SUPPORTING INFORMATION

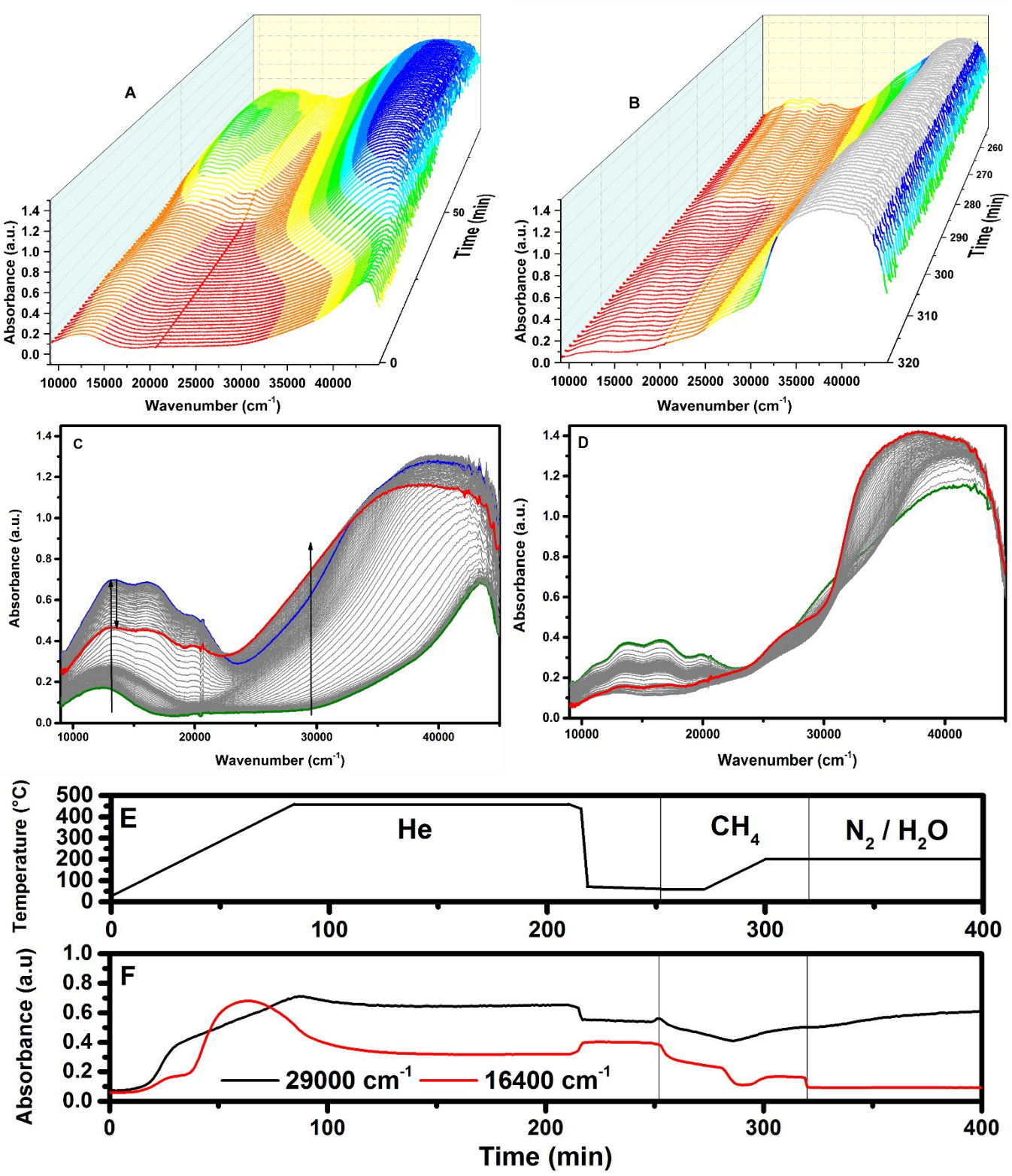

Figure S1. Operando UV-vis spectra of Cu-1.21-Na catalysts, a measurement was taken every minute, the arrow represents the time evolution A) and C) during He activation from RT to 450 °C (1 scan during every 5 °C) B) and D) during CH_4 addition from 60 to 200 °C (68 min or 68 scans are shown), arrows indicate the time evolution, the green spectrum is the starting point of the experiment, while the red spectrum is the endpoint. E) shows the temperature versus time for the operando experiment, while F) shows the development of the d-d band (16400 cm^{-1}) as well as the development of the 29000 cm^{-1} band versus time.

