

THE CO₂ CAPTURE SYSTEM WITH A SWING TEMPERATURE MOVING BED

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Abstract: The reduction in CO₂ emissions is now a very popular topic. According to the International Energy Agency, CO₂ emitted in 2021 was 6% more than that emitted in 2020. Carbon capture and storage (CCS) is gaining popularity as a possible solution to climate change. Experts estimate that industry and power plants will be responsible for 19% of total CO₂ emissions by 2050. This paper presents the design of a semi-industrial-scale system for CO₂ capture based on the moving bed temperature swing adsorption technology. According to the results of laboratory tests conducted by the SINTEF industry, this technology demonstrates high capture efficiency (>85%). The CO₂ capture medium involved in adsorption is activated carbon passing through individual sections (cooling, heating, adsorption), where CO₂ is bonded and then released. The heat and mass transfer processes are realised on the developed stand. The heat exchangers use steam and water as the heating/cooling medium. The paper reviews the existing solutions and describes the developed in-house design of heat exchangers that will ensure heat transfer conditions being a trade-off between economic and efficiency-related issues of the CO₂ capture process. The designed test stand will be installed in a Polish power plant and is expected to meet the method energy intensity target, set at ≤ 2.7 MJ/kg CO₂, with a capture efficiency exceeding 85%. The aim of the work was to develop and solve technical problems that would lead to the construction of a CO₂ capture station with parameters mentioned above. This stand uses an innovative method where CO₂ is captured by contacting the fluid (gases) with solid particles. The heat exchange associated with the heating and cooling of the adsorbent had to be solved. For this purpose, heat exchangers were designed with high thermal efficiency and to prevent the formation of mounds.

Key words: heat and mass transfer, CO₂ capture, moving bed temperature swing adsorption, test stand

1. INTRODUCTION

Reducing CO₂ emissions in line with the European Green Deal has become a very important topic. According to a report by the International Energy Agency, as much as 36.3 billion tons of CO₂ were produced in 2021, which is more than 6% than that produced the year before – which is the highest level ever [1]. The same agency estimates that the share of CO₂ emissions from industry and power plants will increase from 3% in 2020 to 19% in 2050 [2,3]. For this reason, the carbon capture and storage (CCS) technology has become one of the most promising and necessary solutions to combat climate change [4]. Carbon capture (CC) is a solution which makes it possible to continue the use of fossil fuels, minimising at the same time the amount of CO₂ released into the atmosphere, thus mitigating global climate changes. Great efforts are being made around the world to develop a highly efficient CO₂ capture technology consuming little energy.

The CO₂ capture technology mainly includes three methods: pre-combustion capture, oxy-combustion and post-combustion capture [5]. The first method involves avoiding the mixing of CO₂ with air, through conversion processes of carbon contained in the fuel to make it possible to remove CO₂ from the gases.

Such processes include coal gasification or reforming of oil and gas fuels.

Oxy-combustion involves the use of air significantly enriched

with oxygen. Such air is obtained by a prior process which removes nitrogen. The technology makes use of recirculation of a portion of combustion gases consisting mainly of CO₂ and O₂ to lower the furnace temperature and increase CO₂ concentration.

Post-combustion capture can be implemented, for example, by absorption, cryogenic separation, membrane process or adsorption [6,7]. Depending on the applied method, the process requires energy necessary for compression, cooling and extraction for the solvent regeneration and needs to ensure a highly efficient desulphurisation process. However, the main advantage of the post-combustion capture method is that it does not require major modifications to existing power plants [8].

The absorption process involves passing cooled and pre-treated combustion gases through an absorption column, where a significant portion of the CO₂ is absorbed through contact with a solvent. The CO₂-rich solvent then passes through a desorber responsible for releasing carbon dioxide, which is compressed and purified in the next step.

Membrane separation is based on the occurrence of differences in the physicochemical and chemical interactions between the membrane material and the components of the gas mixture, which is related to the difference in the rate of permeation of the gas components through the membrane. The membrane is responsible for dividing combustion gases into the permeating stream and the stopped stream.

The cryogenic method involves compression and cooling of the gas to a temperature of about -56°C , followed by separation of CO₂ in the liquid form. The selectivity of the cryogenic fractionation process results from the different conditions of condensation of individual components of combustion gases.

