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Reply to the 'Comment on "Migration-assisted, moisture gradient process for ultrafast, continuous CO_2 capture from dilute sources at ambient conditions"' by J. Casado, *Energy & Environmental Science*, 2022, 10.1039/D2EE00555G

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Prajapati et al.[1] developed a novel, robust, modular and industrially-scalable CO_2 capture process that u
In a recent comment to our article, Casado[2] argued against the feasibility of the MAMG process for an i
Casado[2] argued that carbonate and hydronium react to form bicarbonate, not the reverse, as shown in Ec

Regarding the comment "the paper neglects the main product of reaction (1) in the overall process: oxygen," all the physical processes, including water-dependent bicarbonate dissociation, anodic reaction, cathodic reaction, and transport processes, are described physically and mathematically in section S2.2 of the supporting information. The anodic reaction and co-evolution of CO_2 at the anode are shown clearly in Fig. 1A and 1B of the article.

Although CO_2 reduction is not a focus of this article[1], Casado[2] raised a couple of concerns about the possible integration of CO_2 capture and reduction unit. To address them, we would like to clarify that (a) CO_2 reduction reaction (CO2RR) can be carried out in a separate electrolyzer with suitable electrodes and bias while using a CO_2-saturated aqueous solution from the MAMG process. (b) CO2RR and MAMG CO_2 capture units can be placed in tandem such that the captured CO_2 on the aqueous side will act as an aqueous reservoir for the CO2RR electrolyzer. There would be no requirement for the voltage reversal on the electrodes with this arrangement (c) On a larger scale, MAMG CO_2 capture, when performed in a commercial electrodialysis (ED) unit, will yield pure streams of CO_2 and avoid mixing with O_2, since the rinse solution of the anode is separated from the dilute (organic solution) and concentrate (aqueous solution) streams.[3] This can be realized by tracing the trajectory of bicarbonate ions, as shown in Figure S.7 in the supplementary information of the paper[1]. Thus, captured CO_2 in the aqueous phase will not contain any oxygen and can be used for CO2RR purposes.[3]

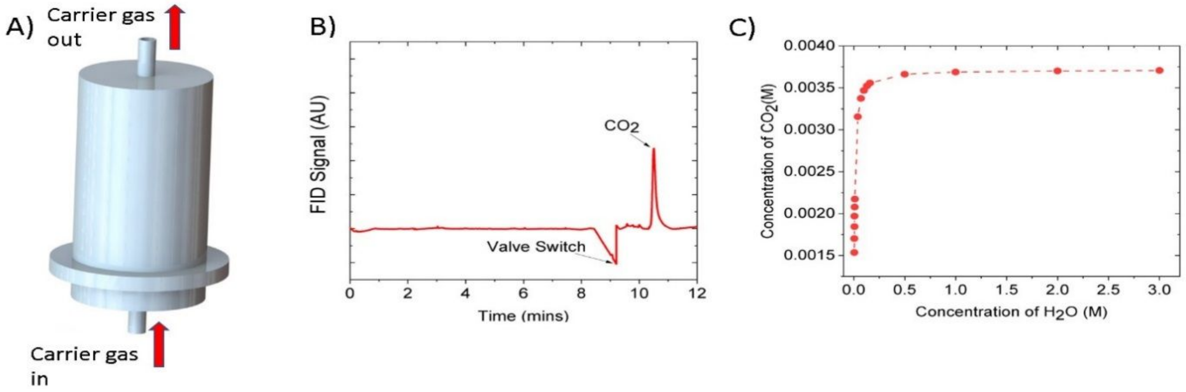

Figure 1: (A) 3D design of the packed bed containing $KHCO_3$ (B) Gas chromatograph for verification of autocatalytic decomposition of HCO_3^- in presence of water. Valve switch occurs during operation of GC (C) Effect of water concentration on concentration of CO_2 evolved

To address another comment, "*A minimal attempt to assess the effect of some impurities is given in the supplementary information.*" [2], an additional MAMG CO_2 capture experiment was carried out in the presence of water vapor and NO gas, and the performance was evaluated using an online pH probe and ion-chromatography. The flue gas composition used for this experiment contained 12 % CO_2, 7% O_2, 0.16% SO_2, <0.1% NO, and balance N_2. Incoming flue gas was bubbled through the humidifier bottle resulting in the outlet stream to 100% R.H. When the humidified flue gas stream was introduced into the organic reservoir of the MAMG

