

Supplementary Information

Increasing Al-Pair Abundance in SSZ-13 Zeolite via Zeolite Synthesis in the Presence of Alkaline Earth Metal Hydroxide Produces Hydrothermally Stable Co-, Cu- and Pd-SSZ-13 Materials

Konstantin Khivantsev ^{1,*†}, Miroslaw A. Derewinski ^{1,2,*†}, Libor Kovarik ¹, Mark Bowden ¹, Xiaohong Shari Li ¹, Nicholas R. Jaegers ¹, Daria Boglaienko ¹, Xavier I. Pereira-Hernandez ¹, Carolyn Pearce ¹, Yong Wang ^{1,3} and Janos Szanyi ^{1,*}

¹ Pacific Northwest National Laboratory, Richland, WA 99352, USA; libor.kovarik@pnnl.gov (L.K.); mark.bowden@pnnl.gov (M.B.); xiaohong.li@pnnl.gov (X.S.L.); njaegeers@berkeley.edu (N.R.J.); daria.boglaienko@pnnl.gov (D.B.); vruatsa@gmail.com (X.I.P.-H.); carolyn.pearce@pnnl.gov (C.P.); wang42@wsu.edu (Y.W.)

² J. Haber Institute of Catalysis and Surface Chemistry, Polish Academy of Sciences, 30-239 Krakow, Poland

³ The Gene and Linda Voiland School of Chemical Engineering and Bioengineering, Washington State University, Pullman, WA 99164, USA

* Correspondence: konstantin.khivantsev@pnnl.gov (K.K.); ncderewi@cyf-kr.edu.pl (M.A.D.); janos.szanyi@pnnl.gov (J.S.)

† These authors contributed equally to this work.

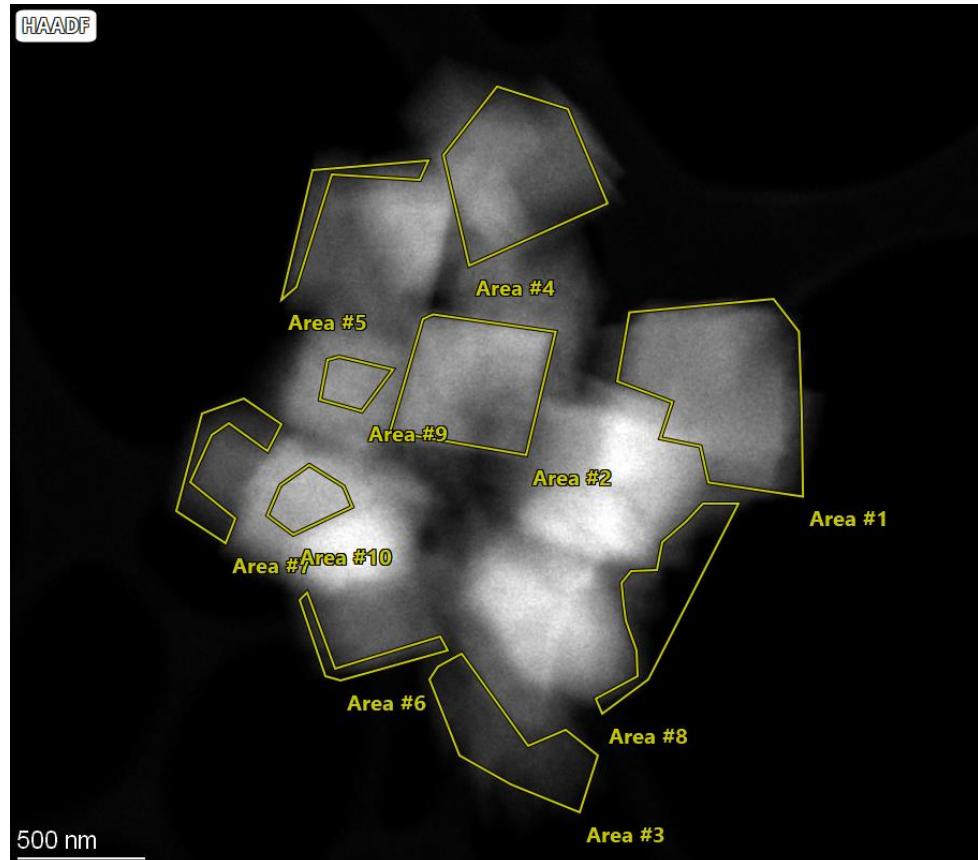


Figure S1. EDS mapping chosen on different areas of H-SSZ-13 (Sr) crystals. The corresponding tabulated values are shown in Table S1.