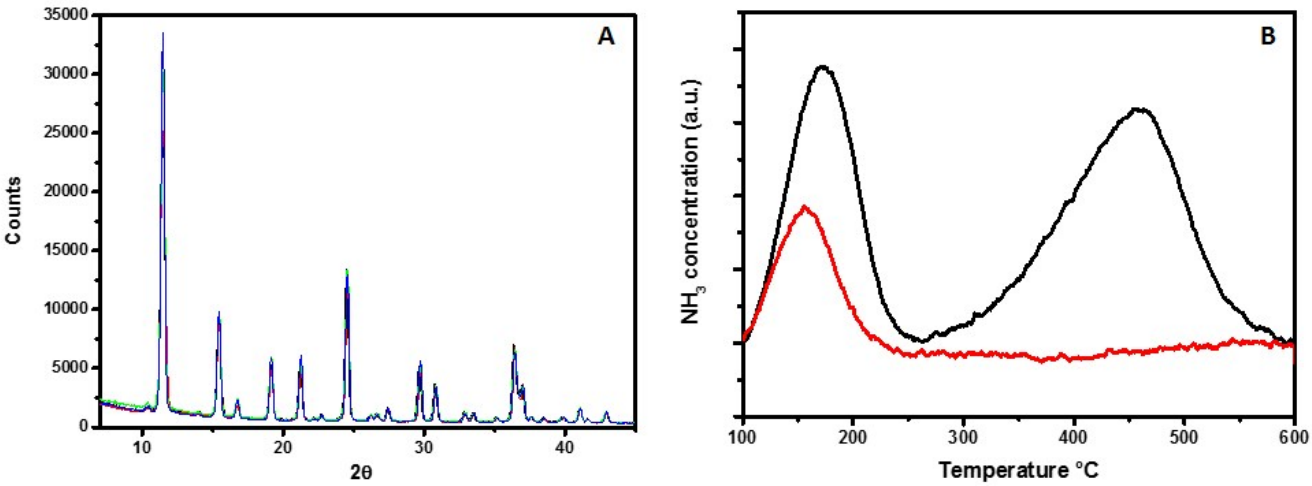

Figure S2. A**)** XRD patterns of the starting H-SSZ-13 material (black), Na-SSZ-13 (red), Cu-1.14-H (green) and Cu-0.91-Na (blue). Note Co k-alpha radiation (1.7XX A) was used. B) NH_3-TPD plots of H-SSZ-13 zeolite (black) and Na-SSZ-13 zeolite (red) before performing the Cu ion exchange.

