A Deposition Study on a Membrane Integrated Chemical Looping Air Separation Unit

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ABSTRACT

A membrane integrated chemical looping air separation (MI-CLAS) process is proposed as an energy efficient air separation unit with the potential to be tailored to oxy fuel power plants. The energy saving of a chemical looping air separation (CLAS) process is 12% of an equivalent cryogenic system however this is further increased by 36% with an integration of oxygen transport membranes into the CLAS. The working principle of the CLAS process involves the cyclic oxidation (O_2 coupling) and reduction (O_2 decoupling) of metallic oxide particles between a set of two interconnected reactors. As metallic oxide particles are in contact with the membranes surface, the success of the membrane integration into the CLAS process largely depends on the degree of deposition. It has been previously demonstrated in the literature that operational problems caused by deposition severely affected the performance of various industrial units such as boilers. Deposition experiments were conducted with coal particles and air in a fluidized bed channel. Effects of various parameters such as air flow rate, membrane pore size and time on the particle deposition were studied. Deposition on the membrane surface was recorded using a Phantom v5 high speed camera and images were processed by Image J software to determine the surface coverage of membranes by particles. Results indicated that particles were transported onto surfaces through deposition mechanisms such as turbulent diffusion, drag force and electrostatic forces. Experimental results gave an insight into the particle deposition at high velocities in two phase flow.

INTRODUCTION

A chemical looping air separation (CLAS) process [1] was developed at the University of Newcastle for the production of oxygen which demonstrated a significant improvement in the energy efficiency over the conventional cryogenic distillation process. More details about the CLAS process can be found elsewhere [1].
An improved process called a membrane integrated CLAS (MI-CLAS) has been developed with enhanced energy savings over the conventional CLAS process. In the MI-CLAS process, oxygen transport membranes are integrated into the CLAS unit which eliminate any need for the utilisation of steam condenser, boilers and additional heat exchangers. The conventional CLAS and membrane integrated CLAS (MI-CLAS)

processes were simulated using ASPEN Plus software version 7.3 [2]. The membrane integrated CLAS systems were studied from an energy savings perspective under ideal (i.e., 100 % oxygen recovery via oxygen transport membranes) and practical (i.e., practical oxygen recovery) conditions. Under ideal conditions, an energy saving of 30 % was achieved via membrane integration into the CLAS process [2]. The energy savings of the membrane integrated CLAS process under practical conditions were almost 10 % and 13 % at low and high temperature ranges, respectively, over the typical CLAS process [3].

As the fundamental concept in the CLAS process involves the circulation of oxygen carriers between two interconnected reactors, membranes may be prone to a particle deposition in the membrane integrated CLAS unit. Therefore, the success of the membrane integration into the CLAS unit largely depends on understanding the phenomenon which could limit the oxygen permeation through the membrane under conditions pertinent to the CLAS process. Of these, particle deposition was considered as the most important phenomenon. The main objective of this study was to examine the degree of particle deposition on the surface of membranes. Deposition experiments were initially conducted in the perspex fluidised bed channel using coal particles and air at room temperature resulting in no reaction. As the rate of the electrostatic charge generation was significantly high during experiments, its adverse impact on the experimental results was inevitable. Consequently, the experimental setup was redesigned and replaced with a combination of conductive glass and aluminium sheets. The conductive glass experimental setup was earthed to dissipate all the electrostatic charge generated. The effects of various parameters such as bed inclination angle, air flow rate, particle size range and time on the degree of particle deposition on the membrane surface were studied. The surface coverage of the membranes was monitored using the Phantom v5 high speed camera and images were processed using the Image J software.

EXPERIMENTAL

Materials

Ceramic membranes with a thickness of 2.5 mm and 47 mm diameter and the pore sizes ranging from 150 kDa to 1.4 μm were purchased from the Sterlitech ® Company (USA). The active layer of ceramic membranes with 150 kDa pore size was titania (TiO_2) while a mixed zirconia-titania (ZrO_2- TiO_2) layer was utilised for membranes with pore sizes of 0.2 μm to 1.4 μm. Specifications of the MOURA coal sample used are reported in Table 1. The particle size range of the coal sample was +90 μm to -150 μm which was measured using the Malvern Mastersizer 2000. Compressed air was supplied by Coregas (Australia). The Phantom v5 high speed camera was used to record particle deposition throughout the course of the experiments and the Image J software was used to process images.

