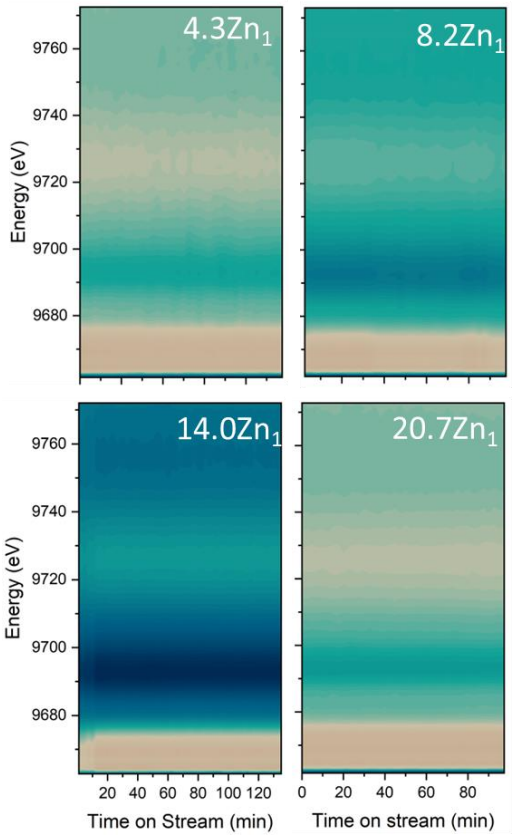
Proposal Code	**CH-6274**
Proposal Title	Unraveling the site density effect of single-atom Zn1/Al2O3-x catalysts as a potential candidate for the hydrogenation of CO2 to methanol and dimethyl ether

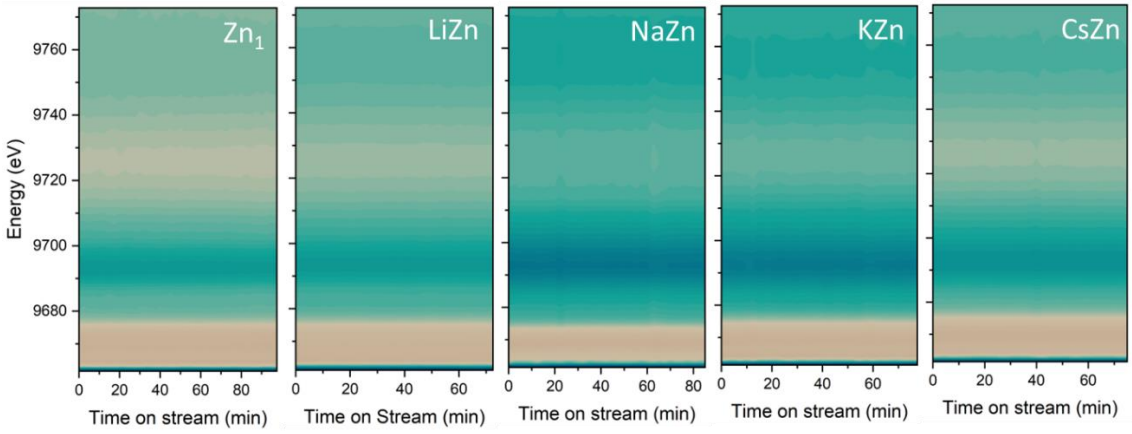
In this experiment, three systems were studied.

The first system, which is the main part of the experiment. Namely, the study of the Zn single-atom density effect. For four samples with different loading densities, 4Zn, 8Zn, 14Zn and 20Zn, we investigated the variation of Zn species with time under a. realistic CO_2 hydrogenation conditions; b. different reaction temperatures; c. different reaction pressures and d. different feedstock atmospheres, using operando methods. Zinc aluminate was used as a comparison sample and tested under operando conditions. Due to the high H-jump values of some of the samples, BN was used to dilute the samples. A part of the data is shown below. the Zn loading density influences the electronic state and coordination environment of Zn in high-temperature and high-pressure reactions, which may be an important reason for the different selectivity. More detailed data analysis and discussion is still in progress.

