



The pilot dual-reflux vacuum pressure swing adsorption unit for CO₂ capture from flue gas



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ABSTRACT

The results of carbon dioxide separation from real flue gas using the dual-reflux vacuum pressure swing adsorption (DR-VPESA) technology have been presented. The mobile pilot CO₂ separation plant consists of two containers: a processing container and a control container. The processing container includes a flue gas treatment section (which comprises dust removal, cooling, deSO_x, deNO_x, gas drying and compression units), as well as a CO₂ adsorptive separation unit. The adsorption unit is made up of four fixed-bed columns, each divided into two sections, operating in a dual-reflux vacuum pressure swing adsorption mode. Two kinds of activated carbon in the total amount of 540 kg were used as the beds filling. The results from the 8-step reference process with no partition of the adsorbers into the upper and lower sections (as one-step separation process with partial product recirculation), and from the 9-step DR-VPESA process (as two-step separation process) with a division of the adsorbers are presented. In the measurement campaign three different feed gas flow rates and five different times of adsorption step were selected to assess their influence on separation results. The results show the optimal parameters of 100 Nm³/h for feed gas flow rate and 300 sec for adsorption step time for both mentioned processes. The 87.5% of purity, 44.6% of recovery, 11.4 kg/(m³ h) of productivity and 978 kWh/Mg_{CO2} of energy demand for gas compression and vacuum evacuation in the case of optimal parameters of 9-step DR-VPESA technology were obtained. Additionally the results from measurement campaign were compared with those reported by other researchers.

1. Introduction

Due to the growing electricity demand in the future and the plan to reduce the energy sector CO₂ emission to zero between the years 2040 and 2060 (to limit the temperature increase by 1.5 °C) [1], some efforts have been specified by the IEA to achieve this goal, which include: the reduction of the electricity demand, the reduction of the output from unabated fossil-fuel power plants, as well as the large-scale deployment of all low-carbon power generation technologies (renewable and nuclear power engineering, and fossil-fuel power plants fitted with CCS). The reduction of the greenhouse effect is possible through the agreements concluded at the international level and then approved and put into effect at the governmental level. The EU greenhouse gas emission reduction regulations [2,3] and the roadmap including perspectives for

a low-carbon economy [4] can be used as the examples. These challenges are being coped with by science, which seeks to find the best solution in the technical, environmental and economical aspects.

Depending on the scenario drawn up by the IEA [1], coal as a fuel for electricity generation will still be present in 2040 (its share of world's total electricity generation will be 27.6% in the New Policies Scenario and 7.4% in the 450 Scenario). At the same time it is assumed that the 70% of the coal-fired power generation in the 450 Scenario is dependent on plants equipped with CCS [1], therefore future investigations into technologies of carbon dioxide capture from flue gas are justified.

A lot of laboratory (absorption [5–7], adsorption [8–10], membranes [11–13], cryogenics [14]), pilot [15–28] and numerical simulation [29–33] studies, as well as technology [34–37] and gas treatment

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Nomenclature**Abbreviations**

C	average carbon dioxide concentration in the CO ₂ -rich gas (purity), [%]
C_F	carbon dioxide concentration in the feed gas, [%]
C_L	carbon dioxide concentration in the CO ₂ -lean gas, [%]
C_R	carbon dioxide concentration in the CO ₂ -rich gas, [%]
F	average carbon dioxide concentration in the feed gas (after gas treatment), [%]
p	pressure, [kPa abs]
R	average carbon dioxide recovery, [%]
t	time, [s]
t_A	time of adsorption step, [s]

t_1	initial time of the analyzed cycle parameters, [s]
t_2	final time of the analyzed cycle parameters, [s]
V_F	feed gas flow rate, [Nm ³ /h]
V_R	CO ₂ -rich gas flow rate, [Nm ³ /h]
V_L	CO ₂ -lean gas flow rate, [Nm ³ /h]
CCS	carbon capture and storage
CFB	circulating fluidized-bed
CSS	cyclic steady state
DR-VPSA	dual-reflux vacuum pressure swing adsorption
IEA	International Energy Agency
PTSA	pressure-temperature swing adsorption
PSA	pressure swing adsorption
TPD	tons per day
VPSA	vacuum-pressure swing adsorption
VSA	vacuum swing adsorption

[38,39] analysis have been carried out so far. Among the above-mentioned methods, attention is paid to the adsorption method, which is characterized by the following advantages [40]: an availability higher than 99%, flexibility, fully automated operation, high product purities and high recovery rates. However, few adsorption tests have been carried out on a pilot scale in industrial conditions on installations with a capacity of over 30 Nm³/h. Eight of them described in the literature were tested in Japan [17–19,24], Korea [20], Australia [21] and China [22,23,25].

