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Advantages and Suitability of a Controlled Movable Packed Bed for the A3c Process

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ABSTRACT
Several carbon capture and storage technologies have been proven to have enormous potential in the reduction of anthropogenic CO_2 emissions; one of such novel technology is the advanced cryogenic carbon capture (A3C) process forwarded by the University of Chester. The A3C process proposes a concept that utilizes a moving bed of metallic beads as the heat transfer media and frost capture surface. The novel process if standardized would not only boast of a high CO_2 capture efficiency but also reduced energy consumption which is a known drawback to most cryogenic carbon capture processes. This paper identifies some challenges and limitations of the current A3C process and proffers some ways with which the A3C process can be improved upon.

Keywords: A3C Process, Cryogenic Carbon Capture

I. INTRODUCTION

Cryogenic carbon capture is a type of post-combustion carbon capture technique that involves freezing exhaust or flue gases below the sublimation temperature of carbon dioxide to generate frost, which may then be thawed and collected for storage or use (Grande, et al., 2017) (Tuinier, et al., 2010) (Cann, et al.), other carbon dioxide post-combustion capture processes include: "**scrubbing technology, membrane technology and adsorption** (Fasili, et al., 2019) (Flude, et al., 2017) (Tuinier, et al., 2010) (Singh, 2010)**,** these listed options will result in energy penalties caused by regeneration of CO_2 loaded absorbents or by recompression of flue gas streams at elevated pressures (Willson, et al., 2019) (Fasili, et al., 2019) (Bestard, et al., 2013) (Tuinier, et al., 2010), while post-combustion is considered the most practical technology in the short-term, the efficiency of the alternatives (pre-combustion and oxy-fuel combustion) may be higher in the long run (Fasili, et al., 2019) (Kvamsdal, et al., 2007). The advantages of cryogenic carbon capture are that; no large pressure difference is required, no chemical absorbent or adsorbent is required and high purity level of the CO_2 is obtained (Tuinier, 2011). In some cryogenic CO_2 capture process, CO_2 will transition directly from a gas to solid phase at atmospheric pressure (Hart, et al., 2009).

II. LITERATURE

BACKGROUND OF STUDY

Economies all over the world have one goal; that is to grow, this economic growth results in an increase in energy demand, and where the sources of this energy are the conventional but limited fossil fuels, the direct consequences are the effects it leaves on the environment as it relates to the carbon dioxide (CO_2) footprint (Fasili, et al., 2019) (Flude, et al., 2017) (Tan, et al., 2017). Carbon capture and sequestration (**CCS**) offers an option for attaining a large scale reduction in the emission of greenhouse gas (**GHG**) (Kang, et al., 2013) (Pawde, et al., 2013) (Jahangiri, et al., 2010). Leung stated that **"amongst the different approaches of CO_2 mitigation, CCS can reduce CO_2 emissions (typically between 85-90%) from large point emission sources, such as power production utilities, and energy intensive emitters, e.g. cement kiln plants"** (Leung, et al., 2014); by far CCS is considered as the most effective solution to the problem of climate change (Deng, et al., 2016). Several CO_2 capture technologies are currently in existence or under development; carbon capture technologies may be

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classified into three main types, **pre-combustion, post-combustion** and **oxy-fuel combustion** (Singh, 2010); in oxy-fuel processes fossil fuels are combusted using pure oxygen, circumventing dilution of CO_2 with nitrogen; in pre-combustion processes fossil fuels are gasified, CO_2 is subsequently captured and hydrogen is fed to the combustion chamber; in post-combustion, the processes are based on capturing CO_2 from the flue gases of conventional air fired power plants (Tuinier, et al., 2010) (Singh, 2010) (Isehunwa, et al., 2006).