The last post-combustion capture method – adsorption – is based on physical attraction of gas and solids. The following materials with a large specific surface area are used as a CO₂ separation medium in the adsorption process: zeolites, activated carbon, aluminium oxide or silica and alumina gel. Three kinds of CO₂ adsorption are used for separation:

- pressure swing adsorption (PSA) – the adsorbent is regenerated through a reduction in pressure,
- temperature swing adsorption (TSA) – the adsorbent is regenerated through a rise in temperature,
- electric swing adsorption (ESA) – regeneration proceeds by passing a low-voltage electric current through the adsorbent.

Currently, adsorption is widely recognised as an attractive method for small and medium-sized CO₂ emitters due to its low energy consumption and low operating cost [9,10]. The moving bed temperature swing adsorption (MBTSA) is a promising technology for capturing CO₂ from combustion gases. In an MBTSA unit, selective adsorbing material circulates between the low-temperature stage, where it removes CO₂ from combustion gases, and the zone with a higher temperature, where it desorbs CO₂ with greater purity. Given these advantages, various solid adsorbents are used to capture CO₂, including zeolites, metal-organic frameworks (MOFs), silica materials, carbon materials and functionalised adsorbents [11,12]. Studies have shown that activated carbon can successfully capture CO₂ because it has a large surface area per unit volume, as well as a large surface area of submicroscopic pores in which adsorption of impurities occurs. Moreover, activated carbon is stable in acidic and alkaline conditions. The activated carbon discussed here, with the trade name G-BAC, was manufactured by Kureha and has a spherical shape. The shape, as well as the particle size, of the activated carbon is significant here because of its adsorption properties. In an experiment conducted by a team from SINTEF [34], this activated carbon showed a high, up to 95%, capture rate. The experiment was conducted for synthetic flue gases with a CO₂ content of 3.5 vol%, which is much lower than that found in flue gases from coal-fired power plants. Simulations were also carried out in gProms software for the composition of the flue gas as in a coal-fired power station, as to ensure that the CO₂ capture efficiency was sufficient. On this basis, an activated carbon adsorbent-based CO₂ capture system was designed. Activated carbon is also cost-effective due to the possibility of regeneration and is therefore suitable for the removal of organic compounds [13]. However, the adsorption bed is a costly and energy-intensive part of the MBTSA system; so, an appropriate design of the heat exchanger responsible for cooling, heating and adsorption of the working medium is of key importance to improving the efficiency of the entire process. Creating gas-granulate or liquid-granulate heat exchangers is a major challenge. Efficient heat transfer is affected by the geometry of the exchanger (shape and arrangement of tubes, hydraulic diameter), operating conditions (fluid flow velocity, the heating/cooling fluid temperature) and the adsorbent specific properties (shape and size of particles, density).

There are three mechanisms of the heat transfer between freely falling granules and the heat exchanger immersed surfaces

– particle convection, gas convection and radiation. In most dense gas-solid fluidised beds, the mixing of solids is the main cause of the convective heat transfer by particles [14].

Several factors are responsible for the convective heat transfer to the flowing sorbent: the contact surface area between the sorbent and the hot surfaces, the sorbent residence time, the flow pattern and also the mixing of the particles. For these reasons, determining the exchanger performance and, in particular, estimating the coefficient of the heat transfer from the sorbent to the medium flowing in the exchanger tubes is a challenge for any designer. In this paper, this coefficient is estimated, and the exchangers intended for the CO₂ capture system are designed.

2. DESIGNING AND TESTING HEAT EXCHANGERS

Researchers all over the world have been making efforts to design more efficient heat exchangers for many years. In this part of the paper, attention will be focused on the literature analysis in terms of designing and testing heat exchangers operating with a fluidised bed.

