## Bimetallic Copper- and Nickel-Rich Cu-Ni Phyllosilicate Catalysts for the Liquid-Phase Selective Hydrogenation of Furfural to Furfuryl Alcohol

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**Fig. S1** XRD patterns of monometallic 20%NiPS and bimetallic Cu-NiPS catalysts reduced at 600 °C compared to the PDF card of metallic Ni<sup>0</sup> and Cu<sup>0</sup>.

Fig. S1 exhibites XRD patterns of monometallic 20%NiPS and bimetallic Cu-NiPS catalysts reduced at 600 °C compared to the PDF card of metallic Ni<sup>0</sup> and Cu<sup>0</sup>. Since the metal phyllosilicate structure consisted of the octahedral layer of metal and tetrahedral layer of SiO<sub>2</sub>, the low crystallinity with small crystallite size of metal ( $d_p < 5.0$  nm) effect to broad diffraction peaks of XRD. However, the XRD patterns of all reduced catalysts were corresponding well with PDF cards of metallic Ni<sup>0</sup> and metallic Cu<sup>0</sup>.



Fig. S2 HR-TEM of monometallic 20%NiPS and bimetallic 15%Cu-5%NiPS catalysts

To clarify the lattice patterns of Ni and Cu metals, the lattice patterns in two types of catalysts: a monometallic 20% NiPS catalyst and a bimetallic 15%Cu-5%NiPS catalyst were determined by High-Resolution Transmission Electron Microscopy (HR-TEM) with the results illustrated in **Fig. S2**. The lattice spacing (d-spacing) of both samples were measured around 30 points for each sample. The d-spacing for Ni in the monometallic 20% NiPS catalyst was found to be 0.204  $\pm$  0.004 nm, which aligns closely with the expected value noted in the PDF file (Ni-PDF: 00-004-0850). Furthermore, for the bimetallic 15%Cu-5%NiPS catalyst, the lattice spacing for the NiCu alloy was measured at 0.206  $\pm$  0.007 nm. This value shows a slight shift from the expected d-spacing for pure Ni (PDF: 00-004-0850) and Cu (PDF: 00-004-0836). These results confirm the formation of a NiCu alloy in the bimetallic catalyst, highlighting the interactions between Ni and Cu at the atomic level.



Fig. S3  $N_2$  adsorption-desorption isotherms of monometallic 20% NiPS and bimetallic Cu-NiPS catalysts



**Fig. S4** XPS spectra of Cu 2p for reduced bimetallic Cu-NiPS catalysts compared with monometallic 20%CuPS catalyst (did not use in this study).



Fig. S5 The selectivity to THFA for all catalysts



Fig. S6 XPS spectra of Ni 2p, Cu 2p, and C 1s of fresh and spent 15%Cu-5%NiPS catalysts

Catalysts	<b>B.E.</b> (eV)		% Atomic Concentration			
	Ni 2p	Cu 2p	Cu/Si	Ni/Si	C/Si	(Cu+Ni)/Si
15%Cu-5%NiPS-fresh	855.1	933.1	0.22	0.02	-	0.24
15%Cu-5%NiPS-spent	856.7	933.8	0.16	0.05	0.53	0.21

Table S1 Surface compositions of fresh and spent of 15% Cu-5% NiPS catalysts

Note: The Ag 3d at B.E. 368.2 eV is used as a standard to determine carbon on the surface