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Enhancing Carbonation Process during Carbon Dioxide Capturing Applying Jetting Fountain Fluidized Bed

تحسين اجراء الكربنة اثناء التقاط ثاني اوكسيد الكربون من خلال استخدام مفاعل المهده المميع ذي النافث النافورى

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KEYWORDS

Fluidized bed, Limestone, CO₂ capture...

المخلص العربي: يتطلب الوضع الحالى لمصادر الطاقة ومعدلات الاستهلاك استمرار الاعتماد على الوقود الاحفورى كمصدر رئيسى للطاقة ، الا أنه يمثل أيضا المصدر الرئيسى لغاز ثاني أكسيد الكربون والذي تتزايد الدعوة للحد من انبعاثه كونه أحد الأسباب الرئيسية لما يعرف بظاهرة الصوبة الزجاجية وبالتالي الارتفاع المستمر لدرجة حرارة الغلاف الجوى وما يترتب عليه من آثار سلبية، وفى هذا الإطار فإن فصل ثنائى أكسيد الكربون وتخزينه يعتبر ضرورة لتقليص الإنبعاثات إلى الحد المطلوب .

يعد استخدام حبيبات الحجر الجيري فى فصل ثاني أكسيد الكربون من غازات العادم أحد الطرق الواعدة حيث تتم عملية الفصل من خلال اجرائين دوريين فى مفاعلين ذوى مهده مميعة منفصلين، فى المفاعل الأول تتم عملية الكربنة عند درجة حرارة 700-650 °C حيث تتفاعل حبيبات أكسيد الكالسيوم مع ثاني أكسيد الكربون لتتحول الى كربونات الكالسيوم ثم تنتقل الحبيبات الى المفاعل الثانى لتتم عليها عملية الكلسنة عند درجة حرارة 800-900 °C حيث تتحول كربونات الكالسيوم الى أكسيد الكالسيوم محررة ثاني أكسيد الكربون والذي يسهل جمعه وضغطه وتخزينه.

يهدف هذا البحث الى تحسين اجراء الكربنة من خلال استخدام مفاعل لمهده مميعة ذى نافث نافورى تم تصميمه وتصنيعه وتركيبه بالمعمل، المفاعل له قطر داخلى 105 ملم وارتفاع 4000 ملم. وقد استخدمت مادة حبيبات الحجر الجيري المكلس لتتم عليها عملية الكربنة. فى تجارب هذا البحث تم تغذية خليط من النيتروجين وثانى أكسيد الكربون بتركيز (CO₂ 15%) خلال مهده من حبيبات مادة الحجر الجيري المكلس، وتم قياس تركيز ثاني أكسيد الكربون بعد المهده لتقدير كميته التى تفاعلت مع الحبيبات وانفصلت عن الغازات، وقد أجريت التجارب فى المفاعل فى صورته التقليدية وفى الشكل المبتكر مع النافث النافورى لعمل دراسة مقارنة، وقد تم دراسة تأثير ظروف التشغيل على أداء المفاعل التى تشمل درجة حرارة المهده، وسرعة التمييع، نسبة هواء النافث، وارتفاع فتحة النافث . وتشير النتائج التى توصلت اليها الدراسة الحالية الى ان المهده المميعة ذى النافث النافورى هو اكثر كفاءة فى اتمام عملية الكربنة مقارنة بالمهده المميعة التقليدية، حيث يزيد من معدلات الكربنة ويقلل من الوقت اللازم لكامل عملية التحويل، وذلك لأنه يحسن كثيرا من كفاءة التلامس بين الهواء وحبيبات المهده. وجد أيضا أن معدل الكربنة يتحسن مع زيادة درجة حرارة المهده وسرعة التمييع. على جانب آخر وعند دراسة تأثير نسبة هواء النافث وارتفاع فتحة النافث ثبت ان هناك قيم بينية تكون عندها لعملية الكربنة قيمة عظمى.

