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Research Article

The effect of Niobium oxide as a promoter on □-Al2O3 based oxygen carrier for chemical looping combustion

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Abstract: Chemical looping combustion (CLC) is known as a low-cost strategy for the capture of carbon dioxide for fuel combustion. In CLC process, oxygen carriers (OCs) are the cornerstone and play a vital role in defining reaction process. The aim of the present work was to investigate the potential of niobium (Nb) as a promoter on the □-Al2O3 and its behavior as an oxygen carrier for chemical looping combustion process. In this work, the Nb loading was varied between 5 – 15 wt. %. Their behavior in CLC was analyzed by TPR, TPO, SEM and TGA. From the TPR results, the reduction temperature for 5 wt.%, 10 wt.% and 15 wt.% Nb loading were 560 ℃, 529 ℃ and 545 ℃, respectively which indicated that reduction reaction occurs around 500 ℃ and above. SEM analysis showed that increasing of Nb loading resulted in some agglomeration and thus lowering the ability of metal oxide to gain and release oxygen. The redox characteristics were carried out using TGA with 5% CH4/N2 was used as the reducing gas, while air was used as oxidizing gas. The highest oxygen transfer capacity was 3.0% which is presented by 5 wt.% of Nb loading. Since the addition of Nb successfully improved the oxygen transport capacity, it can be concluded that Nb is the potential candidate for oxygen carrier in CLC.

Keywords: chemical looping combustion; oxygen carrier; metal oxide

1. Introduction

Climate change caused by greenhouse gases, and it can have a detrimental effect on its structure when gases such as carbon dioxide (CO_2), nitrous oxide (NO_x), methane (CH_4), etc. are in abundance in the ozone. With increasing global energy consumption, growing of greenhouse gas emissions are a major factor contributing to global warming [1]. While several attempts are continuing to establish green energy sources, it is expected that fossil fuels will continue to be the primary source of global energy over the next few centuries [1, 2]. The vast majority of greenhouse gas emissions from the industry is carbon dioxide (CO_2). It is primarily due to the burning of fossil fuels during power generation and causes an effect on the temperature of the Earth. One main aim of seeking to minimize CO_2 pollution is to reduce CO_2 emissions from main source points [3].

Carbon capture and storage (CCS) is one of the key strategies for mitigating CO_2 pollution from large stationary sources, such as coal plants [4]. Specifically, CCS is a method where CO_2 is segregated from power generation, shipped under high pressure to the storage site and subsequently stored in the deep ground in a way to reach zero carbon emissions in using fossil energy [1]. Various alternative are available for eliminating CO_2 such as pre-combustion, whereby the fuel is decarbonized prior to combustion, combustion of oxy-fuel using pure oxygen extracted from cryogenic nitrogen atmospheric separation and post-combustion, which requires different procedures to separate CO_2 from flue gases [3]. Chemical looping combustion (CLC) is a great carbon sequestration practice where it uses metal oxides, also defined as the oxygen carrier (OC), rather than air to supply oxygen for the combustion process [5]. As illustrated in Fig. 1, CLC is a cyclical mechanism that utilizes two distinct reactors: a fuel reactor and an air reactor. In the fuel reactor, the fuel in the gaseous process is oxidized by a chemical reaction using an oxygen carrier. While in the second reactor, the oxygen carrier is reoxidized to absorb the oxygen from air.

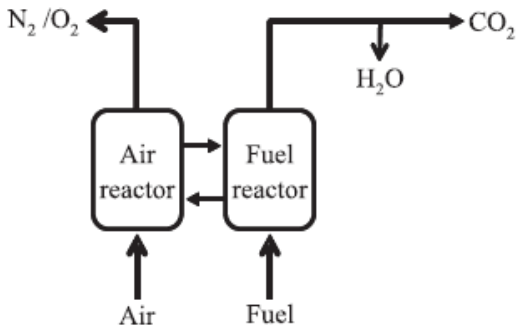


Fig. 1. Schematic diagram of the CLC process [6]

In CLC process, OC is constantly recirculated between two reactors, thus minimizing direct interaction between fuel and air reactor. The key drawback of CLC is that CO_2 emissions to the atmosphere are avoided by the natural isolation of CO_2 and water vapor from the remaining flue gases. In order to be well matched to the CLC process, a good OC material must achieve a range of significant requirements such as enough ability to carry oxygen with high oxygen capacity, stable over periods of repeated reduction and oxidation, high reactivity for redox reaction, economically sustainable and environmentally friendly [7, 8]. Synthetic OCs typically consist of single metal oxides (first-row transition metal oxides are commonly used such as Ni, Cu, Co, Cd, Mn and Fe [6, 9-11]. Even so, for the CLC test, preliminary findings are available for just a several of these metals [12]. While CLC showed great potential for these metal oxides, their efficiency and reactivity would decline after many cycles.