Table 1: MOURA coal sample analysed in accordance with Australian Standards AS1038.3, AS1038.6.1, AS1038.6.2, AS1038.9.3.3 and AS4264.1.

Analysis Basis	**(%)**	**(ad)**	**(daf)**
Analysed moisture	(%)	2.4	
Ash	(%)	9.8	
Volatile matter	(%)	31.5	35.9
Fixed carbon	(%)	56.3	
Total sulphur	(%)	0.42	
Carbon	(%)	73.4	83.5
Hydrogen	(%)	4.67	5.32
Nitrogen	(%)	1.56	1.78
Oxygen (by difference)	(%)	7.8	8.88

Deposition experimental

Perspex and conductive glass experimental apparatus were designed and produced in order to examine the particle deposition phenomenon. Since the preliminary apparatus was made of Perspex, the level of electrostatic charge generation was considerably high which affected the results. Consequently, the secondary apparatus was designed and fabricated using a combination of aluminium and fluorine-doped tin oxide (FTO) coated glass.

Perspex apparatus

The perspex experimental setup consists of a stand and a deposition vessel. The stand was used to securely hold the deposition vessel at desired inclination angles between 0 to 90 degrees from the horizontal surface. A deposition vessel consists of a bottom section, a gas distributor, a channel and a top section. The perspex experimental setup is shown in Figure 1.

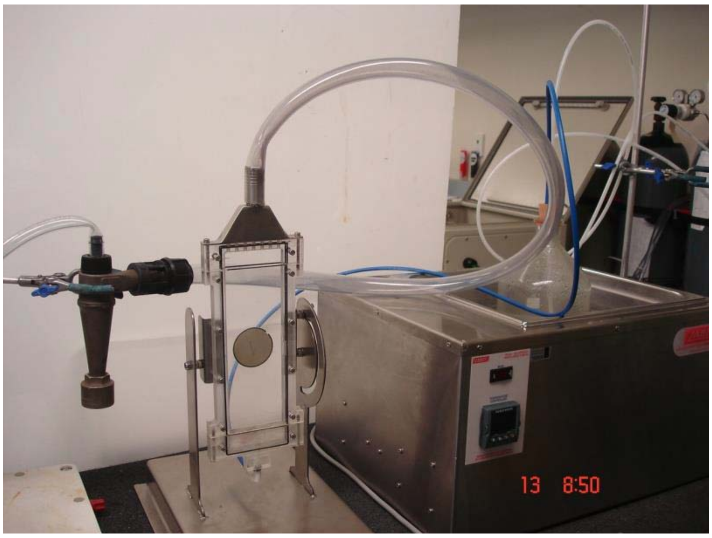


Figure 1: Perspex apparatus

Conductive glass apparatus

In the conductive glass design, fluorine-doped tin oxide (FTO) coated glass (purchased from Sigma-Aldrich ®) was used in combination with aluminium sheets in order to overcome the electrostatic charge issue. The conductive glass experimental setup is shown in Figure 2. The apparatus was earthed to fully dissipate the electrostatic charge to the ground.


Figure 2: Conductive glass apparatus

Experimental procedure

Compressed air (25 °C, 1 bar) was passed through the bed of coal particles resulting in gas solid fluidisation. The flow rate of the air was in the range of 5 l/min to 25 l/min this was achieved using the Dwyer variable area flow meter (Dwyer instruments, Inc., USA). The particles flowing out of the channel were collected using the cyclone as shown in Figure 1. The impact of various parameters such as inclination angle (0-90 degrees), air flow rate (15-25 l/min), membrane pore size (150 kDa, 0.2μm, 0.8μm and 1.4 μm) and time (1-30 min) on the particle deposition was investigated. Surface coverage of membranes was recorded under different experimental conditions via the Phantom v5 high speed camera and images were processed using the Image J software.