Abstract: - Carbon dioxide from fossil fuel burning is the major contributor to the greenhouse effect from human activities. Within the many options and actions for mitigation of greenhouse gas emissions, CO₂ capture and storage is emerging as a viable option to achieve the very deep cuts in emissions that might be needed in the medium term. One promising means of CO₂ capture for fossil fuels based power plants is to use a lime carbonation-calcination cycle. The aim of the current work is to enhance the carbonation process by applying jetting fountain fluidized bed. A jetting fountain fluidized bed reactor has been designed, fabricated and installed to carry out the experimental work. It has 105 mm ID and 4000 mm height. Jabal al-Tair limestone has been utilized as bed materials. During the tests a mixture of nitrogen and carbon dioxide, that simulates exhaust gases (15% CO₂), is fed through a

bed of lime particles. The lime particles are carbonated by capturing CO₂. The concentration of CO₂ is measured after the bed to estimate the quantities of CO₂ that combined with the lime particles. The influences of operating conditions including bed temperature, fluidization velocity, jet air ratio and jet orifice height on the capture efficiency of CO₂ have been studied. The findings of the present work indicate that the jetting fountain fluidized bed is more efficient in carbonation where the CaO conversion rate increases and the time required for full conversion reduces. Applying jetting fountain configuration enhances gas solids contact and improves the interphases mass exchange between bubbles and emulsion. The capture efficiency of CO₂ rate was found to improve with bed temperature and fluidization velocity. On the other side, studying the influences of jet air ratio and jet orifice height demonstrate that there is an

intermediate value at which the capture efficiency records an optimal.

1. INTRODUCTION

Mitigation of CO₂ emissions has growing concerns worldwide since it is one of the main reasons of global warming and climate change [1]. The energy generation that heavily based on fossil fuels accounts for 41% of global carbon emissions [2]. However, the current status of energy resources and demands obliges utilization of the fossil fuels in the mid to long term (2030–2100) [2]. On the other side, alternative or renewable energy sources still have fundamental hurdles to overcome, such as providing sufficient amounts of base-load electricity generation, in order to displace fossil-fuel power. In this context, carbon capture and storage (CCS) is a key principle in an international attempt to mitigate global warming [1].

The term Carbon dioxide Capture and Storage (CCS) refers to a number of technologies that can mitigate CO₂ emissions due to fossil fuels combustion. Development of efficient, economical, and realistic carbon capture and storage (CCS) technologies are demanded for application to fossil fuel-based power plants [3].

In general, three technological pathways are being developed for CO₂ capture: Post-combustion, Pre-combustion, and Oxy-fuel combustion.

Post-combustion capture by chemical absorption with mono - Ethanol amine (MEA), pre-combustion capture from a H₂-rich syngas by chemical or physical absorption and oxy-fuel combustion represent the reference CO₂ capture technologies suitable for short-term applications. Post-combustion capture by the calcium looping process is another emerging technology for mid-term applications that shows some potential advantages in terms of net efficiency and cost of CO₂ avoided [4].

One promising means of CO₂ capture for fossil fuels based power plants is to use a lime carbonation–calcination cycle (Calcium Looping, CaL). This process was originally proposed by Shimizu et al. [5], and uses CaO as a regenerable sorbent to capture CO₂ from combustion flue gases. Other processes that use CaO in combustion systems have been proposed [6,7], while others have also been considered for H₂ production routes [8–12]. CaL involves the separation of CO₂ using the reversible carbonation reaction of CaO and the calcination of CaCO₃ to regenerate the sorbent. Regarding to the large flow of flue gas treated in a CaL system that needs to be put into contact with CaO, a typical configuration for this process would consist of two interconnected circulating fluidized beds (CFB), calciner and carbonator, operating under atmospheric pressure (see Fig.1). Flue gases leaving the boiler of an existing power plant are fed into the carbonation unit, operating at temperatures between 873 K and 973 K, where the CO₂ reacts with the CaO coming from the calciner to form CaCO₃. Solids from carbonator are sent back to the calcination unit where CaCO₃ is calcined to form CaO, which is re-circulated again to the carbonator, and CO₂ as a concentrated stream.

A number of fundamental studies on the properties of Ca-based sorbent (reaction kinetics, sorption capacity, stability, poisoning, doping, and reactivation techniques)

have been conducted. Lab-scale installations have been successfully started both in discontinuous batch operations [13–17] and in continuous operations with interconnected carbonator–calciner fluidized beds [18–22]. Larger demonstration installations of 200 kW_{th} at Stuttgart University [23], 1MW_{th} at Darmstadt University [24] and 1.7 MW_{th} at “LaPereda” power plant near Oviedo [25], have started operation or are under construction.

A key issue for any of the previous processes is how fast reactions take place in the reactors as the gases have a limited residence time within the bed. Increasing the rate of reaction has different positive impacts on the reactor design and performance including compactness, effectiveness and efficiency. The aim of the present work is to apply the jetting fountain fluidized bed (JFFB) for carbonation process. Apply JFFB should increase the rate of carbonation reaction by enhancing the gas solid contact and the mass transfer within the bed. JFFB was proposed by Okasha and presented in many articles [26–33]. JFFB is characterized by excellent gas-solids contact due to creating a jet in the upper part of the bed, establishing a fountain in the freeboard and moderating bubbles size in the main bed. The present work presents a comparison between the JFFB and the conventional fluidized when applied to carbonation process. The influence of different operating conditions including bed temperature, air flow rate, jet air ratio and jet height have been tested.

