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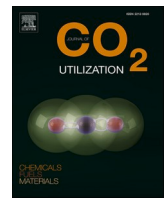
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Retrofitting hollow fibre carbon capture systems to decarbonise surface transport

Collette Larkin^a, Kyriaki Lampri^a, Simona Mazzone^a, Fermín Oliva^b, Kang Li^c, Francisco R. García-García^{a,*}

^a School of Engineering, Institute of Materials and Processes, University of Edinburgh, Robert Stevenson Road, Edinburgh EH9 3FB, UK

^b Repsol S. A., Calle Méndez Álvaro, 44, 28045 Madrid, Spain

^c Department of Chemical Engineering, Imperial College London, Exhibition Road, London SW7 2AZ, UK

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ABSTRACT

On-board carbon capture and storage (CCS) is being recognised as an essential transitional solution for transport decarbonisation. However, due to inherent space constraints, retrofitting current CCS technology to vehicles is challenging. Herein, the feasibility of a novel hollow fibre carbon capture system impregnated with a carbon xerogel for on-board CCS was assessed. The working capacity of four carbon xerogels with varying surface area/surface groups, over ten temperature swing adsorption (TSA) cycles, was studied in a packed bed adsorption unit (PBAU). The highest-performing carbon xerogel (*i.e.* CX) was deposited inside a hollow fibre support to form the hollow fibre adsorption unit (HFAU). Under typical vehicle exhaust gas compositions (*i.e.* 14 vol% CO₂ + 3 vol% H₂O balanced in air) and at mild operating conditions (*i.e.* adsorption at 25 °C, 1 atm; desorption at 125 °C, 1 atm), the HFAU exhibited a 1.3 times greater working capacity than the equivalent PBAU (*i.e.* 4.7 mmol g⁻¹ and 3.5 mmol g⁻¹, respectively), due to minimized mass transfer limitations. In addition, the results obtained were used in a study that compared hollow fibre carbon capture systems and packed bed carbon capture systems designed to be retrofitted to three transport scenarios: (1) a light-duty vehicle (car/van); (2) a large regional delivery truck; and, (3) a ferry. For scenarios 2 and 3, the findings of the study showcased the potential of retrofitting a hollow fibre carbon capture system to these large surface transport modes. However, for scenario 1, despite obtaining a hollow fibre carbon capture system that was 3.5 times smaller, 5.7 times lighter and 6.6 times cheaper than the packed bed carbon capture system, it cannot be feasibly retrofitted without significant improvements in weight. Furthermore, a short-hand simplified heat exchange approach was presented to obtain the TSA operation temperatures of the carbon capture systems.

1. Introduction

Transport is a major CO₂ emitter whose emissions are projected to rise beyond its current global contribution at 24.5%, making 2050 targets unattainable [1–4]. This is despite of governing bodies adopting better policies, improvements in engine efficiency and advancements in renewable fuels [1,2,5]. Thus, to ensure transport emissions are reduced on time, on-board carbon capture and storage (CCS) is proposed as the most economical and practicable decarbonisation solution for the short-to-mid-term [6,7]. This is because on-board CCS involves retrofitting a capture system, rather than undertaking the costly and complicated switch to renewable fuels. In particular, for heavy-duty vehicles and shipping vessels where the technology readiness level of

renewable fuel is low [8–10].

However, current CCS technology cannot be feasibly retrofitted to transport as it is designed for large emitting point sources. The challenge in mobile applications is to develop a compact, lightweight and cheap system that has a high CO₂ capture rate and can handle intermittent emissions of CO₂ at low concentrations and pressures [3,11].

For these reasons, research has focused chiefly on CCS for shipping vessels since the space and weight limitations are less constrained than other transport modes [3,12]. For example, Long et al. recently studied on-board CCS for a 3 MW diesel engine-powered cargo ship. Using the benchmark solvent, 30 wt% aqueous monoethanolamine (MEA), they reported that capturing a 99% pure CO₂ stream was feasible at a CO₂ recovery of 87.4% [13]. However, when considering applications where

* Corresponding author.

E-mail address: francisco.garcia-garcia@ed.ac.uk (F.R. García-García).

the user is close to the CCS technology, amine scrubbing is not suitable due to the health issues posed by toxic amines.