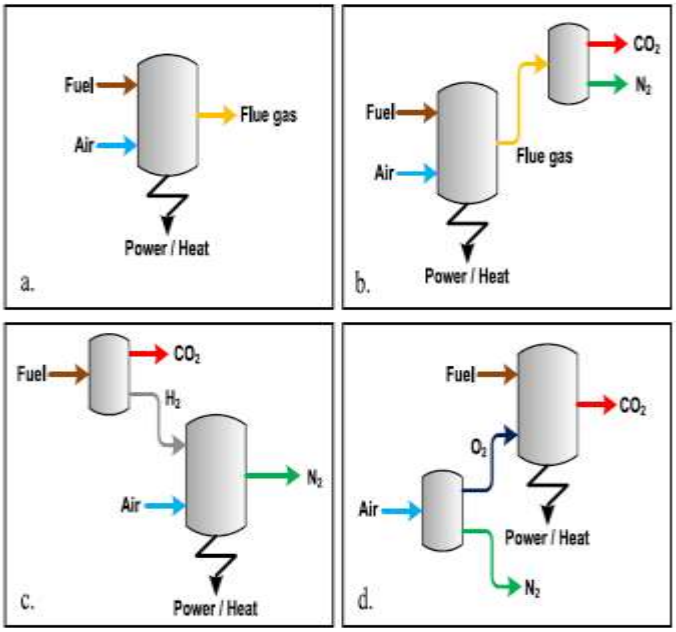

Figure 1: Pathways to CO_2 capture; a. Conventional Combustion without CO_2 capture. b. Post-Combustion capture. c. Pre-combustion Capture. d. Oxy-fuel Combustion capture. **(Tuinier, 2011)**

As no single technology option will adequately provide for all of the emission reduction required for stabilization; it is crucial that the selection of a capture technology is based on the following:

1. Concentration of CO_2 in the gas stream,
2. The pressure of the gas stream
3. The fuel type,
4. Adaptability of current technology with existing technology
5. The purity level of CO_2 obtained

Other economic factors important in the selection of a CCS alternative are the fuel price and the selling price of CO_2 (Blanch, et al., 2016). The desire to reduce the energy penalties and cost of CCS is a major driving force in existing and developing CO_2 capture and storage systems (Wilson, et al., 2019) (Singh, 2010). Low temperature CO_2 separation has been considered as one of the CCS technologies for extracting CO_2 from process gas streams (Wilson, et al., 2019) (Cann, et al.) (Bestard, et al., 2013) (Tuinier, 2011).

A typical low temperature CO_2 separation is based on a phase change, which allows the CO_2 gas to be separated from the flue gas in the form of a liquid or solid with high purity (Wilson, et al., 2019) (Bestard, et al., 2013). Unfortunately, low temperature CO_2 separation is regarded as a high-energy-consuming option owing to the cooling duty required (Wilson, et al., 2019) (Lychnos, et al., 2018) (Bestard, et al., 2013) (Tuinier, 2011).

BRIEF DESCRIPTION OF THE A3C PROCESS

Several cryogenic CO_2 capture technologies have been proposed by various research groups however two base cases on which the A3C was conceived were by **Coldic et al** and **Tuinier et al**; Coldic proposed a cryogenic system which used a refrigeration cascade to separate CO_2, the process however required that all the water content present were removed prior to the CO_2 capture stage; this was to prevent ice formation before CO_2 is captured as frost on the surface of a low temperature frosting evaporator (Willson, et al., 2019) (Cann, et al., 2018) (Clodic, et al., 2003), Tuinier overcame the problem of deposition of CO_2 layers and reduced efficiencies of Coldic's process by proposing an advanced configuration which used cryogenically cooled but dynamically operated packed beds; Tuinier's configuration separated the water and CO_2 on the packing surface without the need for additional drying stages; this was based on

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a cycle of bed cooling, CO_2 capture and a recovery step (Willson, et al., 2019) (Cann, et al., 2018) (Tuinier, 2011). Unfortunately the associated energy consumption of both **Coldic's** and **Tuinier's** CO_2 capture processes were quite high and necessitated the need for a low cost cold source (Willson, et al., 2019) (Cann, et al., 2018), this was confirmed by Bestard et al, where he stated that **"despite the advantages of low temperature CO_2 separation, it is still considered to be a highly-energy consuming alternative, mainly due to the cooling duty required"** (Bestard, et al., 2013).

