CO_2 Sorption Accelerated by Mechanical Grinding.

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Significance and Relevance

The most widely used CO_2 capture technologies are based on adsorption technology. Although the use of amine solutions is a commercially successful technology, an attractive alternative is solid adsorption processes. Solid-state reactions of alkylamine to carbamate have been already investigated, but the kinetic of the reaction is slow at room temperature for ambient CO_2 reaching full conversion only after 1000 h. In this study the kinetics of conversion of the amine to ammonium-carbamate in ambient CO_2 is strongly accelerated by ball milling with full conversion achieved after only 4h.

Preferred and 2nd choice for the topic: CO_2 utilization and recycling ; Green chemistry and biomass transformation, renewable resources conversion
Preferred presentation: (Oral only)

Introduction and Motivations

The reaction of ammonia with CO_2 to give ammonium carbamate is a well-known process,[1] which in the last two decades was proposed as a process for CO_2 capture.[2] The most widely employed sorbents for CO_2 capture are aqueous solutions of organic amines, (mainly mono ethanolamine), which can selectively form alkylammonium carbamates.[3]

Solid-state reactions of alkylamine surfactants to ammonium carbamates were thoroughly investigated by Belman and coworkers, also showing the relevance of this reaction to nanocrystal synthesis.[4] These studies showed complete alkylammonium carbamate formation, which is also fast for high CO_2 concentrations. However, the kinetics of carbamate formations are slow at room temperature for ambient CO_2. The aim of this work is to explore the ability of mechanochemistry to accelerate the CO_2 capture from ambient air by a crystalline alkylamine surfactant.

Materials and Methods

Octadecylamine (ODA) was purchased by by Sigma Aldrich and used without any further purification. Conversion of ODA to the corresponding carbamate (OAOC), in different conditions, was studied by four different techniques: WAXD, FTIR, TGA, and DSC

Results and Discussion

This study explores possible procedures to accelerate CO_2 capture from ambient air by a crystalline alkylamine surfactant (ODA), leading to the corresponding crystalline ammonium-carbamate. Kinetics of reactions of crystalline ODA with CO_2, leading to OAOC, can be easily obtained by DSC scans, by evaluating melting enthalpies of residual ODA. The kinetics of conversion of the amine to ammonium-carbamate in ambient CO_2 is strongly accelerated by ball milling with full conversion achieved after only 4h, while the reaction kinetics of ODA powder simply exposed to ambient CO_2 is much slower, with full conversion only after nearly 1000h. A substantial increase in kinetics of the solid-state ODA reaction with ambient CO_2 can be also achieved by increasing the temperature up to 50°C, However, the time for full conversion remains much higher than for room temperature ball-milled ODA powder (roughly 100h vs 4h). It is apparent that ball-milling is an effective procedure to accelerate the reaction of solid amines with low-concentration CO_2.

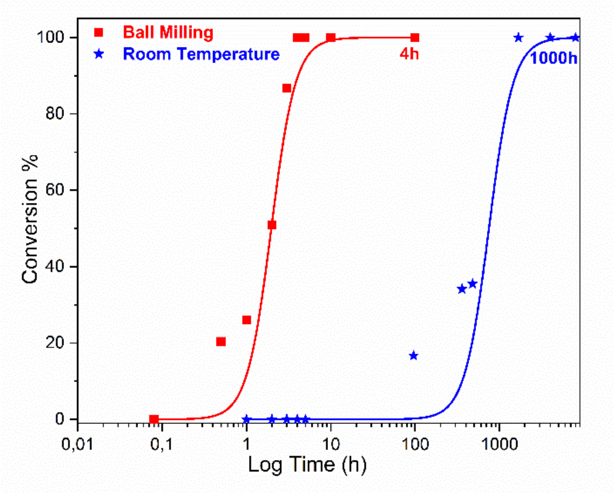

Figure 1 . Kinetics of reactions of crystalline ODA with CO_2 leading to OAOC, as evaluated by DSC scans, from melting enthalpies of residual ODA. All reactions are with ambient CO2: Red line and squares by exposure to air at room temperature; blu line and stars by ball-milling at 100 rpm at room temperature.

References

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