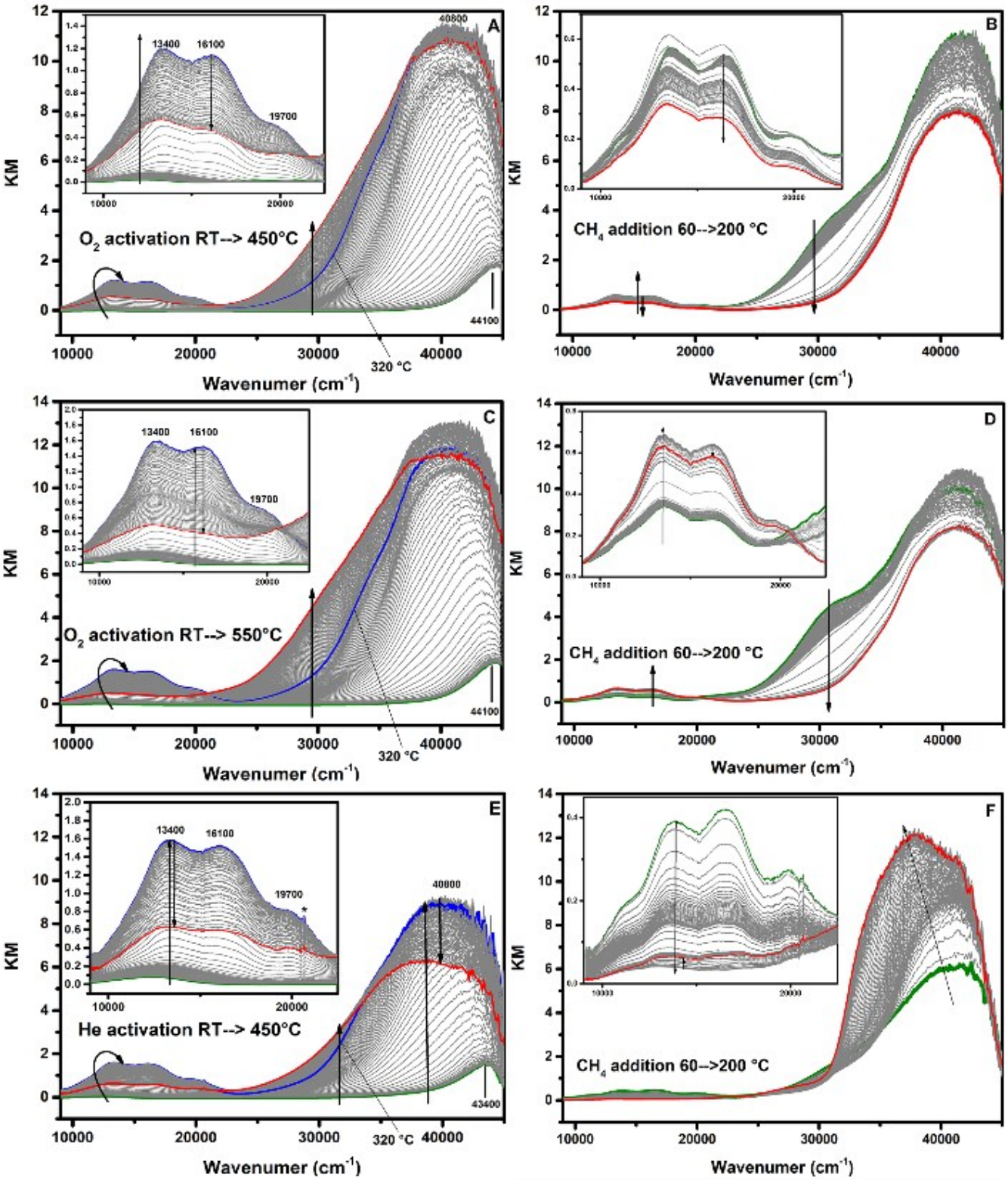

Figure S3. *Operando* UV-vis spectra of Cu-1.21-Na catalysts, a measurement was taken every minute, the arrow represents the time evolution A) during O_2 activation from RT to 450°C (1 scan during every 5 °C), the inset shows a zoom in on the Cu d-d region B) during CH_4 addition from at 60 to 200 °C (68 min or 68 scans are shown), arrows indicate the time evolution, the inset shows a zoom in on the Cu d-d region. C) and D), same catalyst and conditions but for 550 °C oxidation temperature. E) and F), same conditions and catalyst as A and B, but using He instead of O_2. * represents an artifact is the measurement.