Other researchers have focused on temperature swing adsorption (TSA) with packed bed adsorbents for shipping vessels. This is because TSA can provide the benefit of reducing regeneration costs by implementing low-grade waste heat recovery from the hot exhaust gas [3,14,19,20]. For instance, Balsamo et al. [14] reported a CO₂ recovery of 43% using raw and alumina-supported K₂CO₃ under typical marine diesel engine exhaust compositions and temperatures. Erto et al. [15] built upon this work further by assessing the CO₂ capture performance of K₂CO₃ supported on porous alumina under the same typical marine exhaust compositions. After subjecting the K₂CO₃-based material to ten carbonation/regeneration cycles at 60 °C and 120 °C, respectively, the researchers reported a CO₂ recovery of 30%. Both of these studies pose an interesting solution for shipping vessels, however, they are not applicable for road transport as they would be too large to retrofit.

To overcome this, researchers are striving to optimise the reactor design by developing monoliths with high surface-area-to-volume ratios. In this configuration, the adsorbent is thinly coated on the channels of the monolith to produce smaller adsorption units with improved pressure drop [16]. Nevertheless, monoliths, suffer from an accumulation of unused material in the outermost corners of their structure, decreasing their efficiency and increasing their operational cost [17,18].

An alternative solution to monoliths are hollow fibre-based systems which show surface area-to-volume-ratios more than one order of magnitude higher than monolithic structures with improved mass transfer [19,20]. Indeed, in our recent proof-of-concept study [21], we demonstrated how hollow fibre carbon capture systems produce cheaper, more compact and more efficient on-board TSA adsorption systems with enhanced lifetime than the equivalent packed bed system. Additionally, selecting an adsorbent material with a high CO₂ working capacity is crucial to obtaining a compact CCS system. In the CO₂

