binder material, which provides oxygen and increase the mechanical properties of oxygen carrier respectively [5]. One of the oxygen carriers that can perform good chemical reactivity in CLC application is the NiO/NiAl₂O₄ combination [6]. The NiO/NiAl₂O₄ had been attracting many researchers due to its properties that can withstand high temperature. The operating temperature of CLC application can be up to 1200°C [7].

Preparation method of oxygen carriers plays an important role in CLC application since it determines the performance of oxygen carriers. Ishida et al. [8] prepared NiO/NiAl₂O₄ using a dissolution method and obtained a spherical particle with diameter that is larger than 97 µm. Zhao et al. [9] produce NiO/NiAl₂O₄ powder using sol-gel method and obtained particles with sizes ranging from 125-180 µm. However, both methods consume a lot of time (30 – 40 hours). Cho et al. [10] synthesized NiO/NiAl₂O₄ using a freeze-granulation method, which reduced the preparation time, but the resulting particles had low mechanism strengths. Rate of redox reaction in CLC application was found to increase when smaller particle size was used due to the decreases in diffusional resistance [8]. Moreover, nanostructured oxygen carrier can improve the CLC performance since it can reduce the formation of sulfur content [11].

Therefore, this research focuses on synthesizing NiO/NiAl₂O₄ using solution combustion method with aloe vera as fuel. Solution combustion method is capable to produce nanostructured powder with high purity powder using simple tooling requirements [12]. The usage of aloe vera as fuel is more environmental friendly and capable to produce NiO/NiAl₂O₄ powder at low cost and releases non-toxic gases [13]. In this research, various calcination temperatures were varied in order to determine the formation of NiO/NiAl₂O₄ since very few extensive studies were found on synthesizing this powder using aloe vera as fuel. Then, the powder was characterized using X-Ray Diffraction, Field Emission Microscopy and Energy Dispersive X-ray to determine the crystalline phase, morphology and elements of the obtained powders.

2. Methodology

In this research, solution combustion method was used to synthesize NiO/NiAl₂O₄ powder using aloe vera as fuel. Aloe vera is an environmental friendly source that can act as fuel that produces non-toxic gases during combustion process.

Firstly, few leaves of aloe vera was taken from aloe vera plant and washed gently to remove any impurities and dirt. Then, the top layer of aloe vera leaves were cut in order to obtain the aloe vera gel (5g). After that, this aloe vera gel was placed inside beaker together with 10ml distilled water and stirred for 30 minutes in a room temperature prior to obtain the clear solution of aloe vera. This entire process was known as renewable fuel extraction process.

Then, aluminum nitrate (Al(NO₃)₃·9H₂O) and nickel nitrate (Ni(NO₃)₂·6H₂O) were mixed together with the aloe vera solution prior the heating process. The mixture was heated on top of the hot plate until the combustion process occurred. This mixture was stirred continuously throughout this heating process. The solution was found to be combust at 300°C about 40 minutes. This heating process was conducted in ambient air. The as-synthesized powders that were obtained after combustion process were collected and grinded by using mortar in order to get fine particle powder prior the calcination process. Then this as-synthesized powders were introduced inside muffle furnace at various calcination temperatures which were 600°C, 800°C, 1050°C and 1300°C.

The crystalline phase of NiO/NiAl₂O₄ 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 [14],

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

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 NiO/NiAl₂O₄ powder were observed using Field Emission Microscopy, FESEM and Energy dispersive X-Ray respectively (Hitachi SU8030).