Combining other metals will improve the performance, stability and reactivity of OCs [13]. According to the literature, niobium oxide can acts as active metal, support, solid acid catalyst, promoter as well as redox compound [14]. Papulovskiy et al. [15] studied the effect of niobium oxide on alumina using impregnation method. They found that with addition of 16 wt.% of Niobium oxide, results favored the formation of high-coordinate Nb (7) and Nb (8) surface species that were loosely bound to the alumina surface. Jasik et al. [16] reported a study Ni catalyst supported on Niobium oxide for hydrogenation of benzene. A strong interaction between metal and support was obtained when using niobium oxide as support. However, the use of Nb_2O_5 in chemical looping combustion as a promoter has not been sufficiently studied. The use of Nb_2O_5 as a promoter of the support in OC can therefore be a new alternative to the reduction and oxidation reaction. In the present work, niobium-promoted on the support with 5 wt.%, 10 wt.% and 15 wt.% niobium (Nb) loading on the □-Al_2O_3 were prepared using wet impregnation method to study the redox behaviour of the compound. The reactivity of the OCs is measured by thermogravimetric analysis using 5 % methane balance with nitrogen.

2.0 Methodology

2.1 Synthesis of oxygen carriers

The OCs, Nb/alumina were prepared by wet impregnation according to the method proposed by [11] using Nb_2O_5 as the Nb promoter. Aqueous solutions of □-Al_2O_3 and Nb_2O_5 were prepared separately in deionized water by stirring for 10 min. Subsequently, the two solution of Nb_2O_5 and □-Al_2O_3 were mixed in a beaker. The mixture obtained were dried in a dry oven at 120 °C for 12 h and calcinated for 3 h at 500 °C. In this study, OC Nb/alumina impregnated with 5 wt.%, 10 wt.% and 15 wt.% of Nb loading were denoted as 5NbA, 10NbA and 15NbA. Nb represents niobium while A represents alumina support. All OCs prepared were subjected to characterization techniques.

2.2 Oxygen carrier characterization

Temperature-programmed reduction (H_2-TPR) and temperature-programmed oxidation (TPO) experiments were carried out using a Thermo Science TPDRO1100 fitted with a thermal conductivity detector. The experiments were carried out using a calculated quantity of samples (50 mg). All the OCs were put in a U-shaped quartz reactor and heated in the presence of H_2 (pre-mixed gases, 5.06 vol% H_2 in N_2) for TPR analysis and O_2 (pre-mixed gases, 5.00 vol% O_2 in Helium) with a flow rate of 30 mL/min at a rate of 10 °C/min from 100 to 1000 °C.

The morphology of OCs was investigated using a scanning electron microscope (Hitachi TM3030 TableTop). Before analysis, the OCs were coated by sputtering (Emitech Quorum SC7620) at 20mA for 60s with gold using a coating machine.

2.3 Chemical looping combustion

The reduction and oxidation were carried out using TGA model Q50 Thermal Analyzer. The calcined sample was loaded on the platinum pan and heated up to 900 ℃ at a rate of 20 ℃/min in nitrogen (20 mL/min). The OCs sample was then exposed to 5 % methane balance with nitrogen at a flow rate of 80 mL/min for 3 minutes after reaching the set temperature to allow reduction process. In order to avoid the mixing of reduction and oxidation gases, the TGA chamber was then purged with 20 mL/min of nitrogen for 3 minutes, and the sample was exposed to for 10 minutes using flowrate 80 mL/min of air for oxidation reaction. The reduction and oxidation segments were repeated for 10 times, respectively. All redox reactions in this paper were at least duplicated. The oxygen transfer capacity (OTC) of redox cycles was calculated using the following formula:

$$R_o = \frac{m_{ox} - m_{red}}{m_{ox}} \quad (1)$$

wherem_{ox} is the OC weight after its completely oxidized and m_{red} is the mass of OC when it is fully reduced.