Wang et al. [15] proposed a transient model for the analysis of cryogenic desublimation of CO₂ in a gas mixture. The model includes a counter-current tube-in-tube heat exchanger with three control volumes, a nitrogen (or helium) cooling medium, a wall with a solid CO₂ layer and a gas mixture. The deposition distribution, the capture rate and the energy consumption for the CO₂ desublimation process are analysed in different operating conditions. The Nusselt number in the simulations is calculated using equations derived from the model function. Bahrehmand and Bahrami [16] described a 2D analytical model for the design of sorbent-bed heat exchangers of sorption cooling systems. The model covered all relevant thermophysical and sorptive properties, the heat exchanger geometry and operating conditions. Contact thermal resistance at the phase interface in sorption beds was also taken into account. The convective heat transfer coefficient adopted in the computations was calculated based on the Gnielinski correlation. More information on heat exchanger modelling can be found in the paper by Golparvar et al. [17], where a detailed three-dimensional non-equilibrium model was developed to study the effects of the heat and mass transfer in annular and longitudinal finned tube adsorber beds filled with zeolite 13X particles. The convective heat transfer coefficient in this paper was calculated for a fully developed flow using the Dittus-Boelter correlation for smooth tubes. It was observed that larger temperature gradients occur in an adsorption bed with a bigger fin spacing, leading to non-uniform adsorption and desorption processes in the entire heat exchanger. The Dittus-Boelter correlation for smooth tubes was also used to calculate the heat transfer coefficient by Zhang [18]. The paper presents a mathematical model of the conjugate heat and mass transfer in an adsorber in an intermittent adsorption cooling system. The model includes four submodels: the heat transfer in the heating/cooling fluids, the heat transfer in the metal tube, the heat transfer in the fins and the heat and mass transfer in the adsorbent. The model of the heat and mass transfer in the adsorbent (zeolite 13X) is a three-dimensional non-equilibrium model that takes account of the mass transfer both internal and external resistance in the adsorbent. The paper by Clause et al. [19] compares an adsorber cooled indirectly in a TSA system and other adsorbents

classically investigated in TSA systems. A numerical model is presented and then validated using experimental testing results. Based on testing, the advantages of indirectly cooled adsorbers are demonstrated. The paper also describes the possibility of omitting the cooling stage in TSA cycles.

Hofer et al. [20] investigated the heat transfer between bubbling fluidised beds and the immersed surface of heat exchangers with respect to continuous TSA processes used for post-combustion CO₂ capture. Heat transfer coefficients for gas and particle properties were calculated using the models proposed by Natusch and Molerus. Fluidised beds were also investigated by Pirklbauer et al. [21], who studied different process configurations of a multi-stage fluidised bed for continuous temperature swing adsorption. The aim of the studies was to determine the optimum energy of the process. When CO₂ is used as the regeneration medium, the total energy demand of the TSA process increases as compared with a multi-stage TSA system that operates with steam as the separation medium. This research provides clear information on what configurations of continuous TSA processes of CO₂ capture are the most energy efficient. The topic of moving bed adsorption (MBA) was also taken up by Kim et al. [22]. The authors described this process with heat integration. The adsorbent particles and the combustion gases flow in opposite directions in the adsorption bed. In this case, a scheme is also proposed in which the thermal energy removed from the adsorption bed is used in the desorption bed, which increases the presented method efficiency. Post-combustion CO₂ capture was also studied by Schöny et al. [23]. In this case, steam is used as the removing agent on the desorber side. The optimal number of stages on the adsorber and desorber sides is investigated to achieve high energy efficiency of the separation process. It is concluded that the larger the heat transfer surfaces of heat exchangers the higher the CO₂ capture efficiency.

Mitra et al. [24] described typical adsorption heat exchangers, i.e. tube-and-fin assemblies with adsorbents placed between the heat exchange surfaces. The research was conducted for activated carbon as the adsorbent. The results show that the optimal geometry of the heat exchanger depends on the size of the adsorbent particle. Exchangers were also investigated by Hofer et al. [25]. The authors tested devices consisting of a single tube and multiple tubes operating in post-combustion CC processes. The authors developed design requirements for TSA stages based on the heat transfer coefficients achievable in bubbling fluidised beds. The testing results show that the heat transfer coefficients obtained experimentally for single tubes are in good agreement with the modelled values. The results obtained for tube banks could be applied for the heat transfer between bubbling fluidised beds and immersed surfaces of the heat exchanger. Sharafian et al. [26] carried out experimental testing of the effect of fin spacing on the temperature distribution of two adsorption beds using finned tubes with different fin spacing. The works included comparing the adsorption bed/adsorbent mass ratio and the temperature differences between the fins depending on the fin spacing. The researchers concluded that the optimal fin spacing of 6 mm was the best fit design for a finned tube adsorber filled with 2–4 mm silica gel particles.