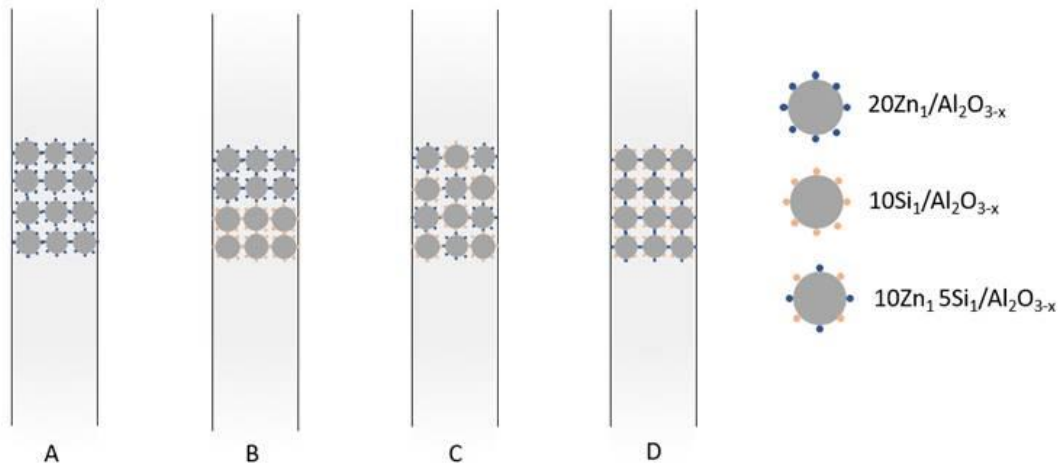


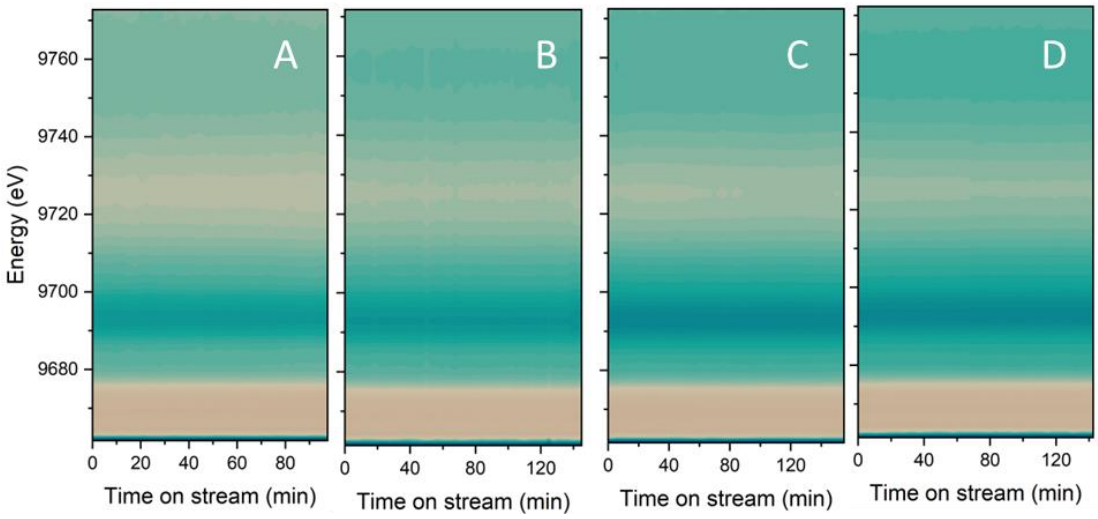
In the second system, four different alkali metal ions, Li, Na, K and Cs, were added to the 20Zn SACs as promoters. Of these, three catalysts, LiZn, NaZn and KZn, increased the methanol yield to varying degrees. Again, these four catalysts were studied under operando conditions and the effects of reaction temperature and atmosphere on the XANES and EXAFS spectra of the Zn species were also investigated. Some data is shown in the figure below. Under realistic

reaction conditions, the addition of alkali metals alters the electronic structure of Zn, to varying degrees. This would result in a change in the adsorption energy of the different carbon species. A more in-depth analysis of the data is in progress.



In the third system, we have designed a Si, Zn tandem single atom catalyst which converts CO2 to methanol and then propylene in a single catalyst bed. As shown in the figure below, we have investigated the effect of the different spatial positions of the two active components, Si and Zn, on the stability of the catalysts. Preliminary operando XAS results show that the hydrocarbon pool formed near Si, when the Zn and Si are in proximity, will change the way in which the Zn species are present in the reaction. This tendency gradually increases with time, leading to deactivation of the catalyst. More data analysis will be discussed in detail in a future article.





With the help of the beam scientists, our experiments went well. The quality of the data obtained was high. The results of the experiment were reliable. However, because of the large amount of data, more valuable data analysis will be presented in the future papers.