RESULTS AND DISCUSSION

Preliminary experiments were conducted using the perspex experimental setup. The pertinent experimental results were affected by the presence of a severe electrostatic charge as shown in Figure 3. Attempts were made to minimise the electrostatic charge to an acceptable level using the methods recommended in the literature [4-12], however this was unable to be achieved. As a consequence, deposition experiments were then conducted using conductive glass. The results of the perspex and the conductive glass experiments are discussed in the following sub sections.

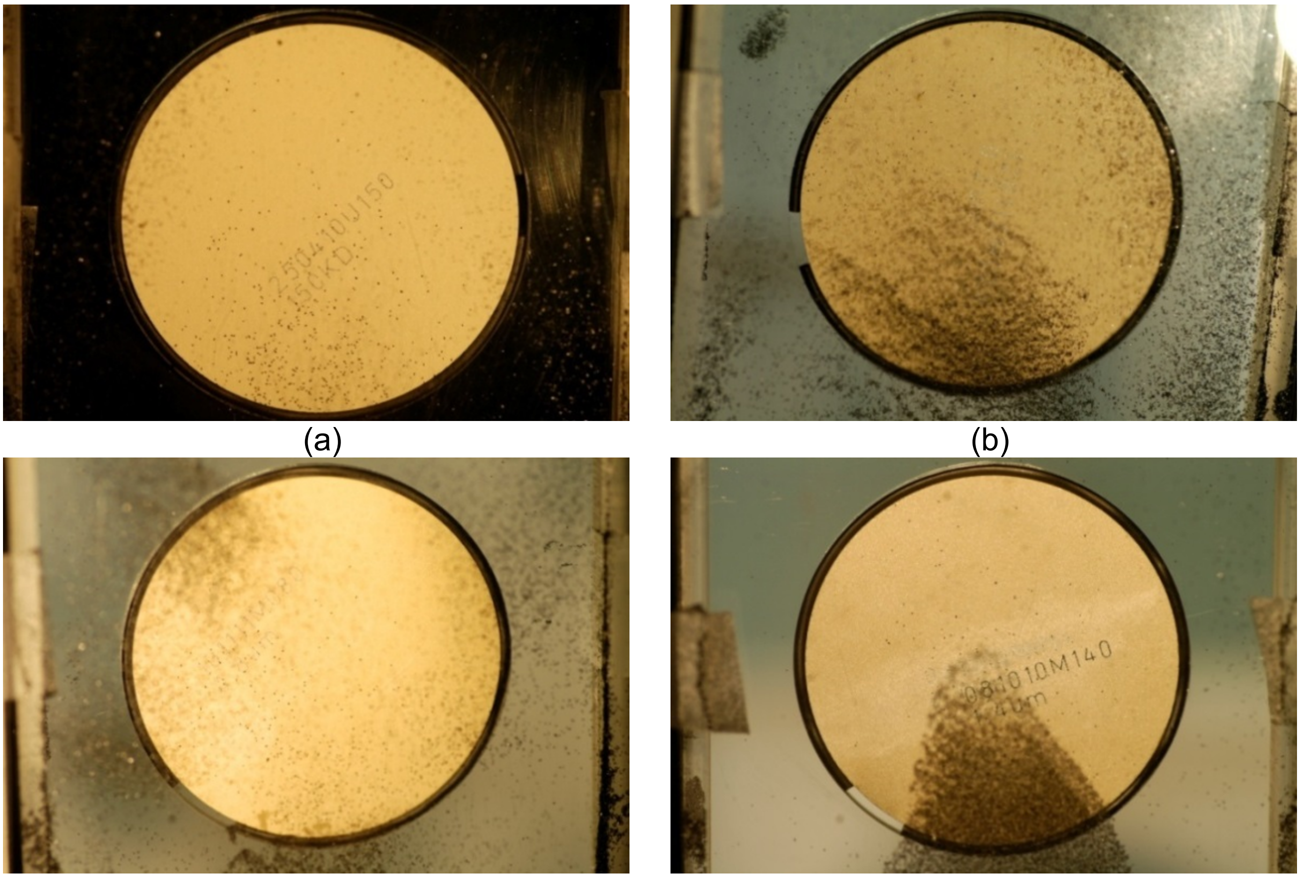


Figure 3: The effect of a sever electrostatic charge on membranes with (a) 150 kDa (b) 0.2 μm (c) 0.8 μm and (d) 1.4μm pore sizes tested using a particle size of 125-150μm at a 75 degree inclination angle and 25 l/min air after 10 minutes.