It follows from the presented literature review that there are various designs of exchangers intended for CO₂ capture plants. Most of them are exchangers with an increased heat transfer surface area (finned tubes). This paper presents a CO₂ capture system based on the MBTSA technology which does not work as

a fluidised bed. In the presented system, the adsorbent is transported to individual sections where it is heated or cooled. In addition to a high coefficient of the heat transfer from the granulate to the surface of the tubes, the exchanger must ensure that the adsorbent flows through freely and is prevented from settling on the tubes. This rules out the use of fins, as they would create spaces for the adsorbent to accumulate, which would render the exchanger inefficient. The developed exchangers must reconcile the adsorbent transport with a high coefficient of the heat transfer from the granulate. The system with the developed in-house heat exchangers will be installed in one of the Polish power plants. The CO₂ capture system with the heat exchanger design is presented in Section 2.1 below.

2.1. Description of the CO₂ capture system

The method of CO₂ capture from combustion gases arising in the process of gas combustion and based on the MBTSA was described, among others, by SINTEF Industry in Norway [27]. The method was adapted to capture CO₂ in the case of pulverised coal combustion in a power boiler [28].

The method is based on using activated carbon in the moving bed adsorption process. This type of adsorbent was chosen for its properties. Activated carbons are considered suitable materials for CO₂ adsorption due to their porous structure - they have well-developed micro-porosity and mesoporosity. The high CO₂ adsorption capacity of activated carbons is due to their large surface area per unit volume, as well as the large area of submicroscopic pores in which impurities can be adsorbed. Carbon materials show lower sorption capacity and selectivity than zeolites but are nevertheless used in CO₂ gas purification processes. This is due to the interactions between activated carbon and CO₂, which are much weaker than the bonding of carbon dioxide to zeolites. This has the effect of requiring less energy to carry out the desorption process and is more easily regenerated. The advantage of activated carbons is that they are regenerative, they do not wear out during multi-stage adsorption/desorption cycles and are therefore ideal for MBTSA adsorption plants. In addition, activated carbon does not require prior removal of moisture from the flue gas [33]. The main advantages of the MBTSA method are low pressure drops in the adsorption zone and the possibility of a faster rate of the adsorbent heating compared with standard adsorption technologies [13]. Preliminary results of the MBTSA simulations [29] allow a conclusion that the degree of CO₂ capture achieved in this method is higher than that of the existing methods based on chemical adsorption (MEA and DAE). In order to examine the method suitability for rapid changes in the power unit load, with a particular focus on a rapid increase in power, it is necessary to design and build a test stand for the proposed method for CO₂ capture from hard coal combustion gases. SINTEF Industry in Norway is now in possession of a laboratory-scale system, a schematic diagram of which is shown in Fig. 1. The system was re-designed to a semi-industrial scale. The exchangers on the laboratory-scale test stand were made using a 3D printer. Considering high printing costs, it was necessary to find an alternative, cheaper solution. Due to that, the semi-industrial system makes use of tube exchangers in a staggered configuration. Fig. 2 and Fig. 3 present the heat exchangers arranged in a triangular configuration. Such arrangement of the exchangers was selected due to the limitations imposed by the room height and to ensure uniform loading of the floor. Activated

carbon in the form of granules with the diameter of 0.7 mm was used as the moving bed. The granules are shown in Fig. 4. Due to the formation of bed mounds, preliminary tests were carried out to eliminate the phenomenon of the adsorbent getting stuck in between the exchangers. As a result, the tube diameter no bigger than 13.5 mm was selected.

The exchanger thermal calculations are presented in Section 2.2.

The test stand will be installed in a Polish power plant. In order to confirm the method suitability for close-to-real conditions, the measurements will be performed during the system operation in steady- and transient-state conditions.

2.2. Determination of the heat transfer coefficient

The heat exchanger design process was based on the classical logarithmic mean temperature difference method for a cross-flow heat exchanger, in which the exchanger power is calculated from the following basic equation (1):

$$\dot{Q} = \frac{\Delta T_{LM}}{R_{tot}}, \quad (1)$$

where: R_{tot} is heat transfer total resistance, ΔT_{LM} is logarithmic mean temperature difference for the counter current system.