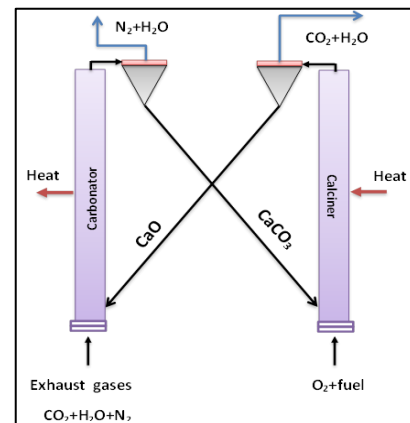


Fig.1 Scheme of the process for CO₂ captures using the lime carbonation–calcination loop.

2. EXPERIMENTAL WORK

2.1 Apparatus

The apparatus used in this work is a bubbling fluidized-bed combustor that has been designed to adopt the jetting-fountain configuration as shown in Fig.2. It has a fluidization column of 105 mm ID and 4000 mm height. The fluidization gases are distributed using a nozzle-type plate. A stainless steel pipe is used to introduce jet air. It proceeds from top to bottom. The pipe has two parts of different diameters. The first part has 2.75 m length and 19 mm diameter to reduce the pressure drop and to have good strength. The second part has 0.75 m length and 10 mm diameter. The later part is curved to allow the jet to issue vertically upward at the center of fluidization column. The tube is designed to be movable in vertical direction. Thus the jet orifice can be adjusted with respect to bed surface and distributor plate. On the other hand the

tube is avoided to move in radial direction as it fixed to the fluidization column at two different points.

Different electric heaters are used to heat the fed air. Three heaters with 5 kW are used to preheat the distributed air and two heaters with 3 kW are used to preheat the jet-air. An orifice meter is used to measure the flow rate of distributor-gas while the flow rate of jet-gas is metered using a Rota meter.

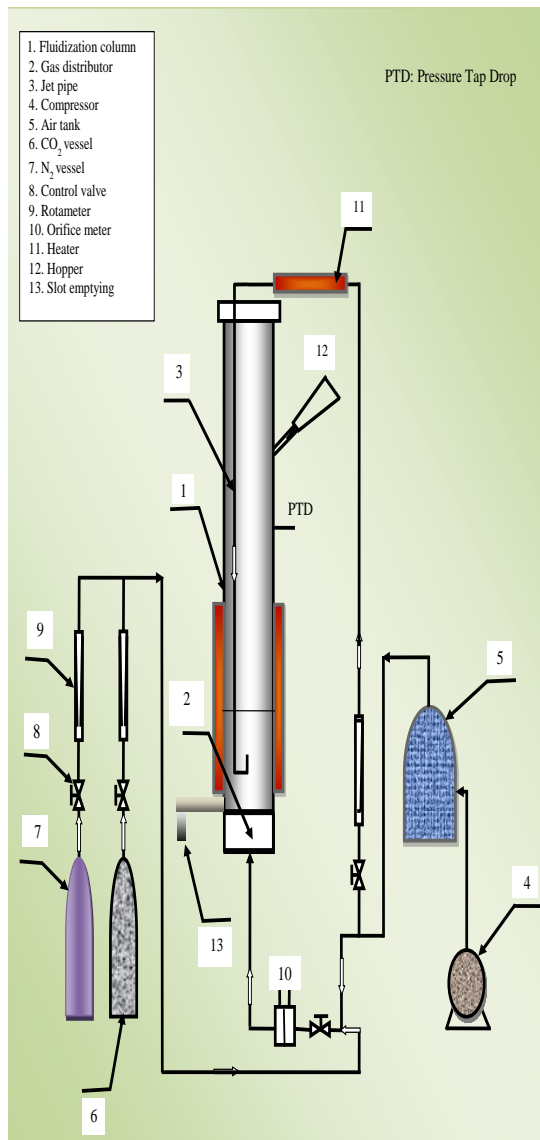


Fig.2 A schematic diagram of the experimental apparatus.