Perspex experimental results

In this section, the effects of the bed inclination angle, particle size, air flow rate, time and membrane pore size on the particle deposition were studied.

The effect of bed inclination angle

The effect of bed inclination angle on the particle deposition is illustrated in Figure 4. The surface of membranes with 150 kDa, 0.2 μm and 0.8 μm pore sizes covered by 125-150 μm coal particles is compared at 75 and 90 degrees bed inclination angles. As seen, a higher surface coverage is observed at the 90 degree bed inclination angle. At the 75 degree bed inclination angle, especially at low air flow rates, the gravity force is more effective on the terminal settling velocity of particles therefore a considerably lower surface coverage is obtained.

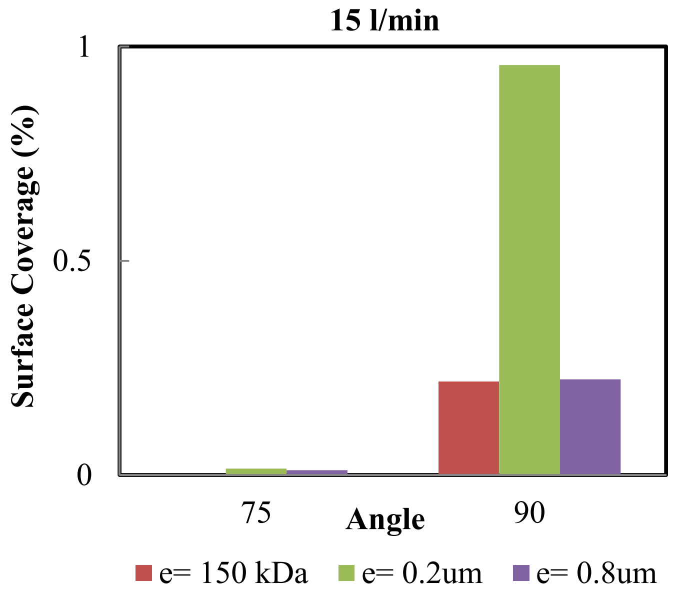
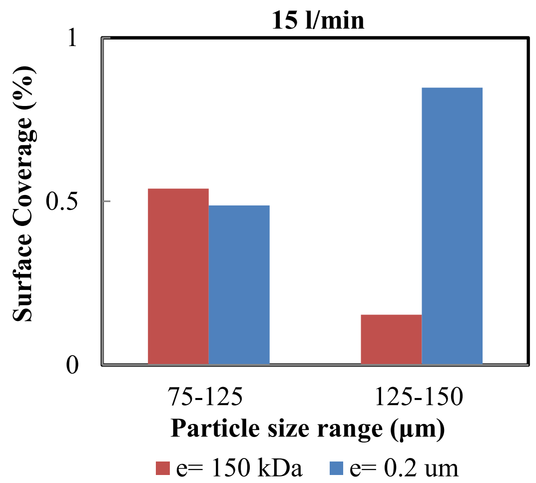


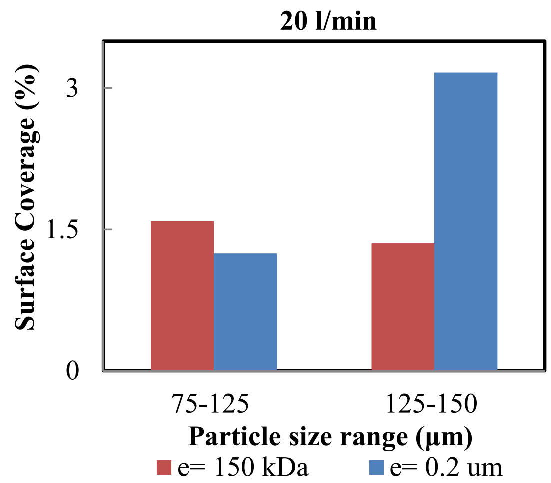
Figure 4: The effect of inclination angle on the particle deposition with the size range of 125-150 μm, at 15 l/min air after 10minutes.