A novel low temperature cryogenic CO_2 separation process known as the A3C (Advanced Cryogenic Carbon Capture) process currently under development at the University of Chester is aimed at reducing the associated energy consumption and cost of CO_2 capture (Wilson, et al., 2019) (Cann, et al.) (Lychnos, et al., 2018). The A3C process described by Wilson et al, Cann et al and Lychnos et al (**Figure 2**) overcame the limitations identified from **Coldic's** and **Tuinier's** processes simultaneously by using a moving bed of metallic beads as a heat transfer medium and frost capture surface (Willson, et al., 2019) (Cann, et al.) (Lychnos, et al., 2018); the A3C separation process was designed to have two stages, each having a circulating packed bed of metallic beads ranging in diameter from 1–5mm, the two stages consisted of a cooling-drying step, and a CO_2-separation step (**Error! Reference source not found.**) and was aimed at reducing the associated energy consumption and cost of CO_2 capture as well as the process equipment size while avoiding the adverse effects of heavy frost deposition and eliminating the need for multiple tower sized beds and associated switching losses (Willson, et al., 2019) (Cann, et al.) (Lychnos, et al., 2018).

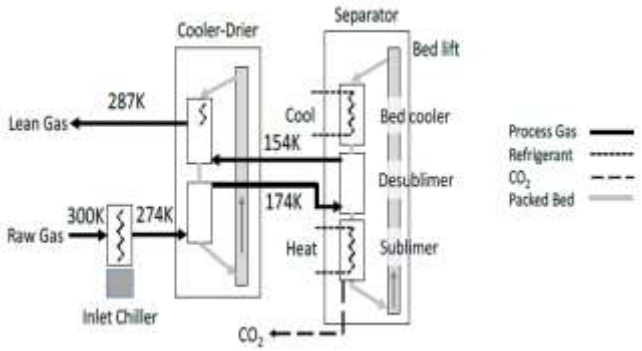

Figure 2: Outline of the two stages of the A3C CO2 separation process **(Lychnos, et al., 2018)**

CURRENT CHALLENGES AND LIMITATIONS OF THE PRESENT A3C PROCESS

Based on the paper by Willson (Willson, et al., 2019) the A3C process showed that it holds enormous prospects and potentials in significantly reducing the associated energy consumption and cost of CO_2 capture from large point emission sources (Willson, et al., 2019) (Cann, et al., 2018) (Lychnos, et al., 2018), however for this process to be successful several technical challenges or limitations must be resolved.

1. In the paper **"Evaluation of the performance and economic viability of a novel low temperature carbon capture process"** (Willson, et al., 2019), it stated that the proposed process uses a moving bed of cold particles with a counter current gas flow and that the CO_2 will be cooled to desublimate at a static frost front on the cold bed as it moves down the column.

Query 1: It is quite possible to have an efficient heat transfer for a counter current process; however the falling mode with which the metallic beads would be moving through the contact column (freely falling or controlled) is of concern; if freely falling then establishing the extent of frost front propagation and efficiency of the bed materials becomes a challenge, as it may be difficult to properly assess the bed materials at varying flow-rates of both bed materials and flue gases; however if a controlled movable bed of metallic beads were employed; the frost front propagation could be examined more accurately and provide insights on how to ensure that the targeted CO_2 does not reach end of the packed bed.

2. It was stated in the poster presentation **"Experimental exploration of carbon capture by frosting on a moving bed"** (Cann, et al.), that moving the bed in counter flow to the flue gas will result in the frost front remaining stationary avoiding the scenario of the capture column being

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saturated with frost and that the cold beads are fed into the cryogenic column to allow CO_2 form frost on the bed.

Query 2: Moving the bed material would prevent the scenario of capture column being saturated with frost or subjected to a freeze-thaw process afterwards; however the question of efficiency of the process still comes to play; because freely falling beads like any reaction needs a minimum residence time in the contact column to efficiently desublimate CO_2, as such to effectively utilize the cold temperature from the cold beads to desublimate CO_2, the contact column must be long enough to account for the minimum residence time, this may not be economical for small scale capture.

3. It was also stated in the poster presentation (Cann, et al.), that two conical hoppers were employed in series with the nature of flow in the hoppers angled at 0˚, 30˚ and 60˚, and a screw conveyor for re-circulation of the cold beads **Error! Reference source not found.** and **Error! Reference source not found.**.