The column contains 13 portals for measuring probes. Two taps, PTD are used to measure the pressure drop from the plenum to the freeboard. Temperatures have been measured in the bed using a thermocouple of type K. The manufacturer's accuracy specification for the thermocouple is $\pm 0.4\%$ of the temperature. Measurement of gases concentrations has been carried out using IMR2800P gas analyzer. The gas analyzer is able to indicate the concentrations of O_2 , CO_2 , CO , SO_2 and NO_x . The measurement accuracy is $\pm 1\%$ for O_2 and $\pm 2\%$ for all other species.

Table 1 the chemical composition of the sorbent (wt. %)

SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	LOI
2.68	1.32	0.82	1.34	45.3	1.21	0.47	46.87

LOI: loss of ignition

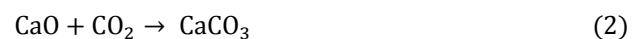
2.2 Materials, Technique and operating conditions

The sorbent used to capture CO_2 is a limestone comes from Jabal al-Tair Quarry in Minya. The chemical composition of the limestone is reported in Table 1. The limestone has 2560 kg/m^3 density and $0.425\text{-}0.600 \text{ mm}$ particle size. The corresponding fluidization velocities are 0.19 m/s and 0.1 m/s at 27°C and 650°C bed temperature, respectively.

The technique pursued in this work to perform an experimental test may be described in the following. The fluidization air is preheated before it is delivered through the distributor plate to the bed. Feeding of air continues until the bed temperature reaches the calcination temperature 850°C . During the calcination process the calcium carbonate converts to CaO by losing CO_2 according the reaction,



The bed is maintained at the later temperature until the CO_2 concentration indicates nearly zero by the gas analyzer. At this point the calcination of limestone particles are fully completed. The temperature controller is regulated to have the predesigned temperature of bed materials. When the bed temperature stabilizes at the test temperature the flow rates of nitrogen-carbon dioxide mixture ($15\% \text{ } CO_2$) are regulated to the pre-design values. At this point the carbonation process starts according to the following reaction,



The concentration of CO_2 in outlet gases is measured and recorded. The measurements continue until the concentration of CO_2 restores its value in inlet gases, i.e. 15% . To this end the current test is completed.

In this work the influences of operating conditions on the effectiveness of carbonation process have been explored. The bed temperatures of 550°C , 600°C , 650°C , 675°C and 700°C have been considered. Fluidization velocity has been tested at three cases 0.4 m/s , 0.8 m/s and 1.2 m/s . In jetting fountain configuration, a part of air is fed through the jet while the remaining part is fed through the distributor. Jet air ratio is varied from 0.2 to 0.6 . The orifice height of introducing jet air above the distributor plate is also an important parameter. It is varied from 8 cm to 16 cm . In all tests the static bed height is fixed to 15 cm .

3. RESULTS AND DISCUSSION

The experimental apparatus system allows the conventional operation of fluidized bed and the novel jetting-fountain configuration. In conventional operation, all air is delivered through the gas distributor. In the jetting-fountain configuration, on the other hand, only a part of air passes through the gas distributor. The

remaining part proceeds through the jet pipe to create a jetting-fountain zone. The ratio of jet-air mass rate to the total air mass rate is defined as jet air ratio, JR. Mathematically, it may be expressed as:

$$JR = \frac{\text{jet air mass rate}}{\text{jet air mass rate} + \text{distributor air mass rate}} \quad (3)$$

Several experimental runs have been carried out to study the carbonation process given in Eqn. (2). The performance of jetting-fountain configuration is also compared with the conventional operation.

In this work, some parameters are directly measured; however, some quantities are calculated. Based on the measured concentrations of carbon dioxide before, C_{CO2i} , and after, C_{CO2o} , the lime bed in addition to gas flow rate, \dot{m}_g , the mass rate of carbon dioxide that reacts with the lime particles, $\dot{m}_{CO2,p}$, may be determined by:

$$\dot{m}_{CO2,p} = \dot{m}_g * \left[C_{CO2i} - \frac{(1-C_{CO2i})}{(1-C_{CO2o})} C_{CO2o} \right] \quad (4)$$

The accumulated mass of CO₂ combined with limestone after time t, $m_{CO2,p}$, is calculated by the integration along the time,

$$m_{CO2,p} = \int_0^t \dot{m}_{CO2,p} dt \quad (5)$$

The CaO conversion, CR, is calculated as the ratio of moles converted into calcium carbonate to the moles of CaO originated in limestone,

$$CR = \frac{m_{CO2,p}/44}{M_L * P/56} \quad (6)$$

Where M_L is the mass of lime particles in the bed, P is the mass ratio of CaO in calcinated limestone (Purity). The number of mole of CO₂ substitutes that of CaO as they react in equal mole according to Eqn. 2. The efficiency of carbon dioxide capture, η is defined as the mass rate of carbon dioxide combined with limestone bed to the mass rate of carbon dioxide fed to the bed.

