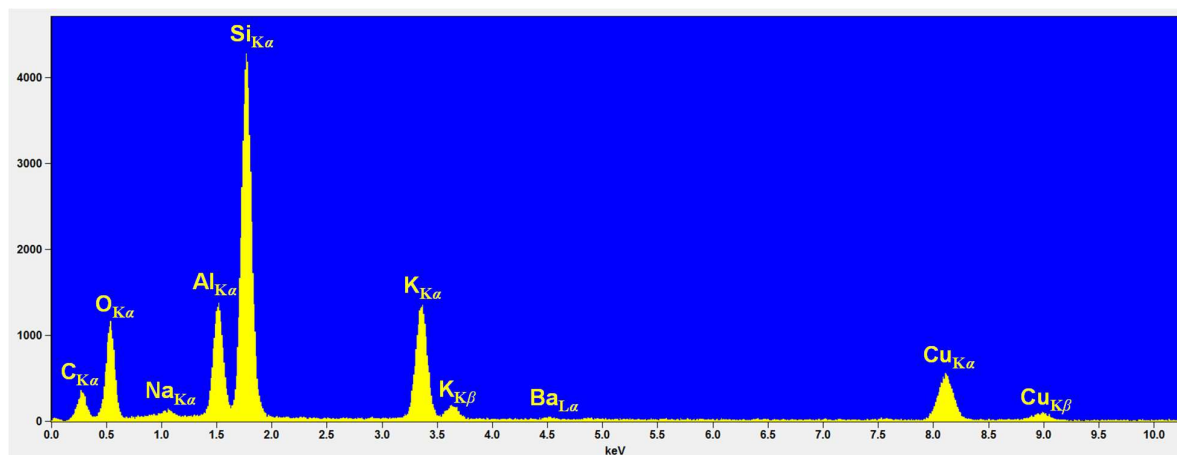
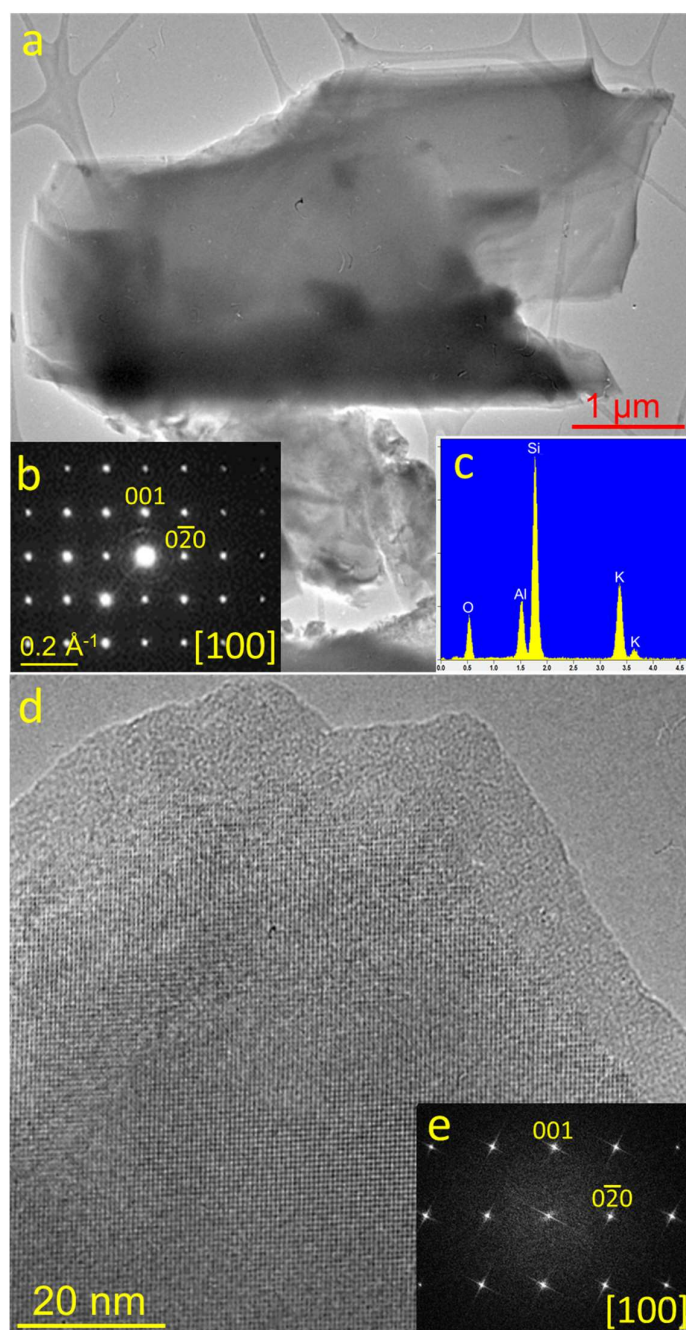


## Supplementary Materials



**Figure S1.** X-ray EDS spectrum from the adularia showing Si, Al, K, Na, and trace amount of Ba in the crystal. Cu peaks are from TEM Cu grid that holds the specimen.