Table S1. Atomic and weight percentages of Si, Al, and O EDS mapped in Fig. 1.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	65.00	4.02	51.50	4.43	0.84
13	Al	K	3.02	0.61	4.03	0.84	0.82
14	Si	K	31.98	4.19	44.47	4.76	0.97

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #1.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	65.30	3.97	51.83	4.39	0.40
13	Al	K	3.22	0.65	4.31	0.89	0.29
14	Si	K	31.48	4.15	43.86	4.74	0.52

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #2.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	69.00	3.75	56.00	4.33	0.55
13	Al	K	2.85	0.57	3.90	0.80	1.05
14	Si	K	28.14	3.89	40.09	4.62	0.16

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #3.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	69.28	3.76	56.32	4.36	0.58
13	Al	K	2.85	0.58	3.90	0.82	4.38
14	Si	K	27.87	3.89	39.78	4.64	2.35

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #6.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	64.99	3.97	51.50	4.37	0.27
13	Al	K	3.41	0.68	4.55	0.94	0.48
14	Si	K	31.60	4.15	43.95	4.75	0.12

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #4.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	66.96	3.82	53.70	4.31	0.77
13	Al	K	3.57	0.71	4.83	0.99	1.61
14	Si	K	29.47	4.00	41.48	4.68	0.46

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #5.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	68.61	3.82	55.54	4.39	0.68
13	Al	K	2.60	0.53	3.55	0.74	3.10
14	Si	K	28.78	3.95	40.90	4.66	1.22

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #7.

Z	Element	Family	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)	Fit Error (%)
8	O	K	65.59	3.96	52.15	4.39	0.57
13	Al	K	3.16	0.63	4.23	0.88	1.76
14	Si	K	31.25	4.13	43.62	4.74	0.50

2023-11-04T22:09:29-0700 Analysis of spectrum: Spectra from Area #10.

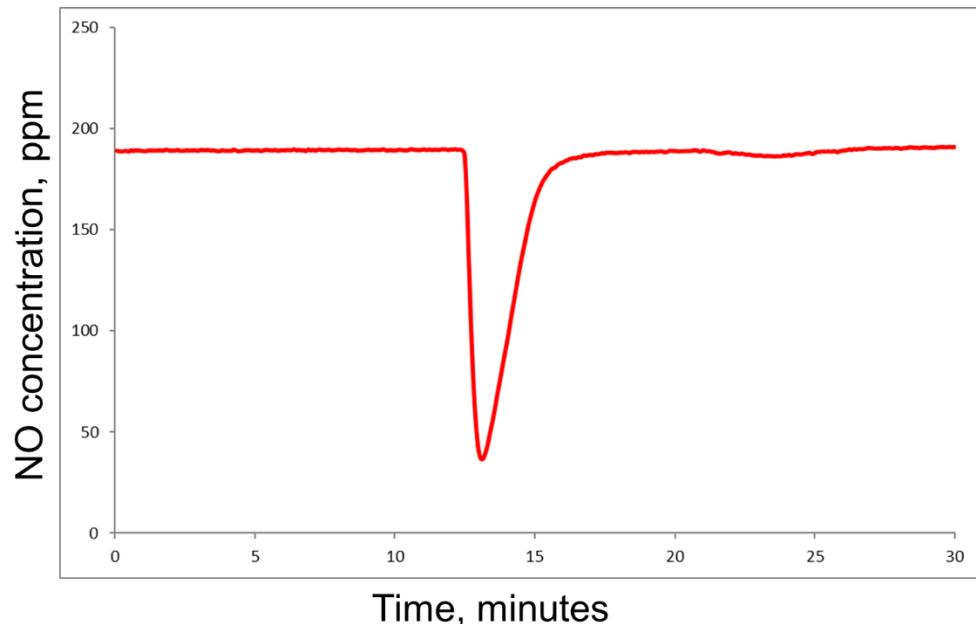


Figure S2. NOx adsorption on Co-SSZ-13 sample (derived from NaOH and hydrothermally aged at 870 °C) at 100 °C for 20 min (after 10 min bypass). The feed gas mixture contains 220 ppm of NOx, 14% O₂ and 300 ppm H₂O. Sample mass 120 mg. Total flow rate 300 sscm/min. It is obvious that only a relatively small uptake of NO is observed, consistent with much lower hydrothermal stability of Co-SSZ-13 (Na) sample. If the structure were preserved, no loss of activity would be observed.