The effect of particle size

The surface coverage of membranes with 150 kDa and 0.2 μm pore sizes was compared at two particle size ranges of 75-125 μm and 125-150 μm. They were fluidised by air at a flow rate of 15 l/min and 20 l/min for 5 minutes at a 90 degree bed inclination angle (See Figure 5). The surface coverage of the membranes with 150 kDa pore size decreased with increased particle size range. In contrast, the surface coverage of membranes with 0.2 μm pore size increased with increased particle size range.



(a)



(b)

Figure 5: The effect of particle size on the deposition fluidised with air at (a) 15 l/min and (b) 20 l/min at 90 degrees inclination angle after 5 minutes.

The effect of time

The effect of time on the deposition of particles with the size ranges of 125-150 μm and 75-125 μm is shown in Figures 6 (a) and 6 (b) conducted under 15 l/min air and 90 degrees bed inclination angle. As seen, time has a diverse effect on the deposition of particles with different size ranges. Surface of membranes with 150 kDa and 0.2 μm pore sizes covered by 125-150 μm coal particles increased with the course of time. However, the surface coverage conveyed a decreasing trend for the coal particles with the size range of 75-125 μm.

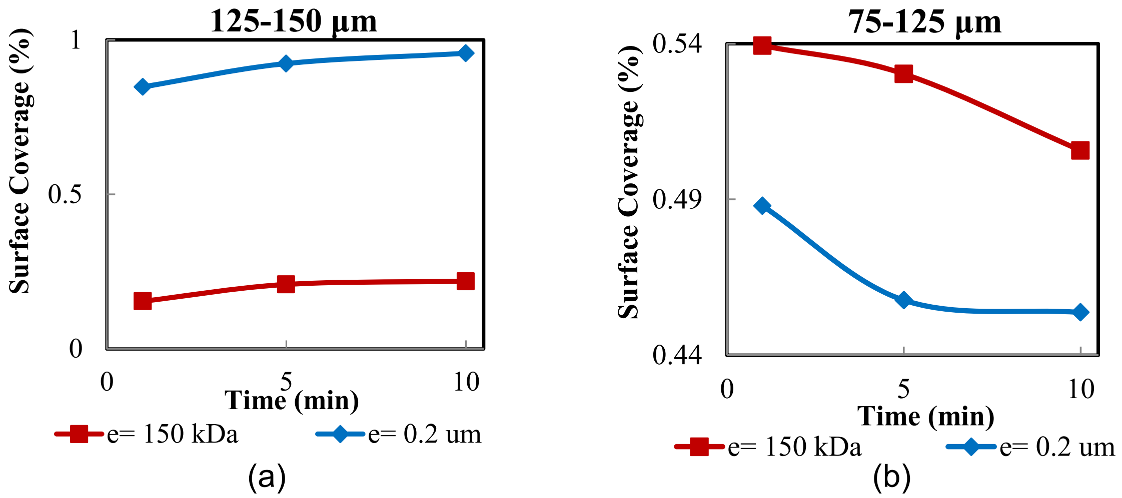


Figure 6: The effect of time on deposition of particle with the size range of (a) 125-150 μm and (b) 75-125 μm under 15 l/min air, and 90 degrees inclination angle.

The effect of gas flow rate

The effect of an increase in the air flow rate on the particle deposition at a 75 degree bed inclination angle is depicted in Figure 7. As shown, the surface coverage increased with an increase in the air flow rate for membranes with three different pore sizes at the 75 degrees bed inclination angle.