Query 3: The cold beads in the hopper with 60˚ angles may provide a higher residence time compared to a hopper with 0˚ and 30˚ angles; however in the hopper with an angle of 60˚; chances are that there may be overtaking of bed materials and even some materials getting stuck for a freely falling bed mode because cold beads close to the walls of the contact column may experience among other things frictional drag.

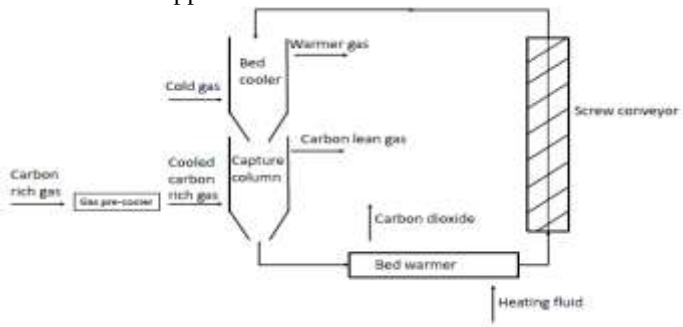

Figure 3: Diagram of 3rd generation rig **(Cann, et al.)**

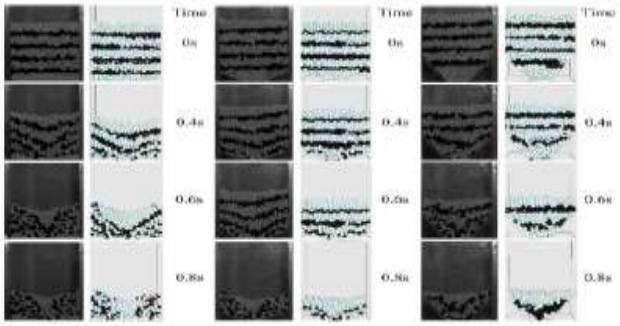

Figure 4: Nature of flow in a hopper with angled walls of 0˚, 30˚, 60˚ **(Cann, et al.)**

4. It was also stated in the poster presentation (Cann, et al.), that the energy requirement to re-cool a section of moving packed bed is not dependent on time;

$$Q = \frac{\dot{m} \int_{T_i}^{T_f} C_p \, dT}{(\Phi_{in} - \Phi_{out})} \dots\dots (1)$$

Query 4: As stated earlier in query 2, the freely falling beads like any reaction or process would need a minimum residence time in the contact column to efficiently capture H_2O in the cooler-drier section and to desublimate the CO_2 from the flue gas in the CO_2-separator; as such it would be necessary to review the rate of heat transfer equation to account for the time variable "t", as the rate of mass deposition and/or sublimation of CO_2 is among other variables a function of time.

III. DISCUSSION AND RECOMENDATION

With reference to the queries 1-4; controlling the falling mode of the bed material is of essence; as such a movable packed bed of battery type with re-circulation for cryogenic carbon capture be adopted considering the following reasons:

a. As it was stated in the paper **"Evaluation of the performance and economic viability of a novel low temperature carbon capture process"**

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that; **"the A3C CO_2 separation process is analogous to distillation and that the desublimation stage has a temperature gradient from lower at the top to higher at the bottom with conditions selected so that the vapor pressure of the desubliming bottom product is in equilibrium with its partial pressure in the gas phase, and also that the gases flow upwards in intimate contact with a falling stream of colder solid phase, carried on a reflux of cold inert bed material, it further stated that the reflux of bed material is cooled, as in an overhead condenser and the bottom product is recovered by sublimation with heating"** (Willson, et al., 2019). Based on the distillation analogy adopted for the A3C process; it would be necessary to establish a suitable feed point for the cooler-drier and desublimation stages respectively such that the movable packed beds could represent plates as in the typical distillation column, however the feed points should be carefully selected sufficiently below the ideal plate(s) which by definition would be where the water vapor condenses out of the flue gas for the cooler section, CO_2 desublimate out of the flue gas for the desublimer section and the CO_2/H_2O free gas is dried in the drier section (McCabe, et al., 2005). This comparison is expressed in heat, mass and continuity equations that are identical to those for a traditional distillation column, with enthalpies of sublimation replacing enthalpies of vaporization (Willson, et al., 2019) (Cann, et al., 2018).