3.0 Results and discussion

3.1 Oxygen carrier characterization

3.1.2 Temperature-programmed reduction and oxidation analysis

The TPR and TPO were performed to study the reduction and oxidation of calcined OCs, the amount of hydrogen and oxygen uptake, interaction between promoter and support as well as reduction and oxidation temperature for redox properties. Fig. 2 and 3 shows TPR and TPO profile, respectively. The reduction peak of 5NbA, 10NbA and 15NbA presented broad peaks of two main contribution at 500-600 ℃ and 850-950 ℃ [17]. For the second region, as the composition Nb increased, the reduction temperature was increased from 894 ℃ to 899 ℃ and to 905 ℃. This finding indicates the strong interaction between the promoter and support. However, forthe first region, reduction temperature different Nb loading was in the range of 529 – 560 °C. The presence of Nb at low temperatures between 500 °C and 600 °C reveals that these Nb species have a poor association with the support. 1st 2ndregi

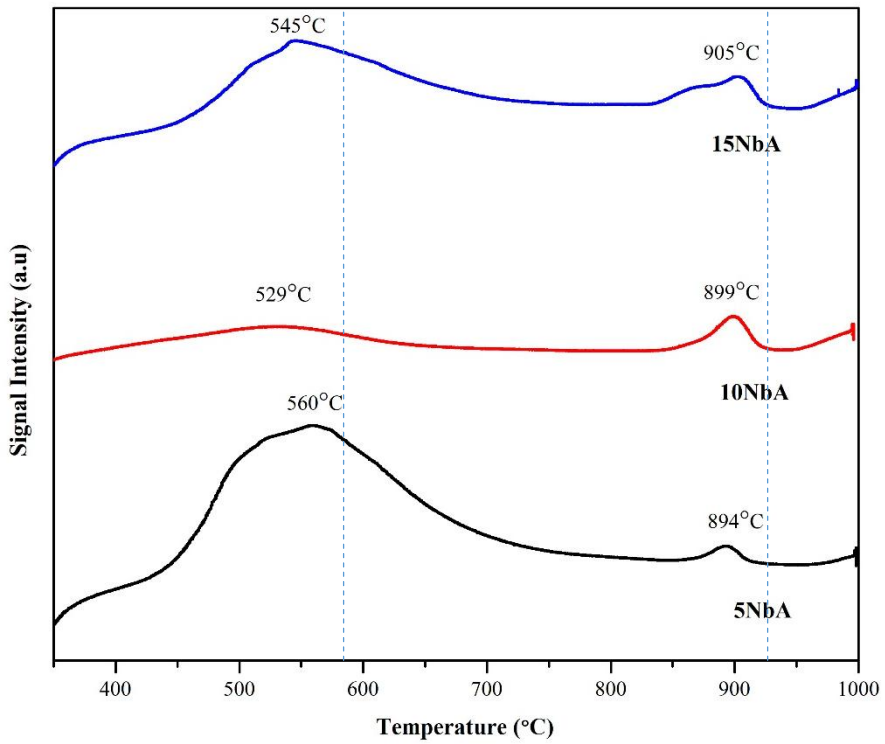


Fig. 2. H_2-TPR profile for synthesized OCs

As shown in Fig 3, the TPO profile revealed the difference between oxidation temperature of three OCs after the TPR experiment. An increase in the Nb loading decreased the oxidation temperature, which is contradict with the reduction process as shown in Fig 2. Similar with TPR, oxidation profile showed two main contribution which are at 400-550 ℃ region and 700 ℃ and above, except for 5NbA. Oxidation temperature below than 550 ℃

indicated lower interaction between promoter and support while above 700 ℃ signifies a strong interaction. It was observed that only one peak was obtained for 5NbA in TPO analysis. The presence of 5% Nb_2O_5 does not practically change the OCs for oxidation in relation to Al_2O_3. Table 1 showed hydrogen and oxygen consumption for all prepared OCs. It was observed that at low Nb loading the hydrogen uptake was the highest followed by 15NbA and 10NbA. The incorporation of more niobium oxide decreased the hydrogen adsorption capacity also reported byF.B. Noronha et al. [18]. The result showed that high hydrogen consumption were shown to result in higher oxygen ability (cf. Section 3.2.1), which is in line with literature study [11]. While for oxygen consumption, 10 wt.%Nb loading showed the greatest number amount of gas adsorbed followed by 15NbA and 5NbA. This variation could possibly due to high interaction Nb with support, hence need more amount of oxygen to oxidize the reduced sample.