$$\eta = \dot{m}_{CO2,p} / (C_{CO2i} \dot{m}_g) \quad (7)$$

The average efficiency of CO₂ capture, η_{av} over the conversion time t is given by

$$\eta_{av} = m_{CO2,p} / (C_{CO2i} \dot{m}_g t) \quad (8)$$

Typical profiles of carbon dioxide concentration in outlet gases are shown in Fig.3. The carbon dioxide concentration appears considerably lower than its inlet concentration, in particular at the first period. In fact, CO₂ reacts with the calcinated limestone, CaO, particles; according to the reaction (2).

The efficiency of the reactor is shown in Fig.4 for the two configurations. The findings demonstrate that the jetting fountain configuration is more effective in carbon dioxide capture process. These results should be ascribed to the better hydrodynamic characteristics by jetting fountain configuration that enables higher contact efficiency between particles and gases. Actually, the

creation of a jet in the upper part of the bed, establishing a fountain of particles in the freeboard and moderating bubbles size in the main bed are very beneficial for contact efficiency heat increases the external mass transfer.

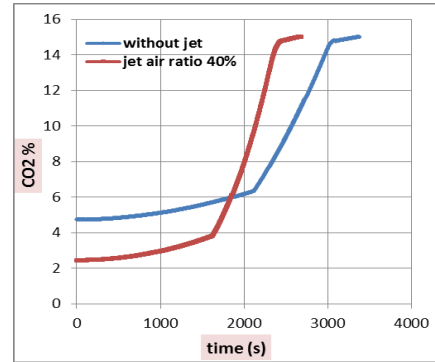


Fig.3 CO₂ concentration in the outlet gases

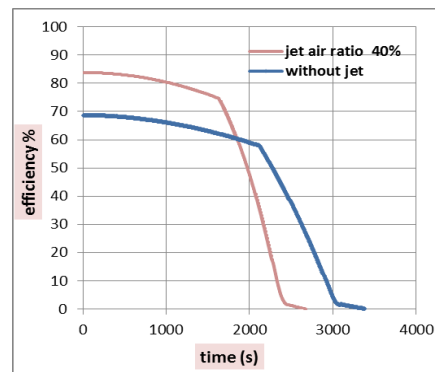


Fig.4 CO₂ capture efficiency as a function of time

Fig.5 illustrates the variation of CaO conversion with time. Near to 60% conversion completes during the first period which is important during continuous operation.

Figs.3-5 indicates that carbonation is characterized by a fast initial reaction rate followed by a transition to a very slow reaction rate. The rate of reaction of the fast stage depends on the surface area of the reacting particle where as in the slow stage is controlled by the diffusion. The reason of this fact is that while CaCO₃ (molar volume of 36.9 cm³/g) is being forming in the CaO (molar volume 16.9 cm³/g) particle, a layer of the former product is created and the diffusion through it becomes the controller of the product formation impeding CO₂ transport.

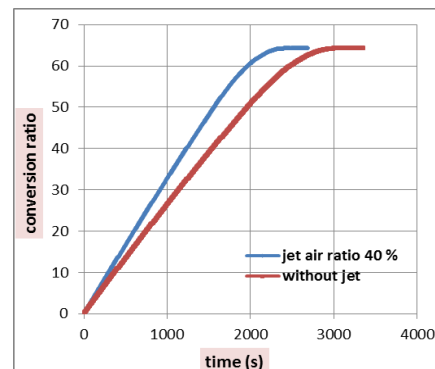


Fig.5 CaO conversion as a function of time

Effect of fluidization velocity

Effect of air velocity on carbonation process has been investigated. Three different velocities have been considered 0.4, 0.8 and 1.2 m/sec maintaining bed temperature at 650 °C for conventional operation. In Jetting fountain configuration 40% of gases are passed through the jet pipe, hence the corresponding fluidization velocities are 0.24, 0.48 and 0.72 m/s.

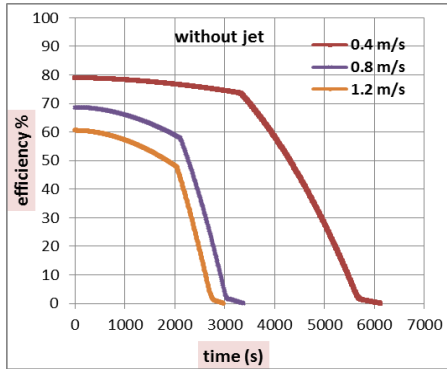


Fig.6 Effect of fluidization velocity on the capture efficiency at conventional operation.