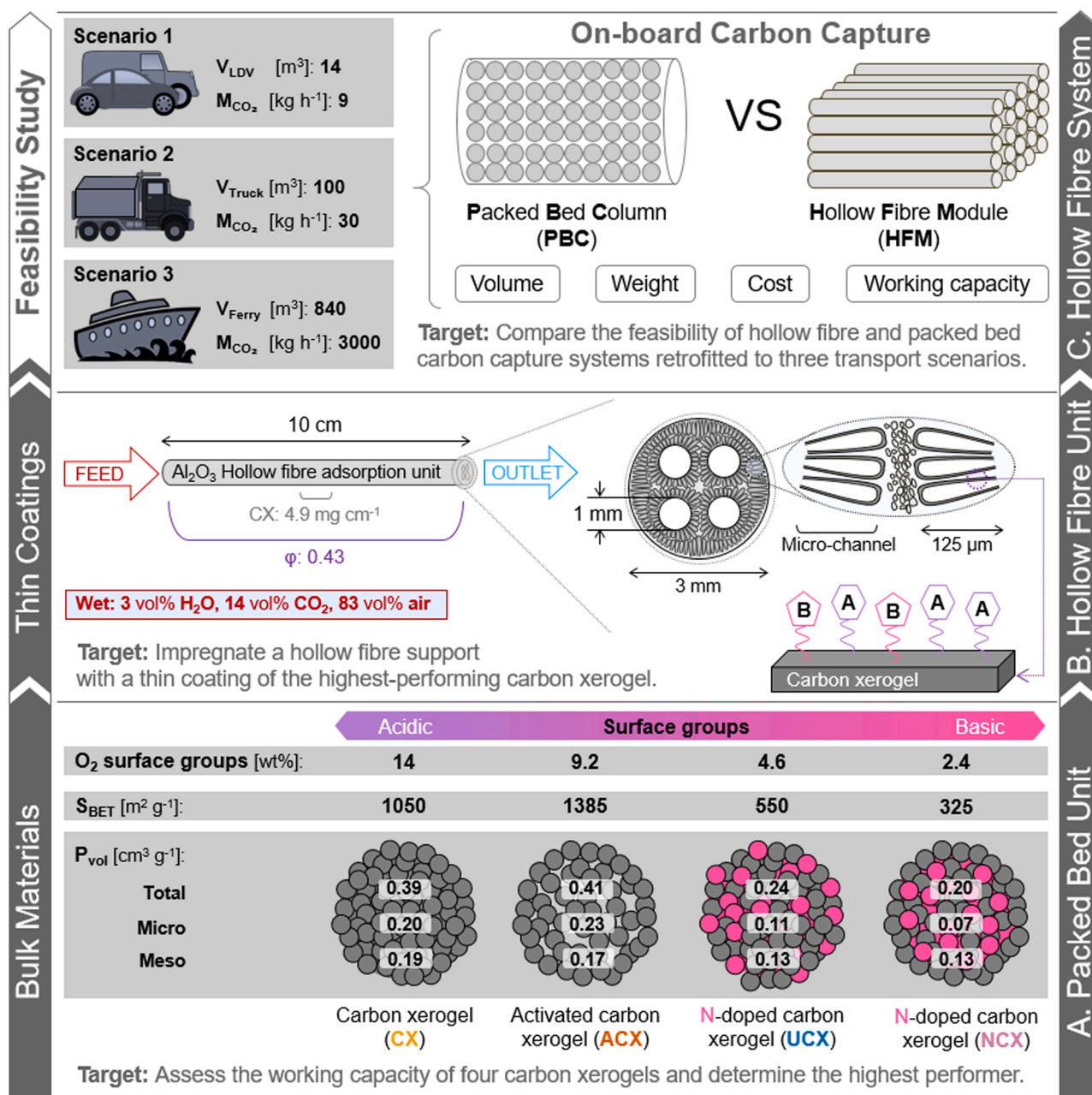


Fig. 1. A summary of the three-step procedure executed in this work. Bottom to top: A) Textural and surface group properties of the carbon xerogels, B) Schematic of the hollow fibre adsorption unit, C) Schematic of the feasibility study undertaken.

capture community, carbon xerogels are gaining interest as they are a cheap, porous material that can achieve high working capacities at mild operating conditions. Moreover, their textural and surface group properties can be finely tuned to suit their application, increasing their working capacity [22,23].

Thus, a promising solution for on-board CCS is the hollow fibre carbon capture system impregnated with a carbon xerogel. In their work, Mazzone et al. [18] successfully impregnated a ceramic hollow fibre support with a thin coating of catalytic material via the sol-gel Pechini method to develop a compact hollow fibre system [18,24].

To that end, a hollow fibre carbon capture system for retrofitting to surface transport was studied in this work using a three-step approach, as outlined in Fig. 1. Firstly, four carbon xerogels with varying surface area/surface groups were tested in packed bed adsorption units (PBAUs) under typical vehicle exhaust gas compositions (*i.e.* 14 vol% CO₂ + 3 vol% H₂O balanced in air) [11,25,26] and their working capacities were obtained, see Fig. 1A. In the second step, the highest-performing carbon xerogel was impregnated as a thin coating inside a four-channelled asymmetric α -Al₂O₃ hollow fibre support to produce the hollow fibre adsorption unit (HFAU), see Fig. 1B. The working capacity of the HFAU was obtained under the same wet feed conditions as the PBAU. Subsequently, in the third step, the working capacities obtained for the HFAU and PBAU were used to complete a case study comparing the volume, weight and cost of scaled-up hollow fibre and packed bed carbon capture systems designed to capture the CO₂ emissions released by three common transport scenarios: (1) a light-duty vehicle (car/van); (2) a large regional delivery truck; and, (3) a ferry, see Fig. 1C.

2. Experimental Procedure

The materials and characterisation methods used are listed in the [Supplementary Information](#).

2.1. Carbon xerogel synthesis

Commonly, the method used to synthesize carbon xerogels consists of the pyrolysis of resorcinol-formaldehyde xerogels. Once formed, the carbon xerogels can undergo an array of treatments to modify their textural and surface group properties [27].

In this work, resorcinol-formaldehyde xerogels were prepared following the sol-gel Pechini method first proposed by Pekala [28]. After which, four carbon xerogels with varying surface areas/surface groups were obtained (*i.e.* pellet size 125 – 250 μ m), see Fig. 1A. The carbon xerogel labelled as CX was obtained by the pyrolysis of the resultant xerogel in a N₂ atmosphere (*i.e.* 100 ml min⁻¹ (STP)) at 800 °C for 1 h. To produce the carbon xerogel with increased surface area, labelled as ACX, a second sample of CX was treated in a CO₂ atmosphere (*i.e.* 25 ml min⁻¹ (STP)) at 800 °C for 1 h. Carbon xerogels containing basic surface groups were obtained by employing two different methods: i) co-precursor method using urea as a nitrogen source in the xerogel starting solution, labelled as UCX; and, ii) post-synthesis activation of a third sample of CX in an NH₃/air atmosphere (*i.e.* NH₃:Air = 1:3, 100 ml min⁻¹ (STP)), labelled as NCX.

2.2. Hollow fibre preparation and impregnation

A 4-channelled asymmetric α -Al₂O₃ hollow fibre support 10 cm in length was fabricated using the phase inversion technique, followed by sintering at high temperatures [29,30]. The sol-gel Pechini method was used to impregnate the hollow fibre support with the highest-performing carbon xerogel, forming the HFAU.