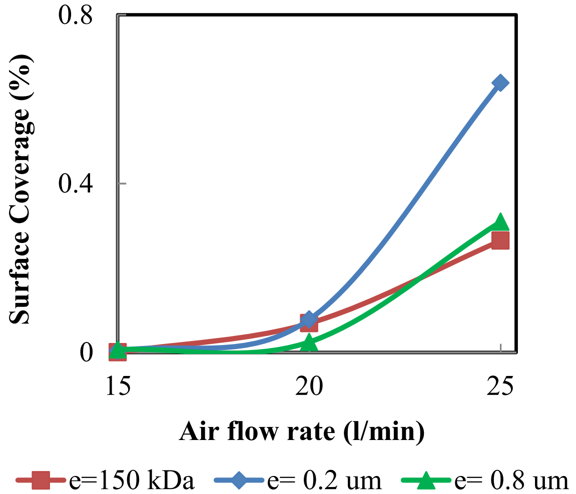


Figure 7: The effect of air flow rate on the particle deposition with the size range of 125-150 µm at a 75 degree bed inclination angle after 5min.

The effect of membrane pore size

Figure 8 shows the surface coverage of membranes as a function of the membrane pore size at 75 and 90 degrees bed inclination angles and also at the air flow rates of 15 and 25 l/min. As seen, a variation in the membrane pore sizes had no significant effect on the surface coverage of membranes at 15 l/min air flow rate and a 75 degree bed inclination angle. However, the surface coverage significantly decreased with increased membrane pore size at 25 l/min air flow rate and 90 degree bed inclination angle. The surface coverage of membranes increased with an increase in the membrane pore size and then levelled off at a 25 l/min air flow rate and 75 degree bed inclination angle. In contrast, the surface coverage increased to a maximum peak and then decreased with an increase in the membrane pore sizes at 15 l/min air flow rate and 90 degrees bed inclination angle.

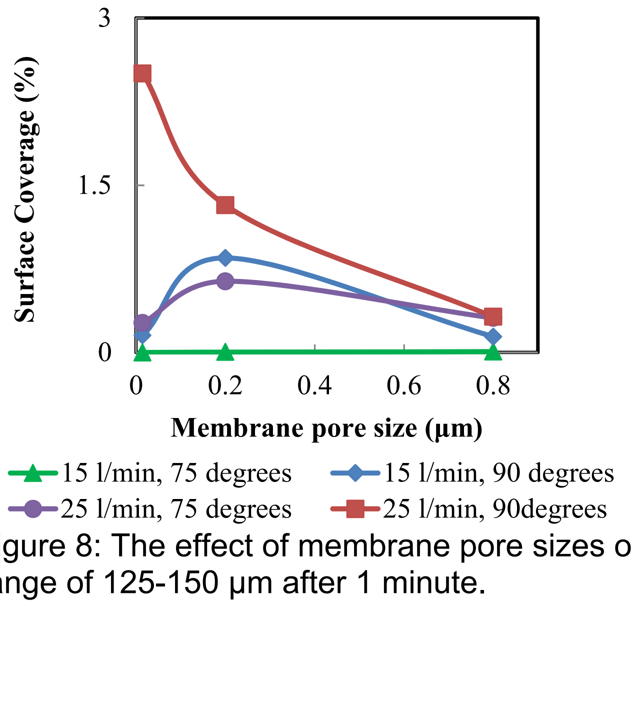


Figure 8: The effect of membrane pore sizes on the particle deposition with the size range of 125-150 µm after 1 minute.

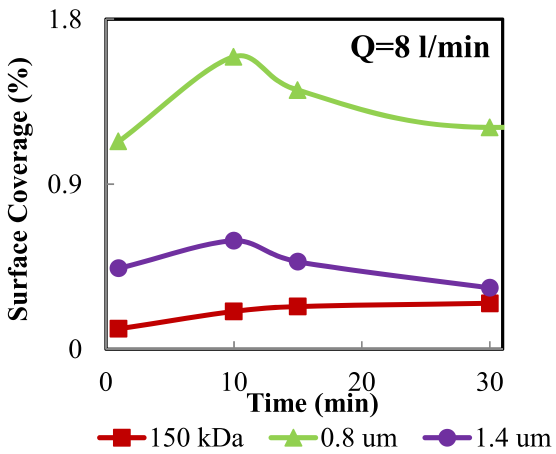
Conductive glass experimental results

In this section, the experimental results of the conductive glass setup are discussed. Results are presented at constant air flow rate, membrane pore size and time.