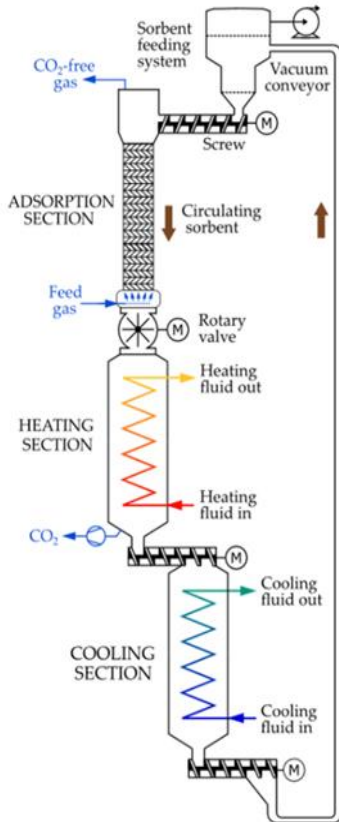


Fig. 1. Diagram of the laboratory system

The method was selected due to the fact that the adsorber temperatures at the inlets and outlets of the heating and cooling sections are known and result from the adsorber CO₂ capture property. According to the art of engineering, the following assumptions are adopted:

- steady-state operation of the exchanger,
 - no heat losses to the environment,
 - identical heat transfer coefficient across the entire heat transfer surface area,
 - constant physicochemical properties of the working mediums.
- Total thermal resistance was calculated from equation (2):

$$R_{tot} = R_w + R_t + R_s + R_{f,i} + R_{f,o} = \frac{1}{A_w \cdot h_w} + \frac{\ln(\frac{d_o}{d_i})}{2\pi \cdot r \cdot l} + \frac{1}{A_s \cdot h_s} + R_{f,i} + R_{f,o}, \quad (2)$$

where: R_w is resistance of the convective heat transfer from the inner surfaces of tubes to water, R_t is resistance of heat conduction through the tube walls, R_s is resistance of the heat flow on the sorbent side, $R_{f,i}$ is resistance of heat conduction through impurities on the inner side and $R_{f,o}$ is resistance of heat conduction through impurities on the outer side.

Equation (2) takes account of the resistance of impurities both on the inner and outer side of tubes. On the inner side, resistance $R_{f,i}$ takes values like for boiling water: 0.0002 (m²·K)/W, whereas on the outer side, $R_{f,o}$, being the resistance of the activated carbon deposit, is adopted according to [30] at the level of 0.002 (m²·K)/W.

The coefficient of the heat transfer from the water flowing inside the tubes is calculated like for a transient flow because the calculated Reynolds number is included in the range from Re=300 to Re=3000. Due to that, a correlation is used based on function $Nu=f(Re,Pr)$ for the following ranges of the dimensionless numbers: $2300 \leq Re \leq 106$, $0.1 \leq Pr \leq 1000$ [31]. The dimensionless numbers Nu , Re and Pr are defined as follows:

$$Nu = \frac{h d_w}{k}, \quad Re = \frac{w_m \cdot d_w}{\nu}, \quad Pr = \frac{c_p \cdot \mu}{k}. \quad (3)$$

The heat transfer coefficient on the side of the granulate was established experimentally on the laboratory stand in Norway and totals ~90 W/m²·K.

Assuming that the flow changes from laminar to transient at Re=2300, for which the Nusselt number is Nu=4.36, the correlation for the heat transfer coefficient is expressed by the following formula:

$$Nu = 4.36 + \frac{\xi (Re - 2300) Pr^{x_1}}{x_2 + x_3 \sqrt{\frac{\xi}{8} (Pr^3 - 1)}}, \quad (4)$$

where friction factor x is given by Filonienko [32] and can be calculated according to Taler [31].

$$\xi = \frac{1}{(1.2776 \log Re - 0.406)^{2.246}} \cdot 2.10^3 \leq Re \leq 10^7. \quad (5)$$