2.3. Working capacity

The working capacity of the carbon xerogels and HFAU was obtained over ten consecutive TSA cycles. Two feed conditions were used during

the adsorption step: dry (*i.e.* 14 vol% CO₂ balanced in air) and wet (*i.e.* 14 vol% CO₂ + 3 vol% H₂O balanced in air). For the wet feed conditions, 3 vol% H₂O was incorporated using a saturator. During the desorption step, N₂ was used as a carrier gas to facilitate the release of the adsorbed CO₂. Adsorption was carried out at 25 °C, 1 atm while desorption was carried out at 125 °C, 1 atm. A total feed gas flow rate of 100 ml min⁻¹ (STP) was used throughout and carefully selected to ensure: (a) the experiment was not external mass transfer-limited at 100 ml min⁻¹ (see Figure S.3 of the [Supplementary Information](#)); and, (b) the space velocity is within the operation window of an automotive catalytic converter (*i.e.* S.V: 25,000 to 150,000 h⁻¹) [31].

The working capacity was defined as the capacity when a breakthrough of 5% of the CO₂ feed concentration was observed in the outlet stream. The gas composition of the outlet stream of each TSA cycle was monitored in real-time and recorded using a mass spectrometer (EcoSys-PTM Mass Spectrometer) connected to a computer.

3. Results and discussion

3.1. Carbon xerogel characterisation and performance study

Fig. 2 A presents the characterisation of the four carbon xerogels which includes SEM imaging at x2700 magnification, CO₂ adsorption/desorption isotherms at 0 °C and the chemical structures of the acidic/basic groups present on their surfaces.

The SEM of fresh samples of the carbon xerogels shows no visible difference in the morphology between the carbon xerogels. This suggests that treatments to increase surface area or modify surface groups have not changed the roughness of the samples.

Notably, it can be seen from the CO₂ adsorption/desorption isotherms at 0 °C, that ACX had the greatest CO₂ uptake at 113 cm³ g⁻¹ while CX had the lowest CO₂ uptake at 79 cm³ g⁻¹. The increase in CO₂ uptake observed by ACX can be attributed to ACX having a 25% larger surface area than CX (*i.e.* 1385 m² g⁻¹ and 1050 m² g⁻¹, see Fig. 1A) and 35% fewer oxygen surface groups than CX (*i.e.* 9.2 wt% and 14 wt%, see Fig. 1A). This result agrees with the findings of Radovic et al. [32] who reported that the strong electronegativity of the oxygen surface group has an electron-withdrawing effect, reducing the affinity of the carbon xerogel surface towards CO₂ [33,34]. In this respect, it is also worth it to mention work undertaken by Xing et al. [22], which suggests that oxygen surface groups enhance CO₂ adsorption via hydrogen bonding interactions. Nevertheless, this effect was not observed in the carbon xerogels based on the.

CO₂ isotherms here reported. Furthermore, it can be seen in Fig. 2A that basic surface groups have a positive effect on CO₂ capture performance as UCX and NCX had higher CO₂ uptake values than CX (*i.e.* 90 cm³ g⁻¹ and 83 cm³ g⁻¹, respectively). This agrees well with literature as it is believed basic surface groups can increase CO₂ uptake via two mechanisms: i) electron-donating effect [23,35,36]; and, ii) formation of carbamates [37]. A schematic diagram of all the different adsorption mechanisms mentioned above can be seen in Fig. 2B.

Total capacity, working capacity and lost capacity for each carbon xerogel over ten TSA cycles under the dry and wet feed conditions are shown in Fig. 2C. Despite ACX demonstrating the highest CO₂ uptake in the CO₂ isotherms, the four carbon xerogels showed an equal working capacity under both dry and wet feed conditions. This can be attributed to a concentration of 14 vol% CO₂ being too low for the benefits of high surface area and absence of oxygen surface groups to be realised. In this respect, as can be seen in Fig. 2A, to use the full potential of ACX, higher concentrations of CO₂ in the exhaust are required (*i.e.* pre-combustion: 37.7% CO₂ and oxy-fuel combustion: 85% CO₂) [12,38].