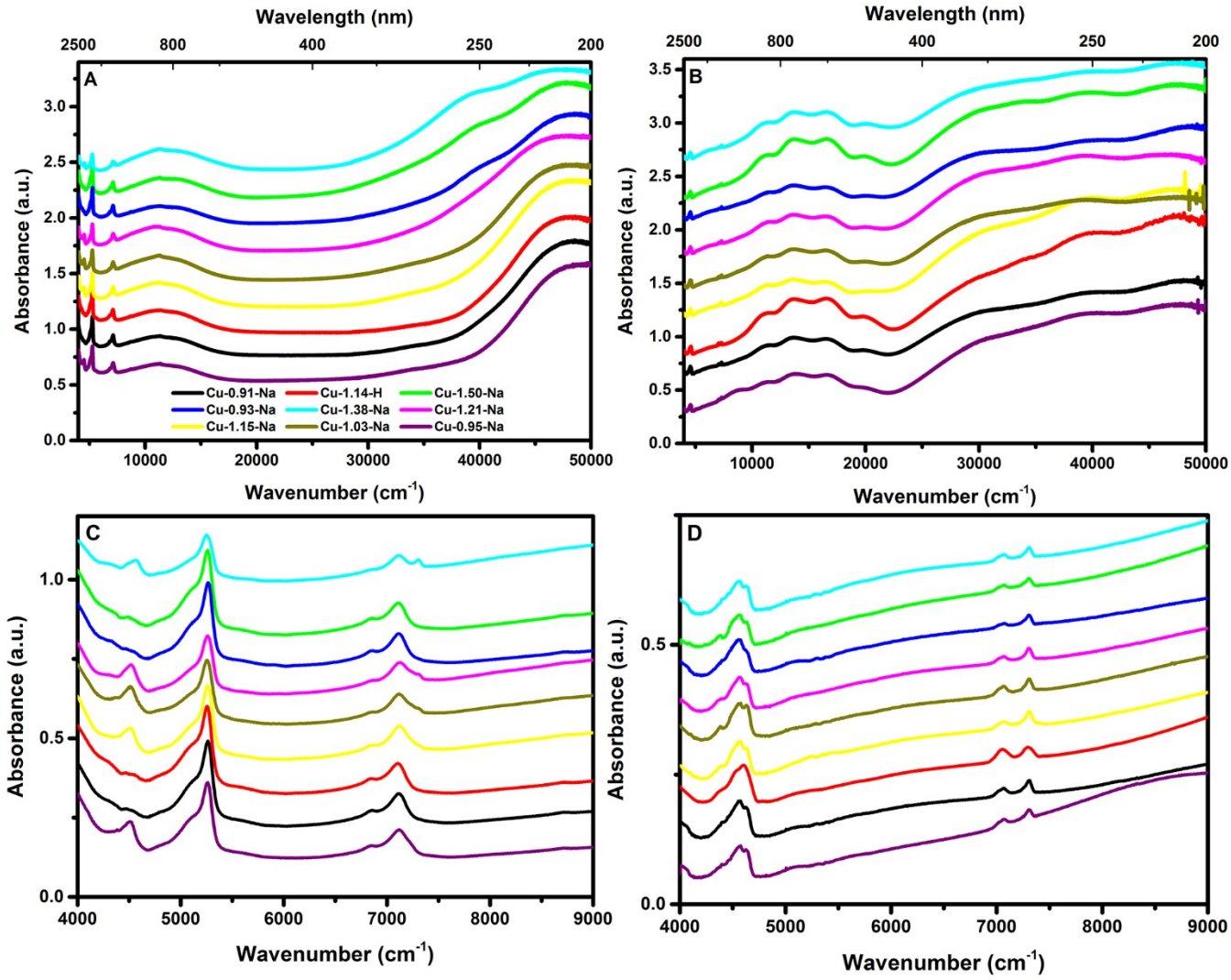

Figure S4. A) UV-vis-NIR DRS spectra of Cu-SSZ-13 in hydrated form. B) UV-vis-NIR DRS spectra of Cu-SSZ-13 after dehydration using dry O_2 at 450 °C for 2h. C) and D) NIR zoom in of the UV-vis-NIR DRS spectra of the Cu-SSZ-13 catalysts, C) hydrated spectra, D) dehydrated spectra.