3. Powder characterizations

3.1. Phase of obtained powder (X-Ray diffraction, XRD)

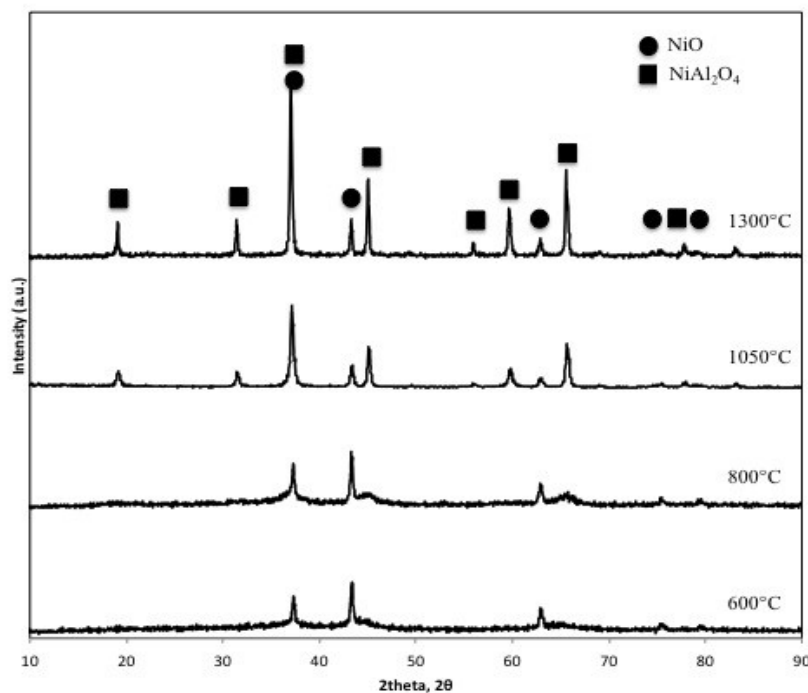


Figure 1. XRD results when NiO/NiAl₂O₄ powder calcined at temperature 600°C, 800°C, 1050°C and 1300°C.

Figure 1 shows XRD results of NiO/NiAl₂O₄ when calcined at 600°C, 800°C, 1050°C and 1300°C. This figure indicates that only NiO phase was shown at 600°C which corresponds to FCC structure [15]. It is in agreement with Ganachari et al. [15] that the formation of NiO is above than 500°C. Calcination at 800°C produces two more peaks which correspond to NiAl₂O₄ peaks (JCPDS 00-010-0339). The formation of NiAl₂O₄ was reported to occur at temperature above than 725°C due to the spinel is non-tolerant structure concerning an excess of Ni [16]. Then, more NiAl₂O₄ peaks were identified at 1050°C and 1300°C. This result suggested that Al³⁺ ions, Ni²⁺ ions and oxygen had been oxidized to form NiO and NiAl₂O₄ phase [17]. However calcination at 1300°C produces sharper peaks than 1050°C which corresponds to a better crystalline phase due to enough heat was introduced at high temperature [18]. In addition, formation of Al₂O₃ was not found at 1300°C even though it is been suggested that Al₂O₃ occurs at temperature above than 1100°C [19]. Formation of Al₂O₃ together with NiO and NiAl₂O₄ is not favorable in CLC application since it can degrade the CLC performance [20]. Therefore, NiO and NiAl₂O₄ powders were formed at temperature above than 800°C without presence of Al₂O₃.

3.2. Crystallite size

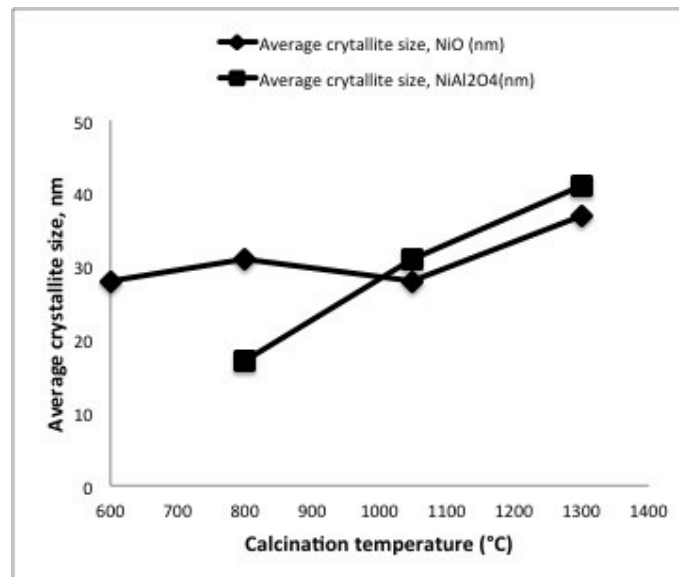


Figure 2. Average crystallite size of the NiO and NiAl₂O₄ powder when calcined at temperature 600°C, 800°C, 1050°C and 1300°C.