The unknown values of friction factor $x = (x_1, \dots, x_m)^T$ in equation (4) can be found using the least squares method:

$$S(x) = \sum_{i=1}^{n_{Re}} \sum_{j=1}^{n_{Pr}} (Nu_{ij}^m - Nu_{ij}^c)^2 = \min, \quad (6)$$

where: $Nu_{ij}^m = Nu^m(Re_i, Pr_j)$ and $Nu_{ij}^c = Nu^c(Re_i, Pr_j)$, $i = 1, \dots, n_{Re}$, $j = 1, \dots, n_{Pr}$ are the appropriately given, and approximated values of the Nusselt, Reynolds and Prandtl numbers are adopted as $n_{Re} = 10$ and $n_{Pr} = 16$. The values of the factors obtained using the least squares method are as follows:

$$x_1 = 1.008 \pm 0.0050, \quad x_2 = 1.08 \pm 0.0089, \quad x_3 = 12.39 \pm 0.0080 \quad [31].$$

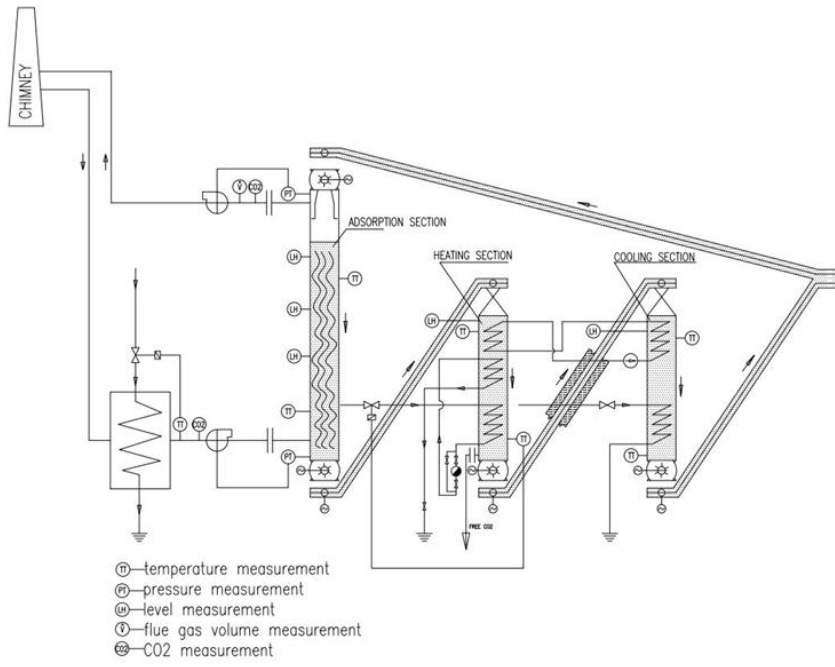


Fig. 2. Diagram of the semi-industrial system

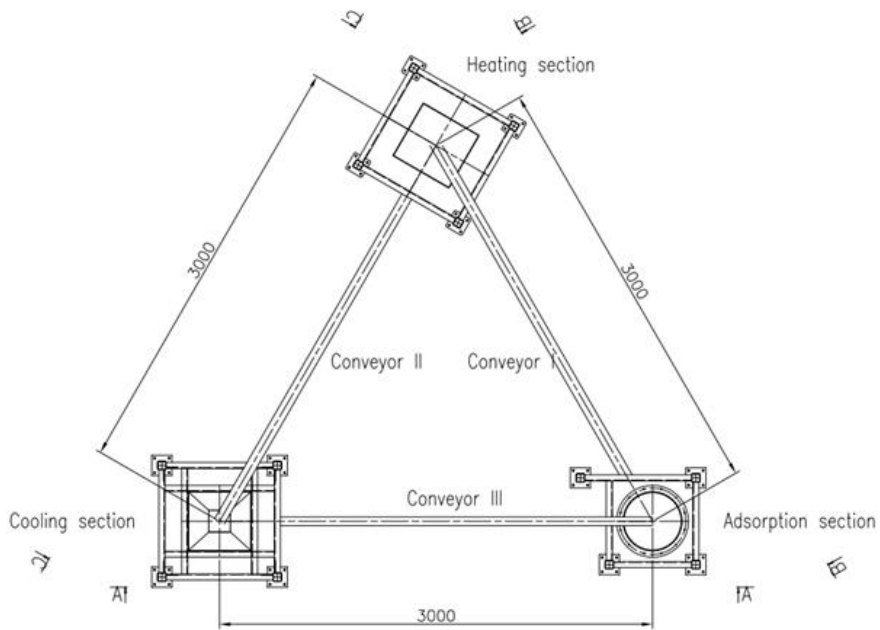


Fig. 3. Vertical projection of the semi-industrial system



Fig. 4. Activated carbon (www.kureha.co.jp)