Fig. 2 C also shows that the working capacities of the carbon xerogels increased by a factor of ten when subjected to the wet feed conditions in comparison to the dry. This increase can be explained by the formation of CO₂ hydrates on the surface of the carbon xerogel which enhances CO₂ adsorption [22,23,39,40]. Such an increase in uptake under wet

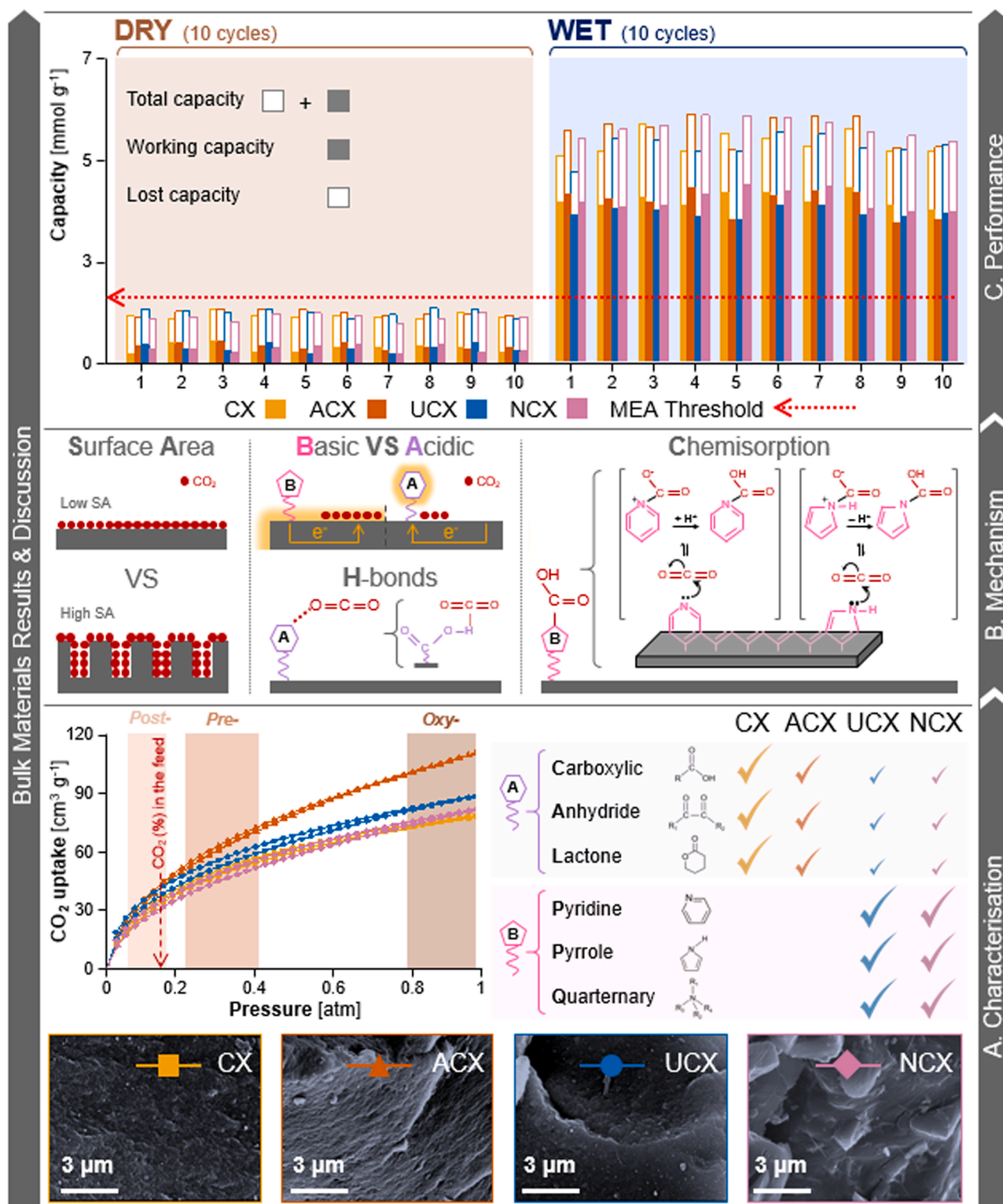


Fig. 2. A) SEM images at x2700 magnification, CO₂ adsorption/desorption isotherms at 0 °C and the acidic/basic surface groups of the carbon xerogels. B) CO₂ adsorption mechanisms that occur on the surface of the carbon xerogels. C) Total, working and lost capacities of the carbon xerogels after ten TSA cycles under the dry and wet feed conditions.

conditions is extremely attractive economically and practically as it eliminates the need for H₂O removal before CO₂ capture.

In addition, it should be noted that under the wet feed conditions, the carbon xerogels exhibited average working capacities above 2 mmol g⁻¹, making them competitive with the toxic industry-standard absorption material, MEA (*i.e.* average working capacities of

3.5 mmol g⁻¹ in the wet feed conditions) [41]. Furthermore, the high CO₂ capture performance of the carbon xerogels at mild operating conditions presents the opportunity for the waste heat of the high-temperature exhaust gas to be recovered. Thus, lowering the cost associated with the regeneration of the sorption material which is often the most costly operation step for the majority of CO₂ capture

technology [12,42,43].