b. Willson et al. also stated that; **"the A3C process would overcome some limitations of previous cryogenic systems by using a moving bed of metallic beads as the heat transfer media and frost capture surface while avoiding the adverse effects of heavy frost deposition, it further stated that the process eliminates the need for multiple tower sized beds and associated switching losses to offer a much reduced energy consumption and process equipment size"** (Willson, et al., 2019). The heat transfer as well as the frost capture capabilities of the moving bed of metallic beads, is one of the key performance indicator for the A3C process; as such adopting the proposed movable packed bed of battery type with which the falling mode is controlled, would not just allow for the phase change between the gaseous CO_2 and solid CO_2 but can provide a means by which the frost front propagation can be measured, it also allows for the packed bed to be represented either as a liquid or solid and the flow-rate adequately accounted for by the time a known mass of the packed bed enters and leaves the contact column, this proposed process further validates the analogy for mimicking the mixing of flue gas and the packed bed and also the separation of CO_2/H_2O from the packed bed without chemical changes to the packed bed, CO_2 or H_2O (Tuinier, et al., 2009) (McCabe, et al., 2005) (Coulson, et al., 1999).

PROPOSED A3C PROCESS

In line with the objectives of the A3C process, this proposed A3C process aims to validate the theory of using a moving bed of metallic beads as the heat transfer media and frost capture surface; while avoiding the adverse effects of heavy frost deposition and reduced efficiency as well as reducing the associated energy consumption of CO_2 capture. The proposed A3C rig would have two stages as in the original design **Figure 5** and **Figure 6**; however both sections shall consist of a full length insulated contact column and a shell and tube column; all with inbuilt gas locks. The contact columns and shell and tube columns shall be integrated with screw rods which would serve as bed lifts for the packed beds of battery type. The contact column in the cooler-drier stage **Figure 5** shall be responsible for capturing moisture (H_2O) from the flue gas, while the shell and tube in the cooler-drier stage **Figure 5** shall be responsible for drying the bed materials prior to entering the cooling section.

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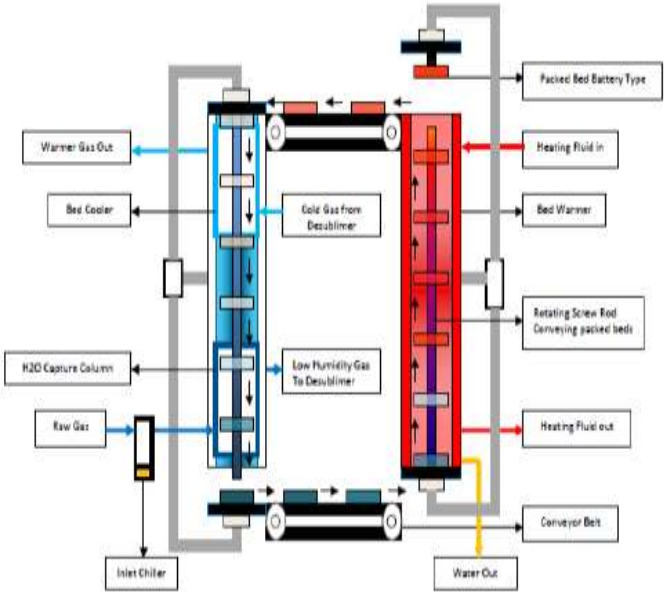

Figure 5: Proposed Cooler-Drier Stage for the A3C process

The contact column in the CO_2-separation stage **Figure 6**, would house the bed cooler 1, the desublimer and sublimer units, this contact column shall be responsible for the desublimation and subsequent capture of CO_2, the shell and tube in the CO_2-separation stage **Figure 6** shall be responsible for re-cycling of the packed beds, however this shell and tube in the CO_2-separation stage can double as a pre-cooling unit for the packed beds in the CO_2-separation stage by utilizing a pinch technology system. As with the original A3C rig, the proposed rig shall employ a refrigeration system that uses conventional refrigeration components for the initial design, however the design is targeted at reducing the cooling duty required without compromising on the efficiency of the CO_2 separation.