2.3. Calculations of heat exchangers

The outcome of the thermal calculations and the applied equation (4) is the heat transfer surface area. The design details and the heat transfer surface area will be the subject of patent proceedings, and for this reason, they cannot be disclosed. Knowing the heat transfer surface area and the diameter of the tubes that will minimise the occurrence of the bed mound formation, it was possible to design the exchanger presented in Fig. 5 and Fig. 6. The described semi-industrial system was developed based on the SINTEF laboratory-scale stand located in Norway. The research system was made as a vertical column with the cooling, heating and adsorption section where the sorbent is fed by pneumatic conveyors (Fig. 1). In the target system, tube exchangers are used in a staggered configuration (Fig. 2), and the sorbent is transported by inclined belt conveyors. Such a solution is cheaper and does not require a big height of the room where it is located. Moreover, the designed exchangers have a module structure, which means that in the event of failure, a single module can be replaced without the need to replace the entire unit. The adopted assumptions made it possible to develop a compact design on a triangular plan, which minimised the system height. The system has three sections: the adsorption, heating and cooling section. The adsorbent with the inlet temperature of 30°C gets into the adsorption section and captures CO₂ from the combustion gases. From the adsorption section, it gets into the heating section, where it is heated to the temperature of 180°C. When this temperature is reached, CO₂ is released from the combustion gases and removed from the system through a connecting pipe. The adsorbent heating is realised in the developed exchangers, where the heating medium is saturated steam with the temperature of ~200°C. After CO₂ is released, the adsorbent gets to the cooling section, where the temperature is lowered to 30°C using cooling water supplied to the exchanger. At the same time, the water in the system is heated to the temperature of ~130°C and returned to the boiler cycle for regenerative heating of feed water. This is a procedure that will make it possible to reduce the method energy intensity to an expected value of less than 2.7 MJ/kg CO₂. The study was carried out according to [13] in gProms software, where the operation of the test stand was modelled. Data related to the flow velocity of the granule in the exchangers are presented in [13] for different operating conditions. The velocity value used in the calculations in the gProms programme determined for the granule transfer through the Mellapak was 5.7 mm/s.

During the development of the test stand, some limitations and challenges were diagnosed and worth considering. The main limitation of the installation is its size and dimensions. The test stand presented in this paper was designed for flue gas volume fluxes of several tens of m³/h. These fluxes under actual power plant operating conditions are much higher, which implies a correspondingly larger CO₂ installation. A solution to the problem may be to install all sections of the test stand vertically (but this will require an enclosed room or a specially placed high container).

Challenges may include designing an efficient sorbent transport. A good solution seems to be (compared with the one proposed in the paper) pneumatic transport. However, due to the internal heat recovery, this transport can generate high heat loss due to the low temperature of the air transporting the adsorbent. It

is also a challenge to minimise the loss of activated carbon through the use of suitable seals. This is related to its granularity (0.7 mm diameter) and any kind of leakage in the transport system which causes its loss.

Another challenge is to maximise the heat available after the adsorbent has cooled from about 180 °C to about 30 C. Under actual power plant operating conditions, there will be large amounts of this heat. It is planned to turn this heat back to regenerate the boiler feed water temperature.



Fig. 5. Fragment of the cooling section



Fig. 6. Cooling section exchangers

3. SUMMARY

This paper presents a CO₂ capture system using activated carbon with sections designed to exchange heat with exchangers ensuring high thermal efficiency.

The design work on the heat exchanger was focused on ensuring a high coefficient of the heat transfer from the side of the working mediums, i.e. the adsorbent, water and steam, minimising at the same time the phenomenon of the formation of granulate mounds over the tubes.

It is assumed that it will be possible to achieve the low energy intensity method of less than 2.7 MJ/kg CO₂ utilising hot water from the cooling section.

The designed test stand is now being built in a Polish power plant. The first tests of the stand are planned in the near future, together with the testing of the efficiency of CO₂ capture from pulverised hard coal combustion gases. In the future, the performance, cost and scalability of the developed system will be investigated and compared with current CO₂ capture technologies.

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