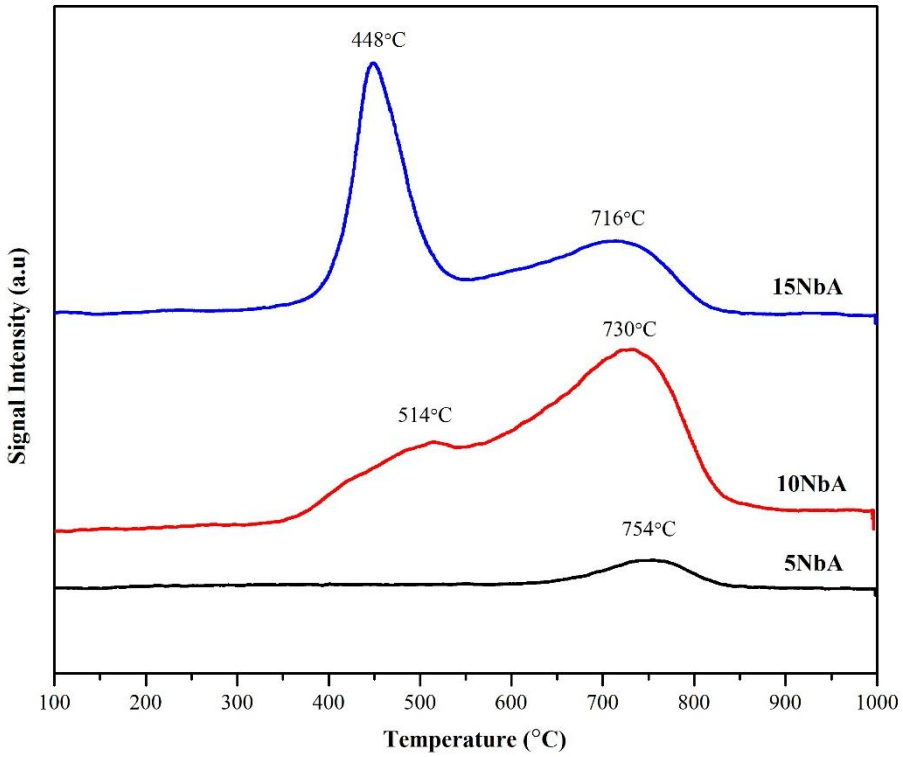


Fig. 3. Temperature-programmed oxidation (TPO) of fresh calcined OCs

Table 1.