The first pilot plant unit [17,18] with a capacity of 1000 Nm³/h separated CO₂ from exhaust gases (ca. 4.5 TPD) in two adsorption stages at the Yokosuka Thermal Power Station in Japan. Boiler flue gas was dried using alumina prior to the carbon dioxide separation process on the Ca-X zeolite. After the first adsorption stage (PTSA), the concentration of CO₂ was in the range of 40 ÷ 50%, and after the second stage (PSA), the purity of the product was equal to 99%. The authors [17,18] found that the addition of this temperature swing in the first step (using a steam gas heater) reduced the electric power consumption by about 11%, while the carbon dioxide removal efficiency greatly depended on the CO₂ concentration in the flue gases [17].

The second two-stage PSA installation [20] with a capacity of 110 Nm³/h was built to separate carbon dioxide from flue gas coming from a residential boiler in Korea. The flue gas containing 10.5% of CO₂ before the separation process was cooled down, dedusted, partially deSO_x in a scrubber and then dried in a two-bed adsorption unit. The tests revealed that two beds were sufficient in each stage of the separation process to achieve the 99% purity of the product with a 80% recovery.

The third VPSA pilot plant investigations in Japan [19] were carried out using two different types of adsorbents in a single-bed (zeolite NaX) and a dual-bed (NaX and Na-A zeolites in varying proportions) configurations. A four-bed eight-step process was carried out. The installation separated boiler exhaust gas containing 13% CO₂. The application of the optimum zeolite mixture resulted in a higher CO₂ concentration (> 50%) without a decrease in recovery (> 80%), depending on the feed gas flow rate used.

The next pilot plant unit located at the International Power's Hazelwood Power Plant in Australia [21] realized a one-stage VSA process. The installation was able to separate approx. 1 TPD of carbon dioxide from flue gas containing 10 ÷ 15% CO₂. Multiple-layered adsorbents for the removal of water, acid gas (SO_x/NO_x) and CO₂ gas were used in the tests. The tests carried out on a three-bed installation operating in a 6-step simple cycle configuration and a not fully optimized process resulted in a purity of approx. 71% and a recovery of approx. 60%.

Other complex pilot plant tests were carried out in China on a single-stage VPSA unit filled with zeolite 5A [22] and then zeolite 13X [23]. In subsequent tests, the installation was equipped with a second

CO₂ purification step on activated carbon [25].

The tests of carbon dioxide separation on zeolite 5A [22] were carried out in a three-bed single-stage VPSA unit. Dehumidified flue gas containing 15.0% CO₂, 76.5% N₂ and 8.5% O₂ at feed flow rate ranging from 32 to 46 Nm³/h was subjected to separation in a seven-step process. A CO₂ concentration of 71–81% was obtained with a recovery of 86–91%. Other tests [23], in which zeolite 13X was used in an eight-step CO₂ separation process, yielded similar performance for carbon dioxide separation (85–95%) as well as purity (73–82%), despite the fact that zeolite 13X has a higher CO₂ equilibrium capacity compared to zeolite 5A. Process parameters, such as flue gas flow rate (32.9–45.9 Nm³/h), CO₂ concentration (15.5–16.5%) and the level of gas pretreatment (dehumidification to a relative humidity value less than 0.5% and the presence of traces of SO₂ and NO_x), were similar.

Slightly different tests of CO₂ capture from exhaust gas (coming from coal combustion) on a pilot scale based on amine-impregnated porous material [24] were conducted by Kawasaki Heavy Industries in a fixed-bed system (adapted to a small-scale plant) and in a moving-bed system (adapted to a large-scale plant). The adsorbent regeneration agent was low-temperature steam (60 °C) obtained from waste heat recovered from a gas engine. The test conducted in a moving-bed system for a feed gas flow rate of 530 Nm³/h and a CO₂ concentration of 13% resulted in the capture of about 3 TPD of CO₂ with a purity of 95% or more, which yielded a recovery rate of above 90%.

By comparing the test results [17–24] one can easily find that only the two-stage separation process or the one-stage separation based on the amine-impregnated porous material regenerated with steam [24] makes it possible to obtain the high purity with a relatively high CO₂ recovery, while allowing the expected medium purity of the product to be achieved with the aim of its further sequestration (> 95%) [41]. This was confirmed in subsequent investigations [25]. The gas pretreated as in the previous investigations [22,23] was concentrated to 70–80% CO₂ on zeolite 13X in the first three-bed VPSA unit, and then further enriched on activated carbon in the second VPSA unit. The total CO₂ recovery amounted to 90.2% with a purity of 95.6% in the product, for a feed gas flow rate of 35 Nm³/h.

The presented results of pilot plant studies, the conclusions obtained by Thakur et al. [42] and Grande and Bloom [43] from numerical simulation studies of CO₂ separation using the duplex (called also dual) pressure swing adsorption method as well as further investigations that have been performed by Shen et al. [44], covering experimental and simulation study based on small two-bed laboratory DR-PSA installation, set the direction for the design and construction of the integrated two-stage four-bed adsorption pilot plant for flue gas CO₂ separation operating in the dual-reflux vacuum-pressure swing adsorption technology. Our DR-VPSA installation characterizes of a compact construction in reference to typical two installations connected in series and is supposed to show the effectiveness as well as the readiness of

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