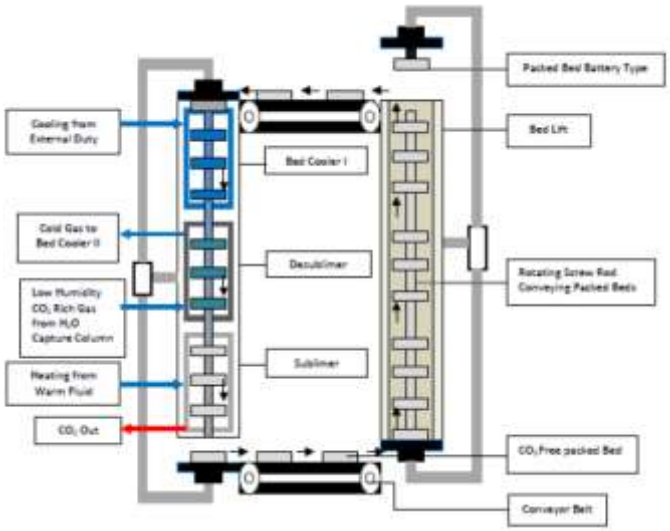

Figure 6: Proposed CO2-Separation stage for the A3C process

ADVANTAGES OF THE PROPOSED A3C PROCESS

Below are advantages as to why the proposed movable bed of metallic beads should be adopted as a suitable method for the A3C process.

1. The proposed movable packed bed of battery type allows for phase change between gaseous CO_2 and solid CO_2 and vice versa; this satisfies one of the analogy made for the A3C process (Wilson, et al., 2019) (Cann, et al.) (Lychnos, et al., 2018).
2. The proposed movable packed bed allows for the process to be represented as a liquid or solid and the flow-rates adequately accounted for by

the time a known mass of the packed bed enters and leaves the contact column (McCabe, et al., 2005) (Coulson, et al., 1999)

3. The proposed process satisfies the analogy for mimicking the mixing of flue gas and the packed bed and also the separation of CO_2/H_2O from the packed bed without chemical changes to the packed bed, CO_2 or H_2O (Tuinier, 2011).
4. The proposed process satisfies the analogy to a conventional distillation column reflecting individual heat, mass and continuity equations (Bird, et al., 2012) (McCabe, et al., 2005) (Coulson, et al., 1999)
5. The proposed process allows for heat exchange between the flue gas and the packed bed (Coulson, et al., 2002) (Coulson, et al., 1999).
6. The proposed process answers the problem of accurate and constant bed to diameter ratio across the contact column as well as providing a means by which the mass deposition rate versus time may be evaluated (Coulson, et al., 2002).
7. The proposed process allows for accurate estimation of the frost front propagation (Tuinier, 2011) (Tuinier, et al., 2010).
8. Based on the distillation analogy adopted for the A3C process, the proposed process allows for accurate selection of feed points for the cooler-drier and desublimation stages (McCabe, et al., 2005) (Coulson, et al., 1999).
9. The proposed process allows for the movable packed bed to mimic the plates as in a typical distillation column (McCabe, et al., 2005) (Coulson, et al., 2002) (Coulson, et al., 1999).
10. Finally, the process allows for the desublimation to be carried out at atmospheric pressure and at low temperature (Wilson, et al., 2019) (Cann, et al., 2018) (Lychnos, et al., 2018).

IV. CONCLUSION

As supporting resources are still limited on this topic, it is assumed that the controlled movable packed bed of battery type should better explain the effects of residence time as it relates to the mass deposition rate of CO_2 on the bed materials, It is also assumed that the controlled movable packed bed of battery type would clarify the extent of frost front propagation formed on the cold beads and proffer a relation between the frost front propagation and the rate of fall of bed materials, and although operating at atmospheric pressure it would be necessary to regulate the flue gas flow rates using flow regulators and gas-locks to ensure the gases entering the cooler section do not propagate towards the drier section so also the CO_2 gas do not propagate toward the top of the contact-column in the CO_2-separation stage, a small gas suction pump could be introduced for any case of rouge gas that may escape the gas-locks, however this would slightly increase the overall duty on the process and ultimately cost of the process.

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