Figure 2 shows the NiO/NiAl₂O₄ average crystallite size as a function of calcination temperature. The crystallite size of NiAl₂O₄ increased from 17 nm to 41 nm, respectively when temperature increase from 600°C to 1300°C. The increase in crystallite size resulted from heat presented when increases the temperature that can produce larger crystallite size [18]. However, the crystallite size of NiO starts to decrease from 31 nm to 28 nm from 800°C to 1050°C. This result suggests that there is a break up in the grain size and causes it to be decreased [21]. However, crystallite size of NiO increases from 28 nm to 31 nm from 600°C to 800°C and 31 nm to 37 nm from 1050°C to 1300°C which in agreement with Leal et al. [18] that increase in calcination temperature causes increases in crystallite size.

3.3. Morphology of the NiO/NiAl₂O₄

Figure 3 shows FESEM images of the NiO/NiAl₂O₄ powders when calcined at 600°C, 800°C, 1050°C and 1300°C. The particles powder calcined at 600°C displays irregular shape with agglomeration and sintered necks with adjacent grain particle due to the large amount of heat occurred during combustion process [22]. At temperature 800°C, the particles started to interconnect with other grain particles to become one large grain size. However, this formation is weak, fragile and could break easily when more heat was introduced since there are still agglomerated particle [22]. However, the particles started to break up to become smaller particle size due to the grain refinement at temperature 1050°C. High crystalline structure started to form at 1300°C. This formation was due to the enough heat to stimulate the formation of crystalline structure [23]. This result also can be seen in figure 1 that sharper peaks were found at 1300°C than other calcination temperature. However, smaller particle size can be obtained at 1050°C, which favorable in CLC application since it could increase CLC performance.

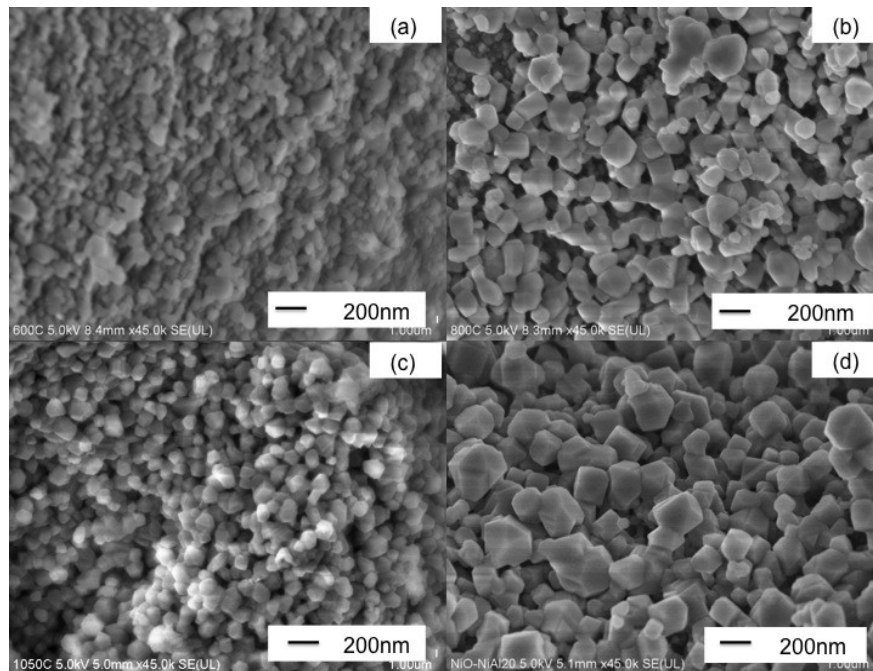
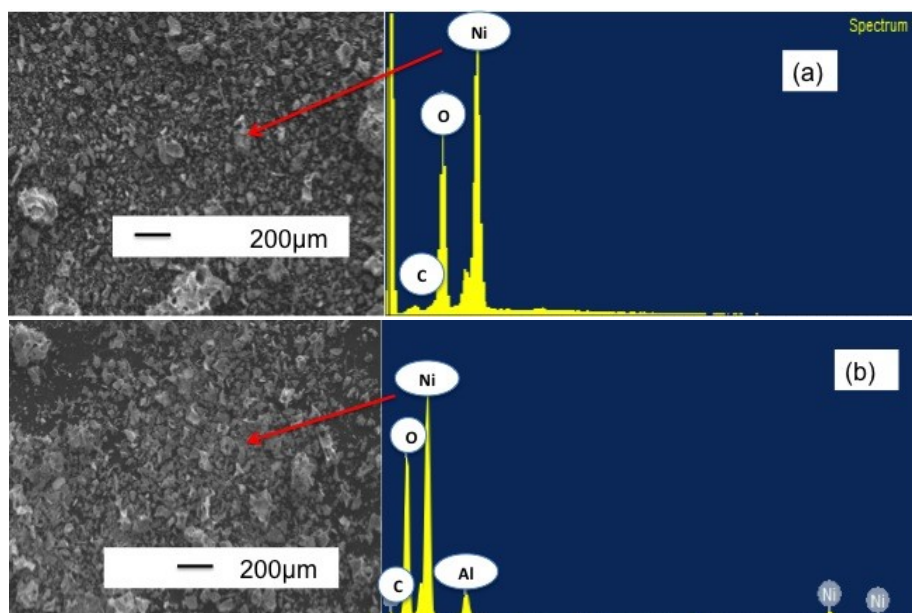