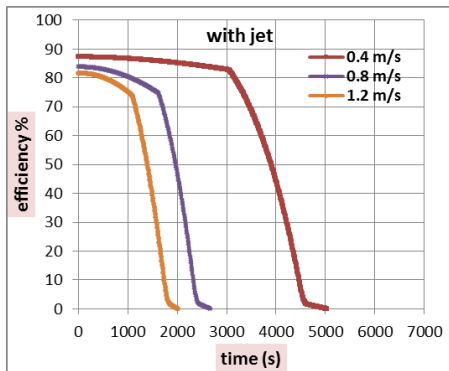


Fig.7 Effect of fluidization velocity on the capture efficiency applying JFFB.

The efficiency of carbon dioxide capture versus time at different fluidization velocities for the two considered configurations is shown Figs.6 and 7. The results indicate that at lower fluidization velocity the efficiency increases, in particular, during the first period as the gas residence time increases. However, the carbonation process needs longer time to attain the full conversion as the available quantity of carbon dioxide is lower. The average efficiency exhibits a notable decrease with the increase in the fluidization velocity as shown in Fig.8.

Fig.8 also gives a comparison between the conventional operation and jetting fountain operation with 40% jet air ratio. The presented results demonstrate that jetting fountain configuration performs better than conventional operation. The average capture efficiency increases from 58% 49.3% and 44.5% for conventional operation to 67.3%. 61.7% and 57.7% for jetting fountain configuration at fluidization velocity of 0.4, 0.8 and 1.2 m/s, respectively. It is obvious that jetting fountain configuration enhances the mass transfer between the bubble and emulsion phases and increases gas-particles contact due to creating a jet in the upper part of the bed, establishing a fountain in the freeboard and moderating bubbles size in the main bed.

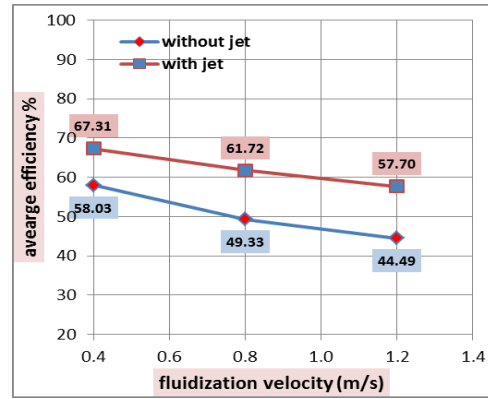


Fig.8 Effect of fluidization velocity on the average capture efficiency

Effect of bed temperature

The temperature has a great impact on the carbonation process. When the temperature rises, reaction kinetics improve, but also the equilibrium CO₂ partial pressure increases causing the reaction to slow down or change direction. To explore the effect of temperature on the CaO-CO₂ reaction, a series of CO₂ capture experiments were performed at five different temperatures of 550, 600, 650, 675 and 700 °C. Figs.9 and 10 present the efficiency of carbon dioxide capture in lime-bed versus time at various bed temperatures in the case of conventional operation and jetting fountain configuration, respectively. It is evident that increasing the bed temperature improves the capture as the efficiency increases and the time for full capture reduces. The optimum temperature is found around 675 °C. It appears worth to indicate that the carbonation reaction takes place when the partial pressure of the CO₂ in the flow stream is higher than the equilibrium partial pressure of CO₂ at a certain temperature. The equilibrium partial pressure of CO₂ increases with increasing temperatures causing a decrease in CO₂ partial pressure driving force which presents the initiation of the reverse reaction (calcination reaction) at a temperature around 700°C.

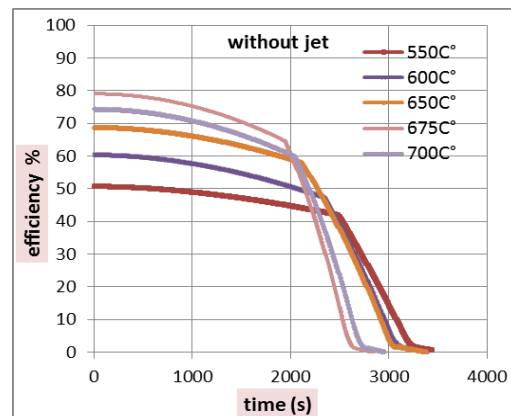


Fig.9 Effect of bed temperature on the capture efficiency for conventional operation.