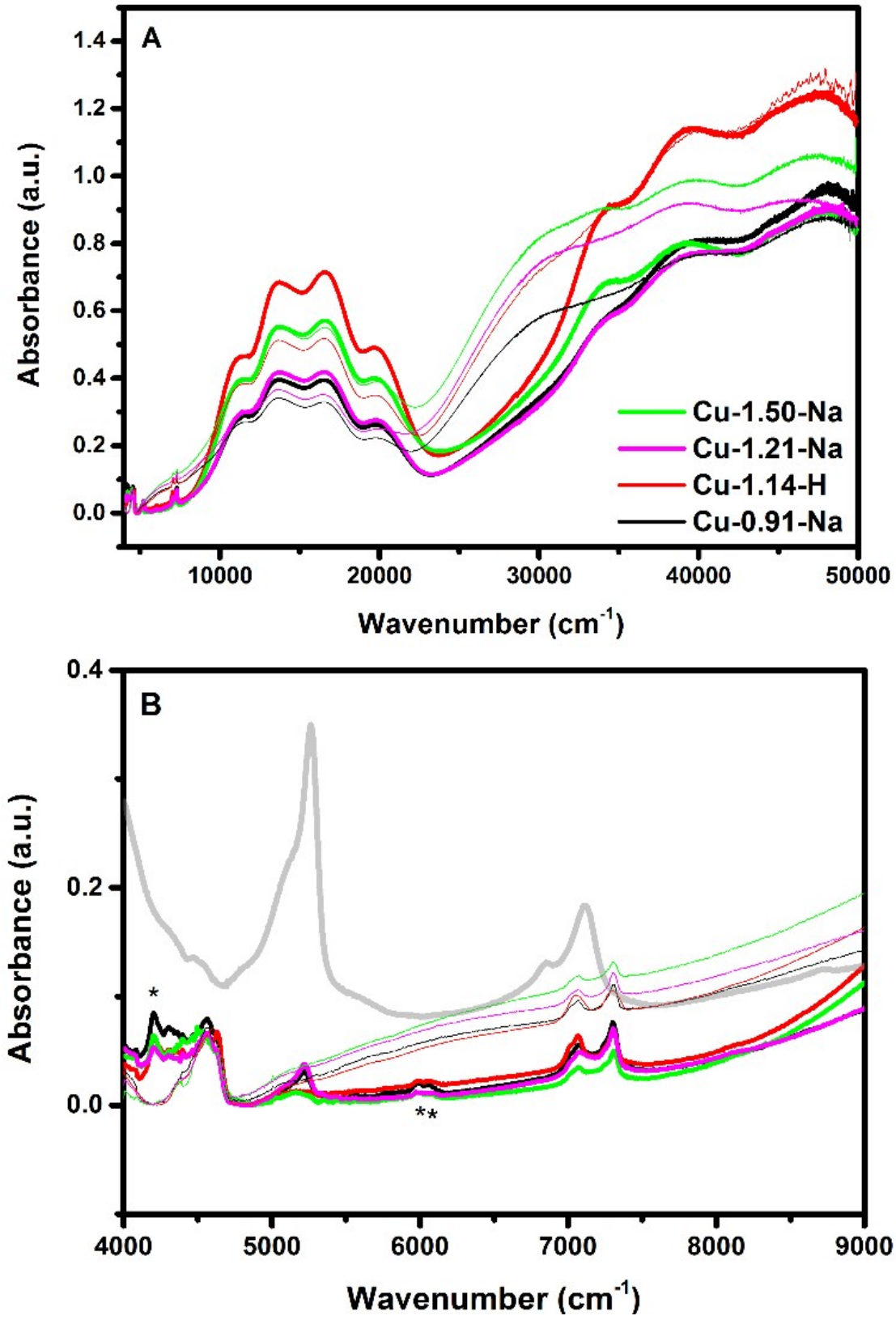

Figure S5. A) **Bold** lines UV-vis-NIR DRS spectra of Cu-SSZ-13 after dehydration using dry O_2 at 450 °C for 2h, thin lines after further addition of CH_4 at 200 °C for 1h. B) Near Infrared (NIR) region of the spectra after CH_4 addition at 200 °C for 1 h. Thin lines; before CH_4 addition, but after O2 activation; **Bold** after CH_4 addition. Bands designated * are due to the CH_3 symmetrical and anti-symmetrical overtone vibrations, showing that at this point in the reaction, CH_3 is present in the Cu- catalysts. As a reference one spectrum was added before CH_4 addition (Grey). See Table 3 for the assignments of all NIR bands.

