Cu nanoparticles CuNP were deposited on high surface area mesoporous silica SBA-15. Using Cu citrate as Cu source, calcination as *thick layer* or *thin layer* resulted in two different types of oxidic precursors for catalysts exhibiting the same Cu loading. Structural characterization was achieved employing N_2 physisorption, DR-UV/Vis spectroscopy, XRD, and XAS. *Thin layer precursors* possessed well dispersed amorphous CuO_x particles, while *thick layer precursors* possessed more ordered and less dispersed CuO_x particles. The mesoporous structure of the support was retained. TPR measurements during activation of catalysts revealed various reducibilities of CuO_x particles depending on calcination mode and Cu loading.

Various CuNP present on SBA-15 were found after activation and during methanol steam reforming. The activated *thin layer catalysts* consisted of smaller and more disordered CuNP. Independent of Cu loading similar Cu metal particles were found. *Thick layer catalysts* exhibited more ordered CuNP increasing in size with increasing Cu content. The Cu surface areas agreed with results from XRD and XAS. In methanol steam reforming (MSR), *thin layer catalysts* showed higher H_2 formation rates and higher H_2 TOFs compared to the corresponding *thick layer catalysts*. Catalytic activity correlated with both, degree of disorder and size of CuNP.

Redox behavior of Cu/SBA-15 catalysts was tested by applying either temporary O_2 addition to MSR feed or redox activation (RA). After both procedures, increased H_2 formation rates of all Cu/SBA-15 catalysts were observed. Additionally, *thick layer catalysts* showed increased H_2 TOFs after RA. Partial or complete oxidation of CuNP and subsequent re-reduction did not lead to increased sizes of CuNP.