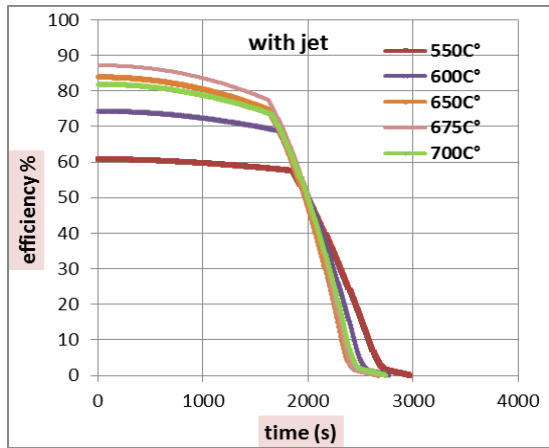


Fig.10 Effect of bed temperature on the capture efficiency for JFFB configuration

Fig. 11 illustrates the average efficiency of carbon dioxide capture versus bed temperature. The figure compares the findings of conventional operation with that of jetting fountain configuration with 40% jet air ratio. The findings indicate that jetting fountain configuration yields greater average efficiency for all considered temperature. It appears that applying jetting fountain configuration enhances the external mass transfer for the reason discussed above.

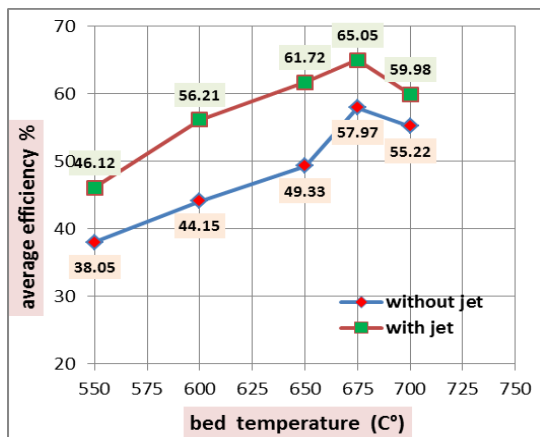


Fig.11 Effect of bed temperature on the average capture efficiency

Effect of jet air ratio

In jetting fountain configuration a part of gases is fed through the jet pipe to create a fountain of particles. Jet air ratio is used to express the fraction of air that is delivered via the jet pipe as discussed above. Fig. 12 shows the influence of jet air ratio on the capture efficiency of CO₂. It appears worth to indicate that the conventional operation of fluidized bed is the case at JR=0.

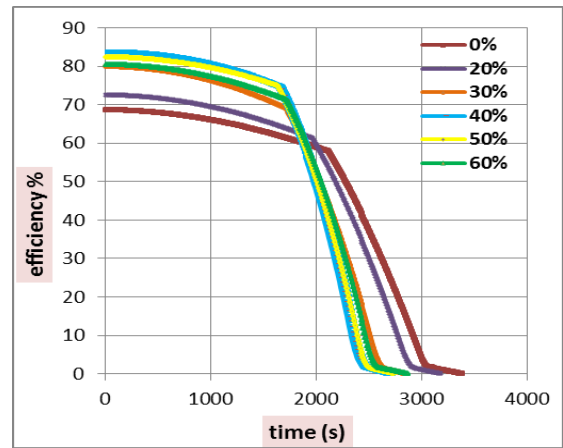


Fig.12 Effect jet air ratio on capture efficiency.

The data presented in Fig. 12 reveals that increasing JR improves the capture efficiency and reduces the time required for full conversion. However, the average efficiency of carbon dioxide capture attains an optimum at 40 % JR, and then decreases for a higher value as shown in Fig.13.

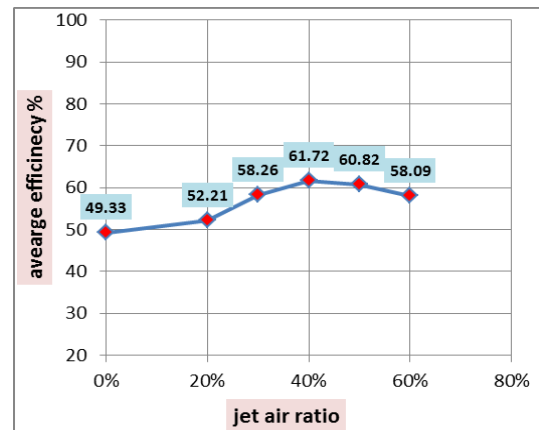


Fig.13 Effect of jet air ratio on average capture efficiency of CO₂.