Figure 3. FESEM images of NiO/NiAl₃O₄ powder when calcined at temperature (a) 600°C, (b) 800°C, (c) 1050°C (d) 1300°C.

3.4. Elements of the NiO/NiAl₂O₄

Figure 4 illustrates elemental analysis of obtained powder using aloe vera as fuel at various calcination temperatures. Only Ni and O elements were shown in figure 4(a). The nuclei of Al₂O₃ does not nucleate yet due to not enough heat presented during synthesis process when calcined at 600°C [24]. Moreover, Al elements start to develop at temperature above than 800°C which can be seen in figure 1. Figure 1 shows that NiAl₂O₄ starts to form at 800°C. However, C element also presented in this research, which is not favorable in CLC application due to the carbon inhibit densification during calcination process [25]. However, amount of carbon content can consider as less and this powder would perform comparatively well in CLC application [25].



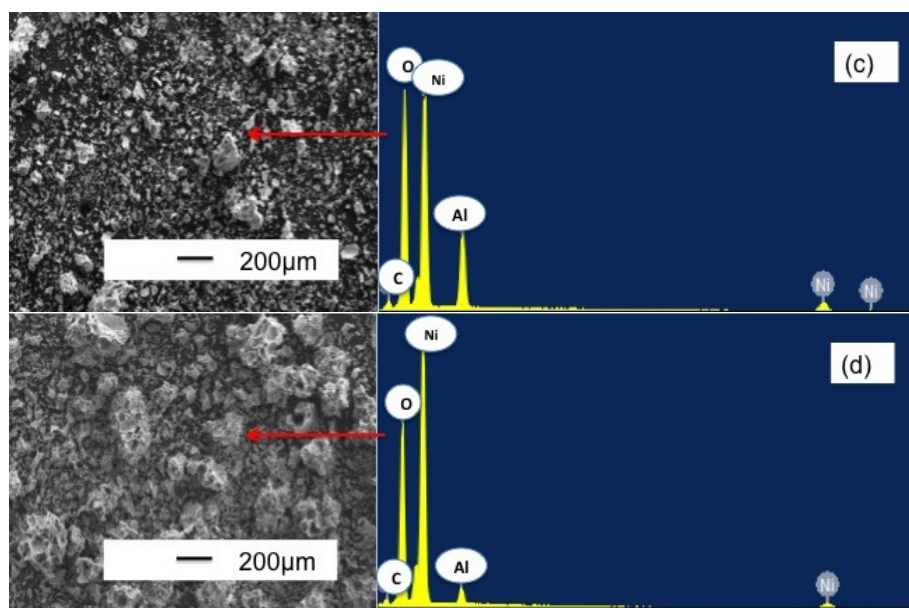


Figure 4. Elemental analysis of NiO/NiAl₃O₄ powder when calcined at temperature (a) 600°C, (b) 800°C, (c) 1050°C (d) 1300°C.

4. Conclusions

Effect of the calcination temperature on the NiO/NiAl₂O₄ powder synthesized using solution combustion method was studied thoroughly. According to the obtained result, NiO/NiAl₂O₄ was successfully synthesized using solution combustion method using aloe vera as fuel at temperature 1050°C, which exhibit smaller size comparing to other calcination temperature and produced oxygen carrier that can be used in CLC application.

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