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An organic solvent-free route for preparing silica-alkoxylated polyethyleneimine adsorbents for CO_2 capture

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Abstract

Post-combustion CO_2 capture (PCC) plays a key role in reaching climate neutrality by 2050[1], since it can be retrofitted and adapted to existing power plants and industrial processes. Mesoporous silica-supported polyethyleneimine (PEI), displaying high adsorption capacity and selectivity, are promising adsorbents for CO_2 capture and, more recently, alkoxylated PEI (APEI) given the most significant improvement in CO_2 capture performance [2], by improving oxidative stability and shifting adsorption and desorption to lower temperatures.

Wet impregnation is the established procedure to prepare silica-PEI, with PEI occupying the available silica pore volume. However, excessive quantities of organic solvents, particularly methanol, have invariably been used for both PEI alkoxylation and mixing the polymer with silica. In all reports, excessive quantities of organic solvents such as methanol have been used for mixing [2] [3] [4]. Typically, 10-20 ml of solvent per gram of substrate with vacuum drying at 40-60 °C is used for 3-12 hrs to reduce the methanol concentration to <2 Wt.% adsorbent. Further, large quantities of organic solvents have also been used for alkoxylating PEI, typically 5-15 ml of methanol per gram of PEI [5].

This study demonstrates an organic solvent-free route for mesoporous silicas prepared from sodium silicate solution, alkoxylation of PEI and the subsequent successful impregnation of PEIs with minimal quantities of water, typically, the mass ratio of water to silica being no more than 1.0. The silica used here possesses a relatively large pore volume (total pore volume is 1.75 ml/g) and Brunauer-Emmett-Teller surface area (BET SA, is 284 m^2/g), with 93% of pore volume contributed by mesopores (1.63 ml/g). It has a particle size of 100-300 microns, which is suitable for fluidisation and has been used in an earlier study with bubbling beds for adsorption and desorption [6]. However, drying on large scale samples needs to be controlled where the dried adsorbents should retain ca. 5 Wt.% moisture to prevent any performance loss.

The results indicate organic solvent-free route for preparing silica-APEI adsorbents for CO_2 capture has successfully been demonstrated at 1g, 2 kg, and 5 kg scales. A 47 wt.% APEI and PEI loading was identified as ideal for the mesoporous silica used in this reseach. Water proved to be an effective sustainable solvent for PEI and APEI impregnation compared to methanol, ethanol, and acetone, with only 1 ml per gram of silica being the minimum amount required to achieve maximum adsorption capacity, while

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larger volumes up to 10 ml showed no negative impact on the isotherms. Alkoxylation can be tailored to maximise APEI's performance for direct air capture and industrial processes at ambient and 50-60 °C, respectively.

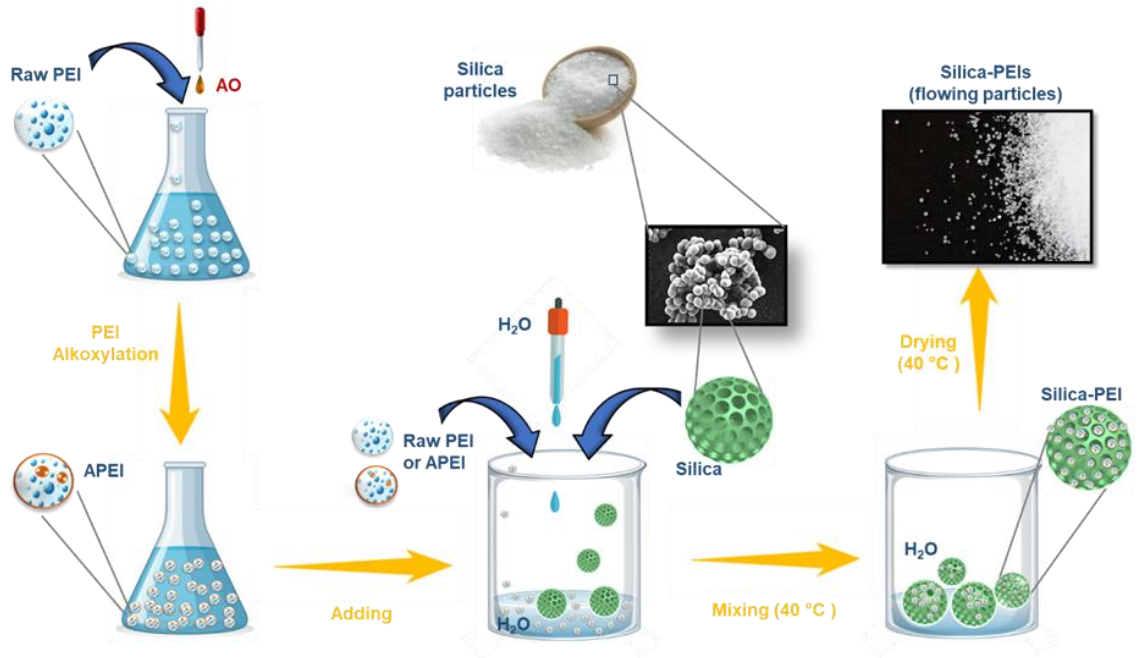

Figure 1. Schematic representation of silica-APEI preparation with the minimal quantity of water for mixing silica and APEI.

Keywords: Silica-PEI; Organic solvent-free, CO_2 capture.

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