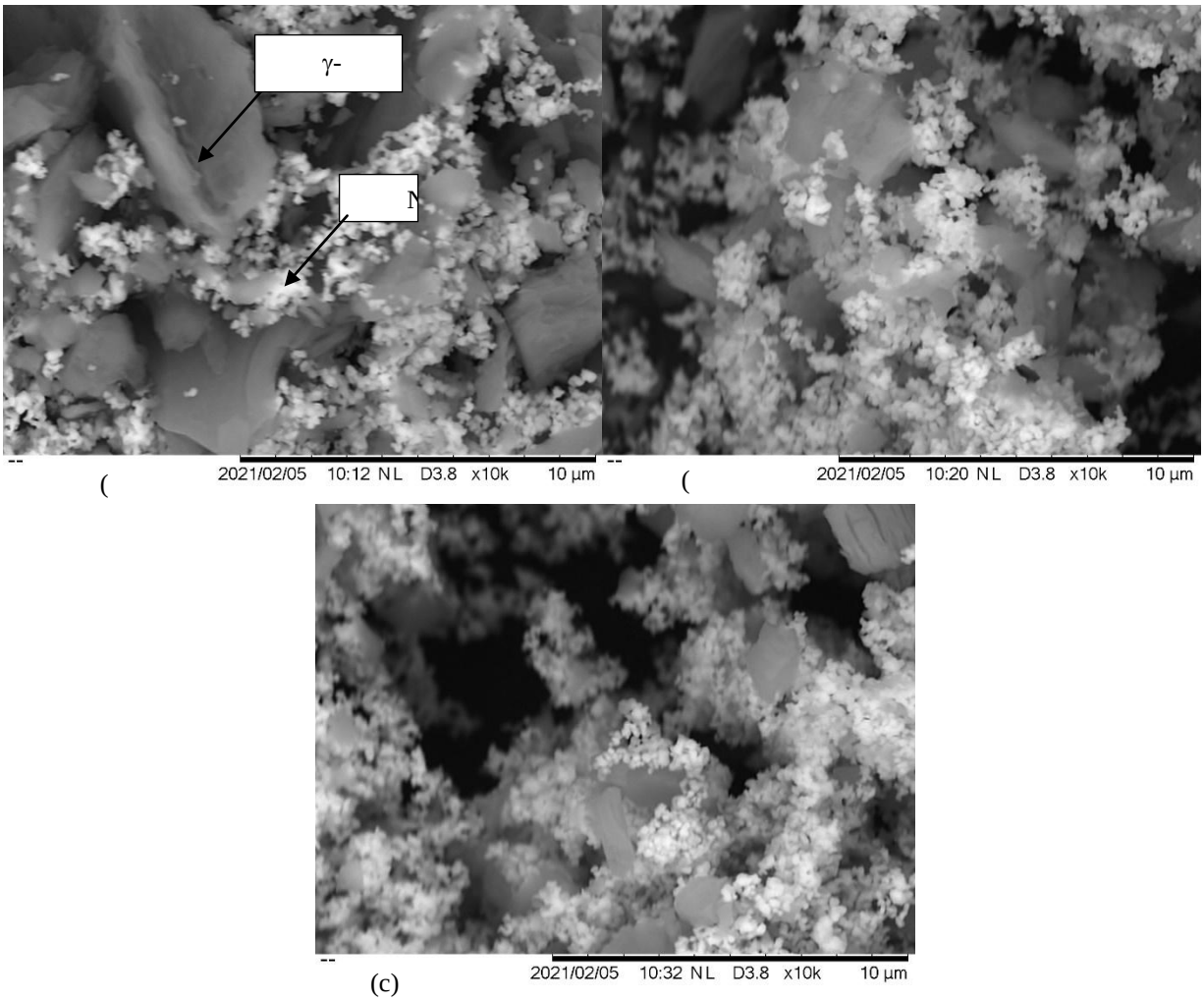
Hydrogen and oxygen consumption of all OCs

Oxygen Carriers	**5NbA**	**10NbA**	**15NbA**
H_2 consumption (µmol/g)	896.40389	34.72038	394.14411
O_2 consumption (µmol/g)	92.47842	451.65445	335.04722

3.1.2 Scanning electron microscopy

Fig. 4 shows SEM images of OCs at different Nb loading. The SEM analysis was applied to compare the dispersion of Nb on the alumina support. As shown in the figure, all three OCs exhibitedsimilar shape and size. Increasing the Nb loading did not give significant effect on the morphological properties of the OCs. From the SEM images, the smooth surface represents the support while the white particles indicated niobium element. Among the OCs, OC prepared at highest Nb loading (15 wt.%) exhibited distinct structure as shown in Fig. 4 (c).

It was observed that the Nb element was poorly distributed on the support and resulted in an agglomeration. This observation indicates that higher metal loading decreased the metal dispersion on the support [19].



(c)

Fig. 4. SEM images of the OCs; (a) 5NbA; (b) 10NbA; (c) 15NbA

3.2 Chemical looping combustion

3.2.1 Reactivity of oxygen carriers

First full cycle reduction and oxidation of three OCs were investigated at 950 °C by using 5% CH_4/N_2 as reducing gas and air as oxidizing gas. To prevent mixing between reducing and oxidizing gas, nitrogen was purged for3 min. Fig. 5 displays the results for first cycle of 5NbA, 10NbA and 15NbA while Fig. 6 summarizes the oxygen transfer capacity (OTC) calculated using equation 1 for all prepared OCs. The reduction and oxidation was performed to see the weight variation of different metal composition (Fig. 5). The result showed that the highest OTC was found to be 0.003 mg for 5 wt.% of Nb loading. This finding could be due to a higher metal dispersion on the support (cf. Section 3.1.2) compare with 10 and 15 wt.% of Nb loading, hence increase the ability of OC to gain and release oxygen. While OTC for 10NbA and 15NbA were 0.00226 O_2/mg and 0.002753 O_2/mg, respectively. This result prove that Niobium material have tendency to gain and release oxygen and can be used as a promoter in the chemical looping combustion process [20]. As shown in Fig. 5, OCs with Nb promoted show weight loss and weight gain around 45 min to 55 min, respectively.