For practical reasons, pure N₂ was used during the desorption step but it is worth noting that air could be used as the carrier gas instead. As it can be seen from Figure S.2, air-TGA profiles of the four carbon xerogels here studied, at the desorption temperature of 125 °C the carbon xerogels were stable. However, for air to be used, the effect the low content of CO₂ present in air (i.e. 0.04 vol% CO₂) has on the capacity should be considered. It can be seen from the CO₂ isotherms in Fig. 2A, that at 0.04 vol% CO₂ the adsorbents have an uptake of 1.9 cm³ g⁻¹ (i.e. 0.004 kg-CO₂ kg-CX⁻¹). Thus, if air was used as the carrier gas during

the desorption step, the capacity would decrease by approximately 6%. Therefore, although a maximum decrease of 6% in the capacity would be observed, it can be assumed that desorbing with air could be used as a compromise.

3.2. Hollow fibre adsorption unit and packed bed adsorption unit performance study

In the second step of this work, CX was deposited as a thin coating inside a hollow fibre support. This was because the carbon xerogels

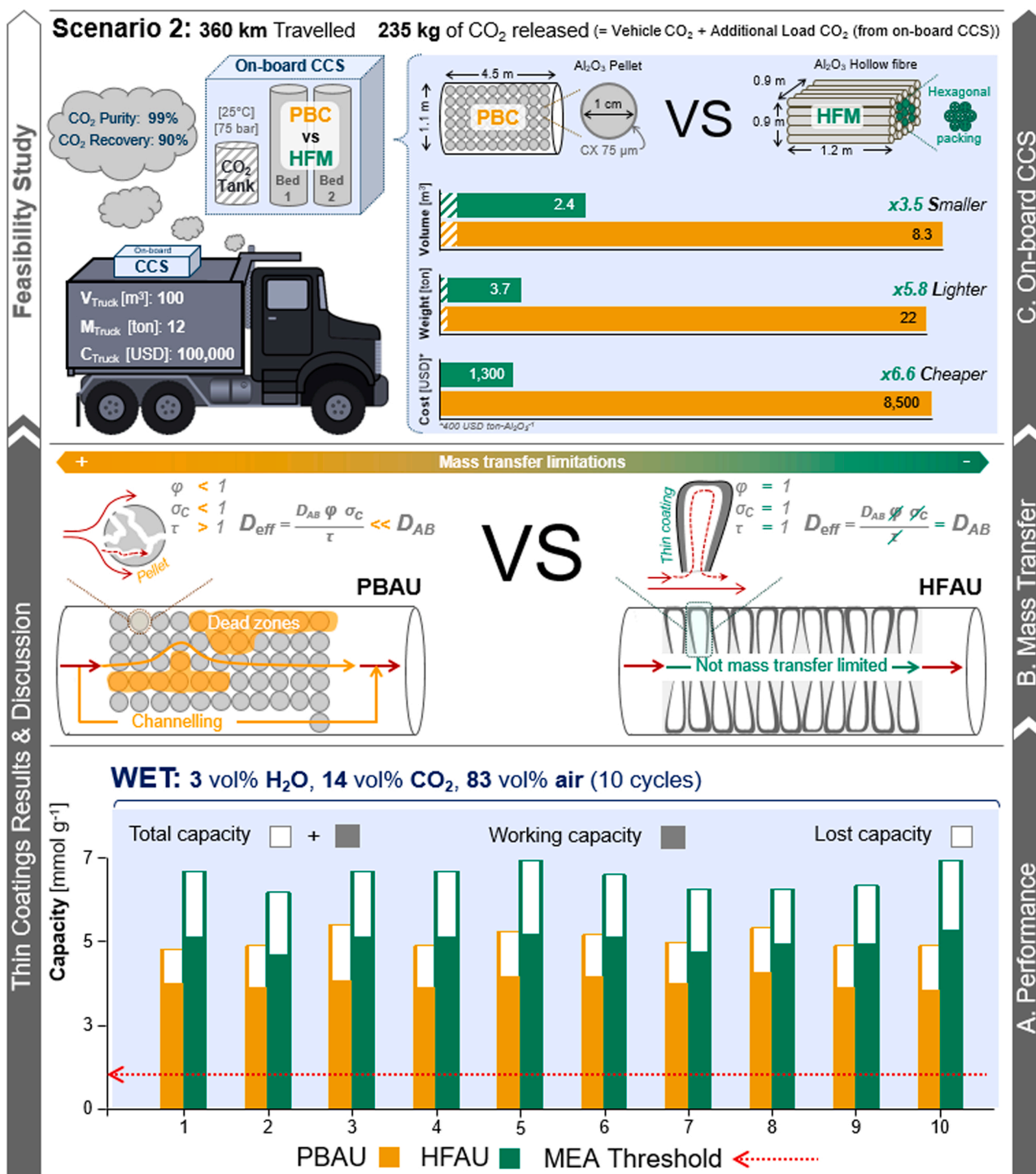


Fig. 3. A) Performance of the PBAU and HFAU after ten adsorption/desorption cycles under wet feed conditions. B) Diffusion mechanisms in the PBAU and HFAU. C) The results of the case study assessing the feasibility of retrofitting CCS systems to capture the CO₂ emissions released by a large regional delivery truck.

exhibited an equal average working capacity at 14 vol% CO₂ and CX had the simplest synthesis procedure, which enabled the number of resources and stages required for the preparation of the HFAU to be minimised.

Fig. 3 A shows the total, working and lost capacities of the PBAU and HFAU over ten TSA cycles for the wet feed conditions. Under the wet feed conditions, the average working capacity of the HFAU is 1.3 times greater than the PBAU (i.e. 4.7 mmol g⁻¹ and 3.5 mmol g⁻¹). The greater average working capacity can be attributed to the morphology of the HFAU minimising mass transfer limitations, see Fig. 3B. The schematic diagram of Fig. 3B shows that in a PBAU, internal diffusion is limited by a complex network of tortuous interconnecting pathways of pore bodies (expressed by porosity, constriction factor and tortuosity) [18,44,45]. In contrast, in a HFAU, internal diffusion limitations are bypassed as diffusion is governed by the open micro-channels of the hollow fibre support such that kinetics as close to practicable to ideal are achieved. Moreover, by depositing the carbon xerogel inside the micro-channels of the hollow

fibre support, deviations from ideal flow patterns such as dead zones and channelling are avoided, an issue regularly experienced by packed bed systems. As a result, the observed working capacity and lifetime of a HFAU are greater than that of a PBAU [17,44].

3.3. Feasibility study