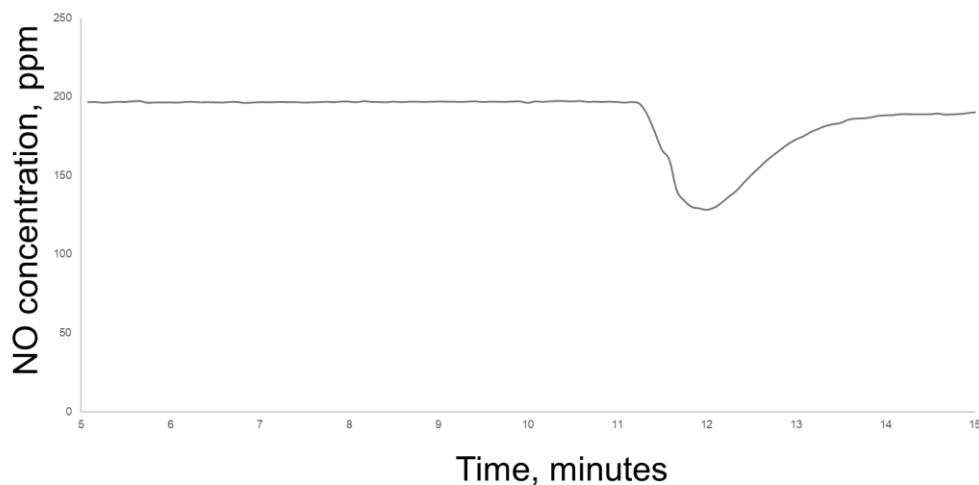


Figure S3. NOx adsorption on 850 °C hydrothermally aged 1.7 wt% Pd/Co-SSZ-13 sample derived from NaOH synthetic route at 100 °C for 5 min (after 10 min bypass). The feed gas mixture contains 220 ppm of NOx, 250 ppm CO, 14% O₂ and 3% H₂O. Sample mass 120 mg. Total flow rate 300 sscm/min. Rather small uptake of NO occurs, consistent with material degradation.

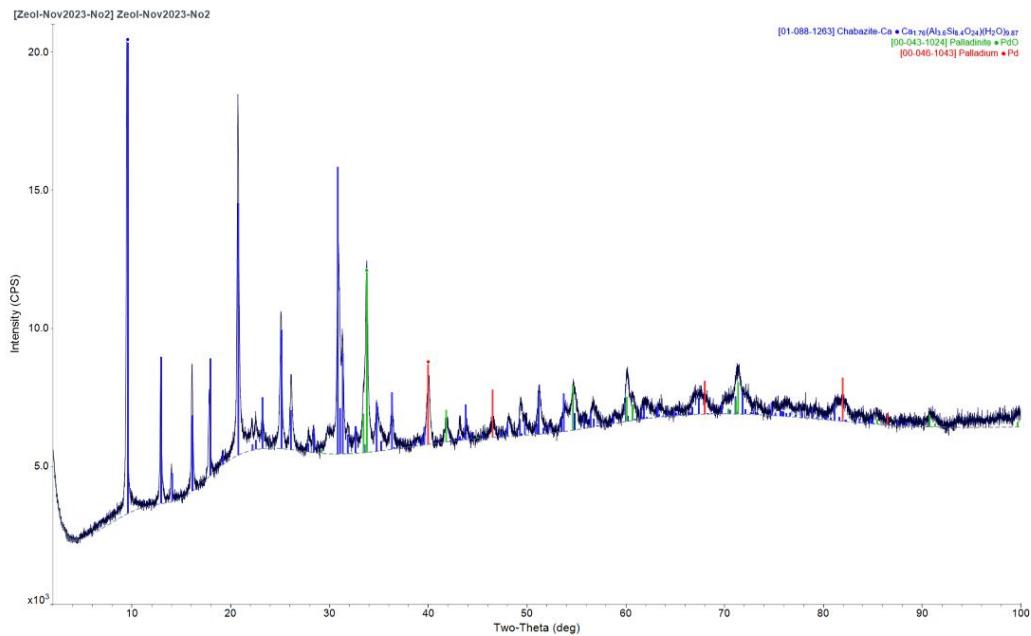


Figure S4. XRD pattern of Pd/SSZ-13 (Sr) sample hydrothermally aged at 930 °C. Blue ticks indicate the SSZ-13 crystalline phase reflections. Red ticks indicate the Pd metal presence. Green ticks indicate PdO presence.

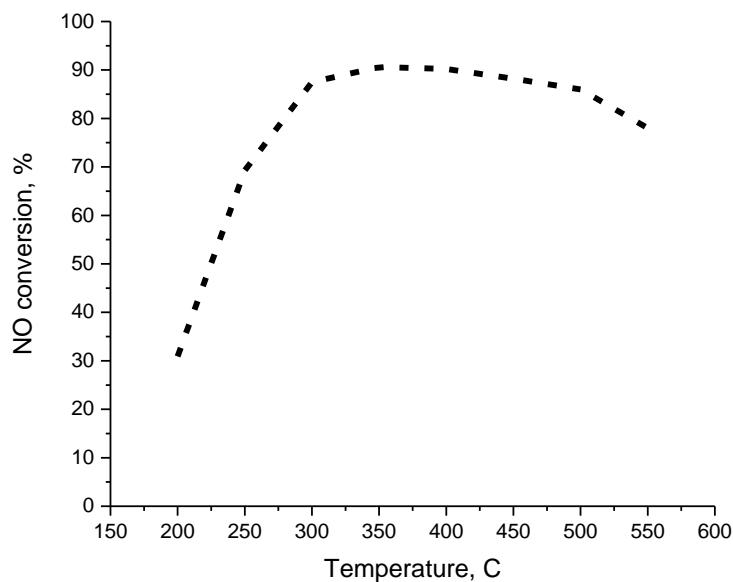


Figure S5. NO conversion vs Temperature. Conditions: 120 mg, 2.5 % wt% Cu/SSZ-13(Sr), 360 ppm NH₃, 360 ppm NO, 14% O₂, 2.4 % H₂O and balanced with N₂. The total flow rate 300 ml/min. GHSV 150 L/g*hr. The sample was aged at 920 °C in air/ 10% H₂O flow for 3 hours before catalysis.