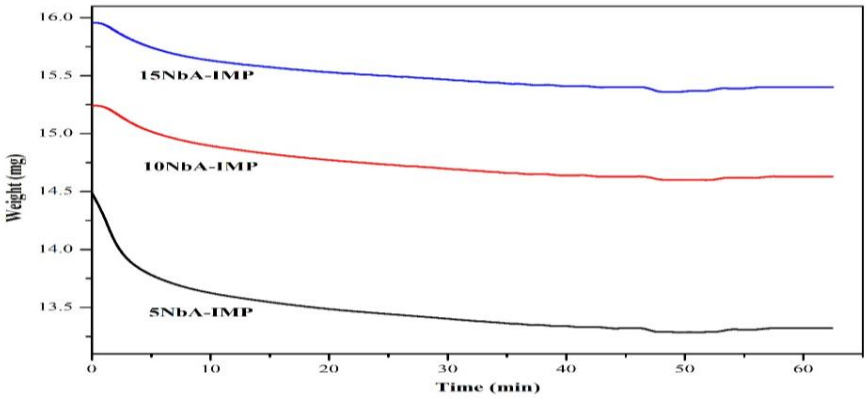


Fig. 5. First redox cycle for OCs prepared via wet impregnation method.

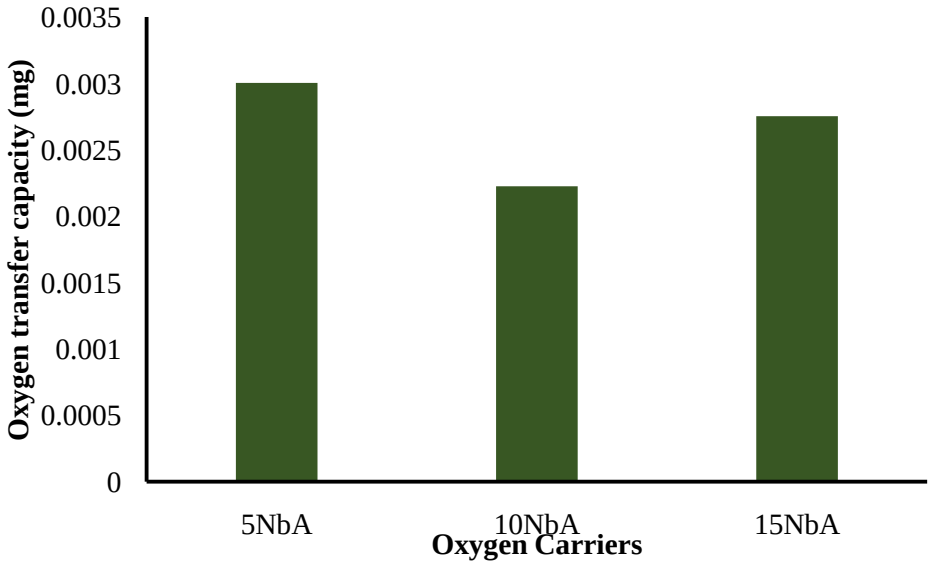


Fig. 6. Oxygen transfer capacity of prepared OCs

4.0 Conclusion

In this study, the effect of Nbloading (5, 10 and 15 wt.%) on the physical properties and oxygen transfer capacity of the Nb/alumina OCs prepared by wet impregnation method was investigated. Nb/alumina OC showed 0.3% oxygen transport capacity (highest) for 5 wt.%Nb loading, while 0.28% for 15 wt.% of Nb loading and 0.22% for 10 wt.% of Nb loading. Similar size and shape were observed for all prepared OCs. The reduction temperature for 5NbA, 10NbA and 15NbA was 560 ℃, 529 ℃ and 545 ℃, respectively, which suggested the reduction reaction takes place about 500 ℃ and above. From hydrogen uptake analysis, it was noticed that high hydrogen consumption (896 μmol/g) resultedin higher oxygen capacity (0.3%). Above all, this study suggest that Niobium promoted on the □-Al_2O_3 can be used for oxygen carrier for chemical looping combustion.

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