Using the working capacities obtained for the PBAU and HFAU under the wet feed conditions, a short-hand study was conducted to illustrate the potential application of CCS systems in the transport sector. In this study, the feasibility to retrofit a hollow fibre carbon capture system and a packed bed carbon capture system to the following three common transport scenarios were compared: (1) a light-duty vehicle (car/van); (2) a large regional delivery truck; and, (3) a ferry, see Fig. 1C. The carbon capture systems consisted of two TSA beds (two hollow fibre modules (HFMs) for the hollow fibre carbon capture system, and two packed bed columns (PBCs) for the packed bed carbon capture system) and a CO₂ storage tank (with an estimated mass of 0.72 kg to store 1 m³ of liquid CO₂ at 25 °C, 75 bar) [3,11]. The carbon capture systems were designed to account for the additional CO₂ emissions that were released as a result of increasing the loading of the vehicle by retrofitting the carbon capture system. As shown in the Supplementary Information, in all of the scenarios here studied, the hollow fibre carbon capture system requires a smaller volume and less adsorbent compared to a packed bed carbon capture system. For instance, as can be seen in Fig. 3 C, to capture 90% of the CO₂ emissions at a purity of 99% released by a truck travelling 360 km, a hollow fibre carbon capture system at 2.4 m³ requires 130 kg of CX. Whereas the equivalent packed bed carbon capture system would be 3.5 times greater in volume (i.e. 8.2 m³) and needs almost one third more kg of CX (i.e. 180 kg). Moreover, summing up the weight of the Al₂O₃ support (i.e. hollow fibre supports for the hollow fibre carbon capture system and pellets for the packed bed carbon capture system) with the weights of the CX, CO₂ captured and CO₂ storage tank, the total weight of the hollow fibre carbon capture system would be 3700 kg while the packed bed carbon capture system would be 22,000 kg. Considering the price of Al₂O₃ to be 0.4 USD kg⁻¹ [46], the hollow fibre carbon capture system would cost 1300 USD while the packed bed carbon capture system would cost more at 8500 USD. As a result, the hollow fibre carbon capture system is 3.5 times smaller, 5.8 times lighter and 6.6 times cheaper, see Fig. 3 C. Using a simplified heat exchange approach, it was assumed that the hot exhaust gas released by the vehicles provided the thermal energy required to heat up each bed to the desorption temperature. For example, looking at the on-board carbon capture system designed for the diesel-powered regional delivery truck, it can be assumed that at a high engine load, the vehicle exhaust leaves the engine at a temperature of 400 °C [47]. After the heat exchange, it was assumed that the exhaust gas was cooled to the adsorption temperature. However, further detailed calculations should be

completed in future research to ensure the adsorption temperature is achieved.