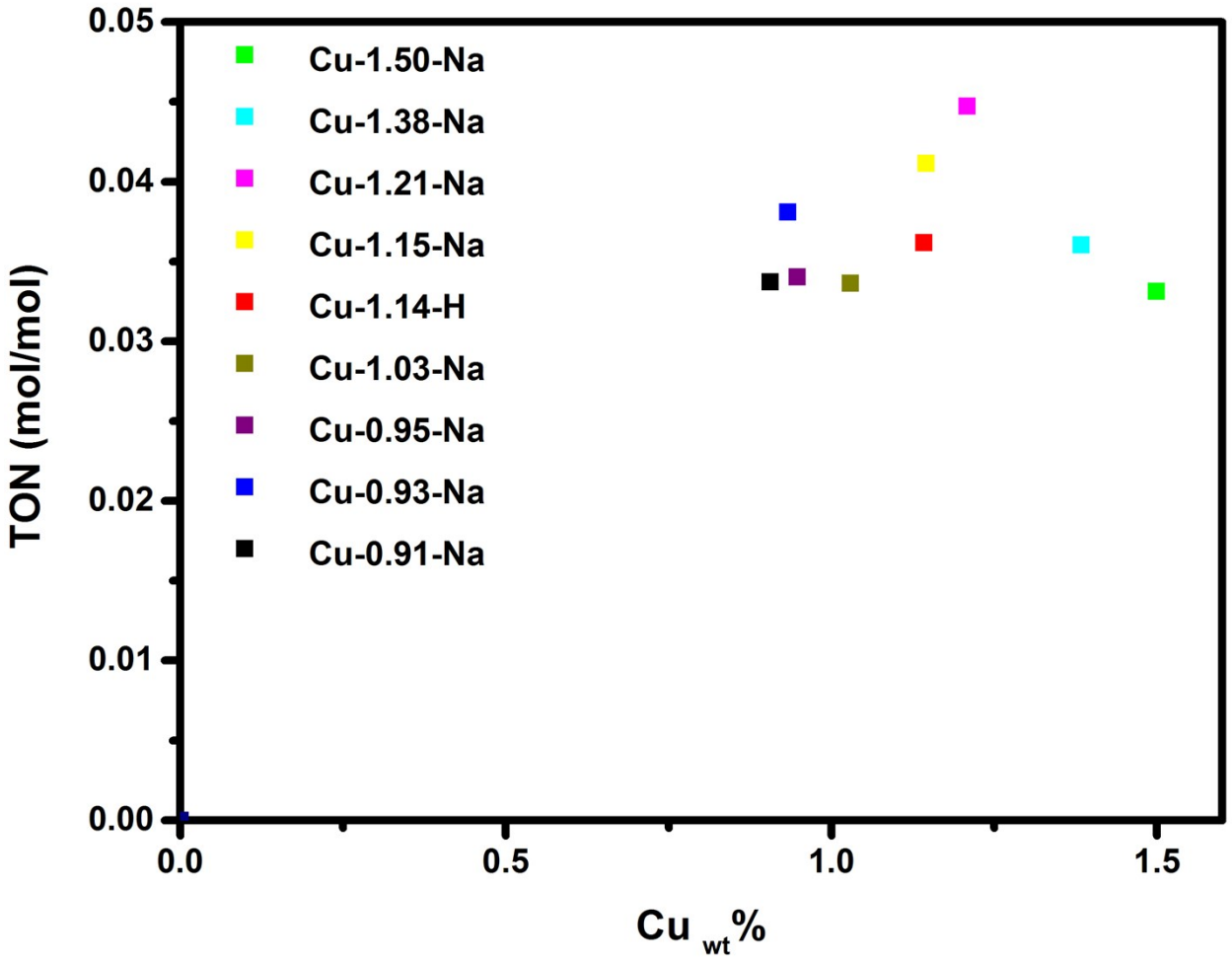

Figure S6. TON for the methane-to-methanol activation in mol methanol per mol copper for the Cu-exchanged zeolites using an oxidation temperature of 450 °C.