THEME

☐ Theme 1 : Synthesis and shaping
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Valorisation of Greenhouse and Acid Gas by Low-Silica Zeolite Catalyst

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The increasing emissions of greenhouse gases such as CO_2 into the atmosphere demand mature technologies to convert CO_2 into value-added compounds. Refineries and petrochemical industries emit 1.24 Pty of CO_2 and process large quantities of hydrogen sulfide (H_2S). The admixture of H_2S with CO_2 is called "acid gas".[1] No existing technologies allow simultaneous reduction of CO_2 and H_2S. A new technology aims to electrify simultaneous conversion of acid gas components into platform molecule carbonyl sulfide COS, which is further converted into CO and marketable Sulphur.[2] The COS formation in continuous mode from CO_2 and H_2S is the first and most important stage of this process ($H_2S(g) + CO_2(g) \rightleftharpoons COS(g) + H_2O(l)$). Zeolites catalyze this reaction via dissociative adsorption, leading to over 70% conversion of H_2S. The benchmark catalyst for this reaction is zeolite NaX[3], yet the literature lacks devoted efforts to optimize this reaction. In this work, we explore different zeolites for COS formation, aiming to develop a catalyst that gives high COS yields per pass and low energy demand for regeneration. A fixed-bed quartz reactor was used for this reaction and four zeolites in two states were used (i) fully dry and (ii) water-saturated.

Table 1. Properties of zeolite catalysts employed in this work.

Material	Si/Al ratio	S_{BET}(m^2/g)	Density (g/ml)	Hydration (%)
13X	1.2	900	0.65	24
4A	0.97	400	0.4	20.6
Y	2.5	700	-	23.8
13X-K (potassium-exchanged 13X)	1.2	900	0.65	21.5

The COS formation is minimum on zeolite Y amongst all dry zeolites. Despite having the same framework as that of zeolite Y, 13X has very high conversion followed by slower deactivation. Zeolite 4A outperforms all the catalysts, giving the maximum COS yield during the first hour of the reaction (Fig 1), followed by a deactivation faster than that of 13X and 13X-K. The activity of 13X-K is intermediate between 13X and 4A. The catalytic tests at a range of temperatures i.e., 45-350°C are carried out to study the effect of temperature. As water is one of the reaction products, zeolite deactivates due to the poisoning of the active sites by water[4], which was confirmed in this work by the negligible activity on water-saturated zeolites. Therefore, we systematically evaluate the hydration level of these zeolites and its role in COS formation.

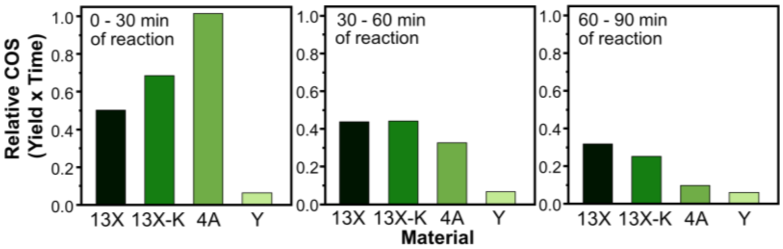

Fig 1. Relative COS formed at 100°C over dry zeolites at different times on stream (TOS).

References

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