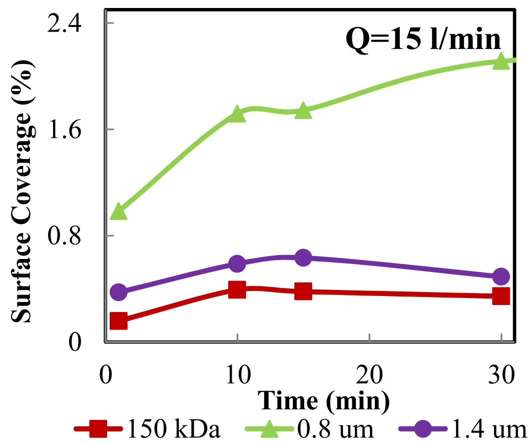
Surface coverage of membranes at constant air flow rate

The surface coverage of membranes are depicted as a function of membrane pore sizes and time at constant air flow rates of 8 l/min and 15 l/min (See Figures 9 (a) and (b)). At the constant air flow rate of 8 l/min, the surface coverage of membranes with 150 kDa pore size gradually increased with time. However, the surface coverage of membranes with 0.8 µm and 1.4 µm pore sizes showed a maximum peak at 10 minutes and then decreased with time.

At the constant air flow rate of 15 l/min, the surface coverage of membranes with 150 kDa and 1.4 µm increased gradually with time and then levelled off after 15 minutes. However, the surface coverage of membranes, with 0.8 µm pore size, considerably increased with time. A comparison between two graphs showed a higher surface coverage of membranes with time at higher air flow rates.



(a)



(b)

Figure 9: Surface coverage at constant flow rates at (a) 8 l/min, (b) 15 l/min.

Surface coverage of membranes at constant membrane pore sizes

The surface coverage of membranes as a function of time and air flow rates are depicted in Figures 10 (a) and (b) at constant membrane pore sizes of 150 kDa and 0.8 µm. Figures 10 (a) and (b) showed a peak in the surface coverage of both membranes at 10 l/min air flow rate. However, the surface coverage significantly decreased at the air flow rates greater than 10 l/min. Figures 10 (a) and (b) showed a considerably higher surface coverage of membranes with 0.8 µm pore size compared with the one with 150 kDa pore size.

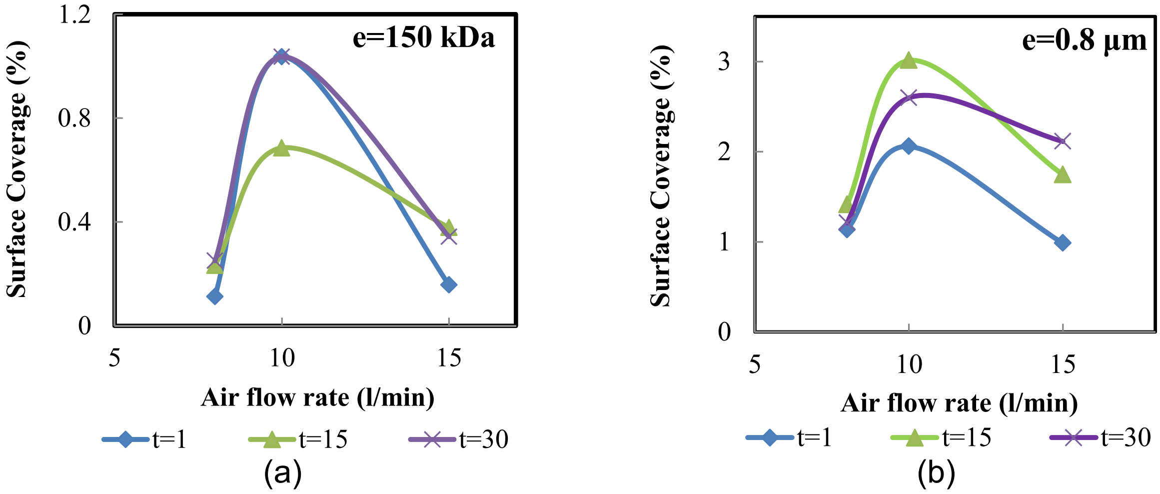


Figure 10: Surface coverage at constant membrane pore size (a) 150 kDa and (b) 0.8 um