It appears that the beneficial of increasing contact efficiency due to creating a fountain and decreasing bubbles sizes start be offset. At high jet air ratio, the jet velocity becomes very high that decreases the air contact time. Moreover, under this condition a large fraction of air bypasses the lower part of the bed without contact. However, lowering the jet orifice down in the bed could allow higher jet air ratio with greater contact efficiency.

Effect of jet orifice height

The jet pipe was designed to be movable in vertical direction to adjust the position of jet orifice with respect to the air distributor. The Effect of jet orifice height above the distributor on capture process has been investigated and the obtained results are plotted in Figs.14 and 15. Fig.14 illustrates the efficiency of carbon dioxide capture versus time at various jet orifice heights. The results demonstrate that the efficiency of capture improves with applying jetting fountain configuration.

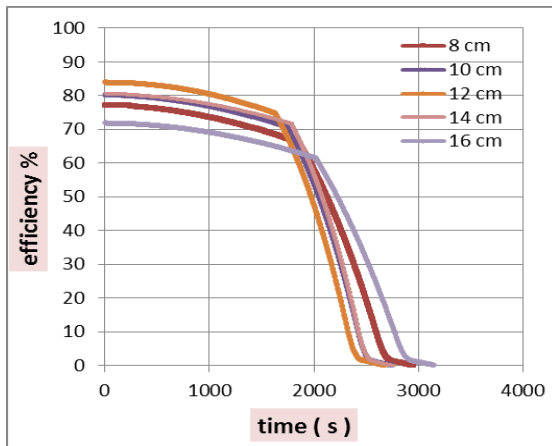


Fig.14 Effect of jet orifice height on the capture efficiency.

Fig.15 shows average efficiency of carbon dioxide capture versus jet orifice height. The trend line is not monotonic but rather exhibits a maximum point at 12 cm jet height. It is evident that there is a certain height for the jet orifice based on the applied jet velocity at which the performance of capture process attains its optimum. It is a matter of compromise for different competitive factors. The enhancement in contact efficiency due to creating a fountain of particles and the reduction in bubble size have positive impacts. On the other side, the lower part of bed that is bypassed by jet gases becomes greater which has a negative impact.

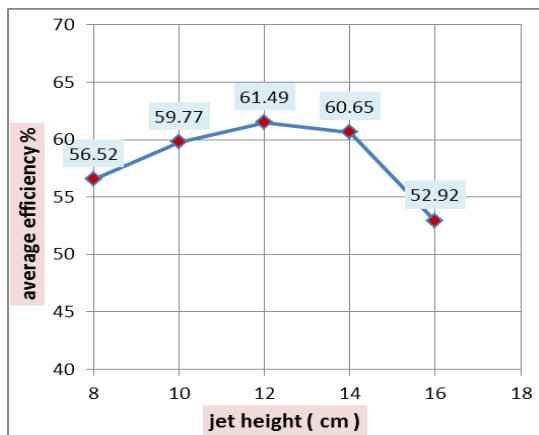


Fig.15 Effect of jet orifice height on the average capture efficiency.

4. CONCLUSIONS

An experimental study on carbonation process in the jetting fountain fluidized bed has been carried out. The experimental tests have been also performed in the conventional fluidized bed for comparison purpose. The effects of different parameters have been tested and evaluated. Based on the obtained results and the above analysis, the following conclusions can be drawn:

- Jetting fountain fluidized bed improves the carbonation process. The capture efficiency of carbon dioxide increases while the time required for complete CaO carbonation reduces. These good findings should be ascribed to the hydrodynamic characteristics of jetting fountain fluidized bed

configuration that promote much better gas-solids contact.

- The capture efficiency of CO₂ increases with the increase in bed temperature due to the improve in the reaction kinetics. This trend continues up to an optimum temperature, about 675 °C. However; the further increase in the bed temperature leads to a drop in the capture efficiency. The drop is mainly due the increase in equilibrium pressure that, in turn, causes a decrease in CO₂ partial pressure driving force which presents the initiation of the reverse reaction (calcination reaction) at a temperature around 700°C.
- Decreasing fluidization velocity increases the capture efficiency, mainly due to the longer gas residence time. However, the required time for full CaO carbonation becomes longer.
- The effect of jet air ratio on the capture efficiency of CO₂ is not monotonic but rather it has an optimum value. The optimum jet air ratio is found to be 40% under the considered conditions.
- The influence of the jet orifice height of on the capture efficiency of CO₂ has also an intermediate optimum value. The optimal jet height is found to be at 12 cm under the considered conditions.

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