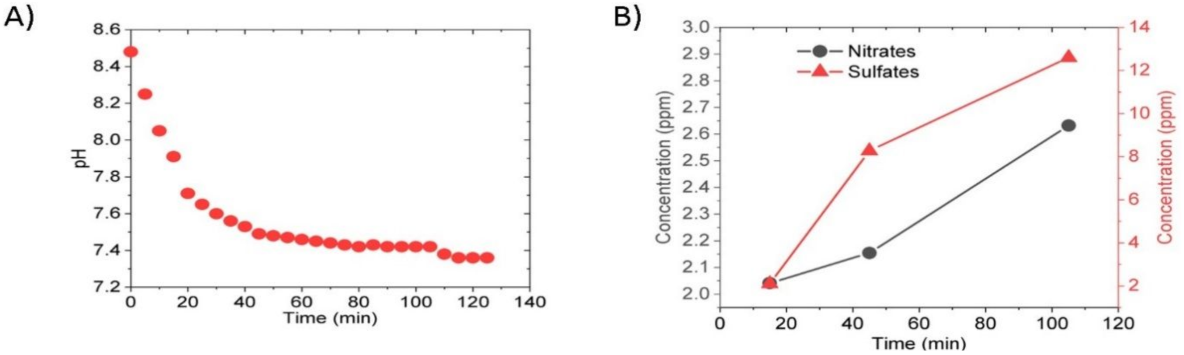

Figure 2: (A)Change in pH for a MAMG CO_2 capture experiment in the presence of impurities in the flue gas. (B) The concentration of ions in the aqueous phase of the MAMG CO_2 capture experiment

CO_2 capture process, migration of bicarbonate ions was still achieved, which is shown in Fig. 2A. Furthermore, the presence of sulfates and nitrates could be confirmed in Fig. 2B. From Fig. 2A, it can be readily seen that the pH decrease results from bicarbonate, sulfate, and nitrate migration. Due to 100% R.H., the dynamics of the process are slightly dampened as compared to Fig 4A of the article. Theoretical discussions for such behavior have already been provided in Fig 5D of article[1]. However, even under extreme flue gas humidity conditions, the MAMG CO_2 capture process shows significant activity and rate. Thus, we believe that the MAMG CO_2 capture process will be robust on an industrial scale.

The scope of the article[1] was to showcase the novelty of the MAMG CO_2 capture process instead of d

We appreciate comment[2],"*No attempt was made to measure the flux and composition of the gas liberated from the aqueous phase.*" Therefore, we provide the data to analyze the gaseous composition liberated from the aqueous phase. In Fig. 3, a theoretical vs. experimental comparison of the evolution of CO_2 liberated from the aqueous phase has been provided. Since the water oxidation reaction occurs at the anode of a MAMG CO_2 capture unit, it is self-evident that O_2 will be a by-product in the incoming stream. However, as explained previously, the evolution of O_2 will be circumvented when stacks of ED units are deployed on an industrial scale for MAMG CO_2 capture.[3]

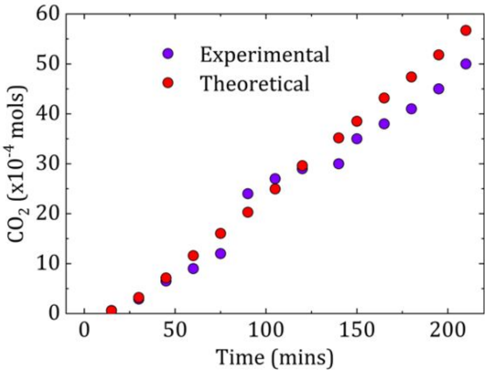

Figure 3: Comparison of the theoretical gaseous CO_2 released from the MAMG process vs. the experimental CO_2 released monitored through GC.

Finally, we want to mention that methanol was preferred over ethylene glycol for FTIR and KF titration experiments only. The reasons have already been provided in two instances of the paper[1]. "CH_3OH is used as a… without the interference of EG" in section 2.1.4 and "CH_3OH was chosen a solvent … seen in an aqueous system" in section 3.1.

In the techno-economic calculations, Casado[2] also pointed out that the additional potential drop caused due to the bipolar membrane (BPM) in electrolyte was not accounted for in our calculations. We want to underscore that the potential drop caused by BPM strictly depends on the operating current densities of the electrodialysis (ED) unit. We have now obtained a commercial BPM ED unit, which consistently shows a potential drop of 1.5 V per membrane pair when operated at 200 mA of current. Also, we strongly disagree with comment[2] regarding the standard price of electricity used for calculations. The industrial price of electricity in 2020 for the USA and Germany was close to \$40/ MWh.[4] Based on this cost of electricity, the new estimate for the cost of carbon capture is \$152 per ton of CO_2.

Conclusion: The data provided in the original work[1] and the additional experiments provided in this response letter prove the feasibility of the MAMG CO_2 capture process. The experimental evidence provided confirms i) water-dependent bicarbonate dissociation, ii) evolution of pure CO_2 using ED unit, iii) minimum impact of flue gas impurity on MAMG process, and iv) technoeconomic feasibility. Hence, we firmly believe in the broad applicability of the MAMG CO_2 capture process on an industrial scale.

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