Surface coverage of membranes at a constant time

The surface coverage of membranes at constant time as a function of air flow rate and membrane pore sizes are illustrated in Figures 11 (a-d). Figures 11 (a) and (b) showed a maximum in the surface coverage of membranes, with 150 kDa and 0.8 µm pore sizes, at 10 l/min air flow rate at constant time of 1 minute and 30 minutes. Figure 11 (a) and (b) also showed a higher surface coverage of membranes with 0.8 µm pore size compared with 150 kDa pore size with increased air flow rate and time.

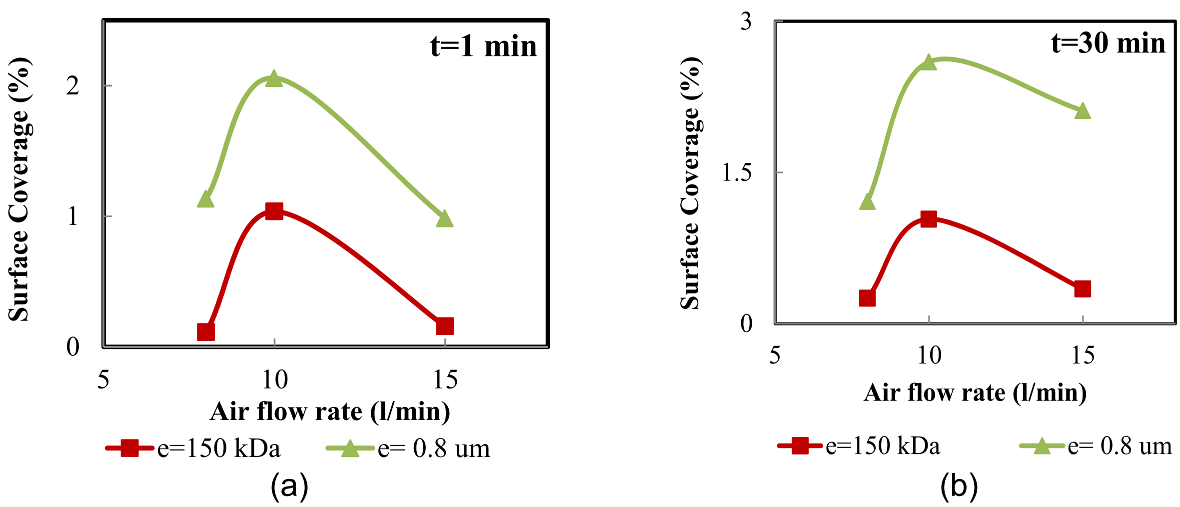


Figure 11: Surface coverage at constant time of (a) 1 min and (b) 30 minutes.

CONCLUSIONS

The particle deposition phenomenon was examined for the membrane integrated chemical looping air separation unit. Deposition experiments were conducted in the fluidised bed channel using coal particles and air at room temperature resulting in no apparent reaction. The effects of various parameters such as bed inclination angle, air flow rate, particle size range and time on the degree of particle deposition were studied. The surface coverages of membranes were taken with the Phantom v5 high speed camera and images were processed using the Image J software. Results showed a significantly lower deposition rate on membranes surface at a 75 degree bed inclination angle and low flow rates of air. Results also demonstrated that the least surface coverage was obtained at a 75 degree bed inclination angle and 15 l/min air flow rate regardless of the membrane pore size range. Among all parameters studied, the air flow rate and the inclination angle had the most impact on the particle deposition on the membrane surface. According to the presented results, the impact of particle deposition on the membrane performance can be ignored as the rate of particle deposition was relatively low.

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