The detailed procedure for the design of both carbon capture systems can be found in the Supplementary Information, along with the results for scenarios 1 and 3. For scenarios 2 and 3, the weights obtained for the hollow fibre carbon capture systems show promising potential for its implementation on-board these large surface transport modes (i.e. 3.7 ton for scenario 2 and 480.0 ton for scenario 3, respectively). Particularly, since for these scenarios, the space and weight limitations are less constrained and the technology readiness level of renewable fuels is low [8–10]. However, currently, for scenario 1, the hollow fibre carbon capture system cannot be feasibly retrofitted without significant improvements in weight (i.e. 1.1 ton for scenario 1). For instance, the weight of a light-duty vehicle ranges from 1.0 ton to 3.7 ton and as a result, retrofitting a carbon capture system that weighs 1.1 ton is not practicable.

With this, it should be noted that further improvements to reduce the volume and weight of the hollow fibre carbon capture system should be explored to increase the competitiveness of hollow fibre carbon capture technology, particularly for light-duty vehicles. One holistic approach that can be implemented to reduce the dimensions of the overall hollow fibre unit volume is to replace 100% carbon-based fuel with carbon-based fuel and carbon-free fuel mixtures. In recent work, we investigated the effect fuelling mode (i.e. single - 100% carbon-based fuel; and, dual - a mixture of carbon-based fuel and carbon-free fuel) has on the volume of a hollow fibre carbon capture system that uses CX. The results of the study showed that the volume of the system can be reduced by using a dual mixture, however, there is a trade-off between decreasing the carbon-based fuel load in the mixture and the working capacity of the adsorbent. Notably, this work was presented as an oral presentation at both the VI UK Emission Control Workshop and the 16th International Conference on Greenhouse Gas Control Technologies. Another approach that can be undertaken is to optimise the loading of the carbon xerogel inside the hollow fibre micro-channels so that for a given length of the hollow fibre adsorption unit, its CO₂ capture performance is maximised. In our previous work, [21], the loading of sorbent inside a hollow fibre support was studied by increasing the number of impregnations of sorbent until saturation of the micro-channels was observed. The results of the study showed that the hollow fibre adsorption unit subjected to three impregnations achieved the highest CO₂ capture performance per unit length. Moreover, the mass of sorbent deposited after three impregnations was approximately double the amount deposited after one impregnation. This demonstrates that the hollow fibre carbon capture system volume can be reduced by optimising the sorbent loading.

4. Conclusions

In this work, a high-performing carbon xerogel (i.e. CX with an average working capacity of 3.5 mmol g⁻¹ at 25 °C, p_{CO₂} = 0.14 atm) suitable for capturing CO₂ under typical vehicle exhaust gas conditions was synthesised. CX was stable under ten TSA cycles (i.e. adsorption was carried out at 25 °C, 1 atm while desorption was carried out at 125 °C, 1 atm). In addition, the impregnation of the CX inside an α-Al₂O₃ hollow fibre support increased its working capacity by a factor 1.3 when directly compared against a packed bed adsorption unit. The enhanced performance of CX arises from the morphology of the hollow fibre minimising mass transfer limitations.

The feasibility study of scaled-up carbon capture systems shows that on-board CCS for large surface transport modes, such as large heavy-duty trucks and marine vessels, can be possible through retrofitting the hollow fibre carbon capture system. For instance, the results of the study showed that the hollow fibre carbon capture system retrofitted to a large regional delivery truck would be approximately 3.5 times smaller, 5.8 times lighter and 6.6 times cheaper than a packed bed carbon capture system. Whereas, currently, for smaller surface transport modes such as light-duty vehicles, further improvements in weight must be

achieved before the hollow fibre carbon capture system can be feasibly retrofitted.

Finally, the short-hand simplified heat exchange approach suggests that heat integration can be engineered to obtain the temperatures used in the desorption step (*i.e.* 125 °C) and in the adsorption step (*i.e.* 25 °C). Therefore, overall, these findings will progress the design and development of novel, compact hollow fibre carbon capture systems for on-board CCS, ensuring transport net zero targets are achieved on time.

CRedit authorship contribution statement

Collette Larkin: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Kyriaki Lampri:** Data curation. **Simona Mazzone:** Data curation. **Fermín Oliva:** Supervision. **Kang Li:** Resources. **Francisco R. García-García:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Project administration, Resources, Supervision, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jcou.2022.102336](https://doi.org/10.1016/j.jcou.2022.102336).

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