**Figure S2.** The bright-field TEM image, SAED pattern, TEM-EDS, High-resolution TEM image with its FFT pattern of adularia at [100] zone-axis, showing no diffuse diffractions along  $b^*$ - direction. The adularia is a weathering product in North American Sub-Cambrian Paleosols ([1]).

**Table S1.** Experimental and crystallographic information of the adularia sample (100K).

	Host	Nano-phase	One phase
Crystal data			
Formula	KAlSi <sub>3</sub> O <sub>8</sub>	KNaAl <sub>2</sub> Si <sub>6</sub> O <sub>16</sub>	K <sub>0.9</sub> Na <sub>0.1</sub> AlSi <sub>3</sub> O <sub>8</sub>
<i>a</i> , <i>b</i> , <i>c</i> (Å)	8.5291(1)	8.5291(1)	
	12.9795(2)	12.9795(2)	
	7.2032(1)	7.2032(1)	
$\alpha$ , $\beta$ , $\gamma$ (°)	90	90	
	116.0521(7)	116.0521(7)	
	90	90	
V (Å <sup>3</sup> )	716.397(18)	716.397(18)	
Space group	<i>C2/m</i>	<i>Pa</i>	<i>C2/m</i>
Data collection			
Diffraction Source	Mo K $\alpha$	X-ray	Mo K $\alpha$ X-ray
Total reflections	16915		8567
Independent reflections	4359		1139
Observed [ <i>I</i> > 3 $\sigma$ ( <i>I</i> )]	2199		1109
<i>R</i> <sub>int</sub>	4.22		4.7
$\theta$ values (°)	1.57		3.09
	30.53		30.53
Range of <i>h</i> , <i>k</i> , <i>l</i>	<i>h</i> = -12→12		<i>h</i> = -12→12
	<i>k</i> = -18→18		<i>k</i> = -18→18
	<i>l</i> = -10→10		<i>l</i> = -10→10
Refinement			
Twin Percentage *	80%	11.4%, 8.6%	100%
wRobs (%)	3.56		5.09
wRall (%)	3.79		5.12
GOF (all)	1.2		2.33
GOF (obs)	1.64		2.35
No. of parameters	298		63
No. of constraints	65		25
$\Delta Q_{\max}$ , $\Delta Q_{\min}$ (eÅ <sup>-3</sup> )	0.33, -0.36		0.44, -0.33

\* The crystal contains 80% of end-member orthoclase and 20% of the nano-precipitates with *Pa* symmetry in twinning relationship (11.4% and 8.6% for each orientation).

**Table S2.** Fractional coordinates for the adularia average structure with *C2/m* symmetry (100K).

Label	Atom	Occ	x	y	z	U <sub>eq</sub>
M	K	0.9	0.28189(5)	0	0.13643(7)	0.01451(18)
	Na	0.1	0.28189(5)	0	0.13643(7)	0.01451(18)
T <sub>1</sub>	Si	0.5895	0.00914(5)	0.18368(3)	0.22418(5)	0.00925(17)
	Al	0.4105	0.00914(5)	0.18368(3)	0.22418(5)	0.00925(17)
T <sub>2</sub>	Si	0.9105	0.29366(5)	0.11748(3)	0.65551(5)	0.00881(17)
	Al	0.0895	0.29366(5)	0.11748(3)	0.65551(5)	0.00881(17)
Oa1	O	1	0	0.14426(10)	0	0.0158(5)
Oa2	O	1	0.6319(2)	0	0.2857(2)	0.0159(5)
Ob	O	1	0.17431(15)	0.14360(8)	0.77266(17)	0.0177(4)
Oc	O	1	0.46745(14)	0.18946(7)	0.74132(16)	0.0143(3)
Od	O	1	0.18304(14)	0.12537(7)	0.40745(15)	0.0131(3)

Note: See Supplementary cif file for the electronic file.

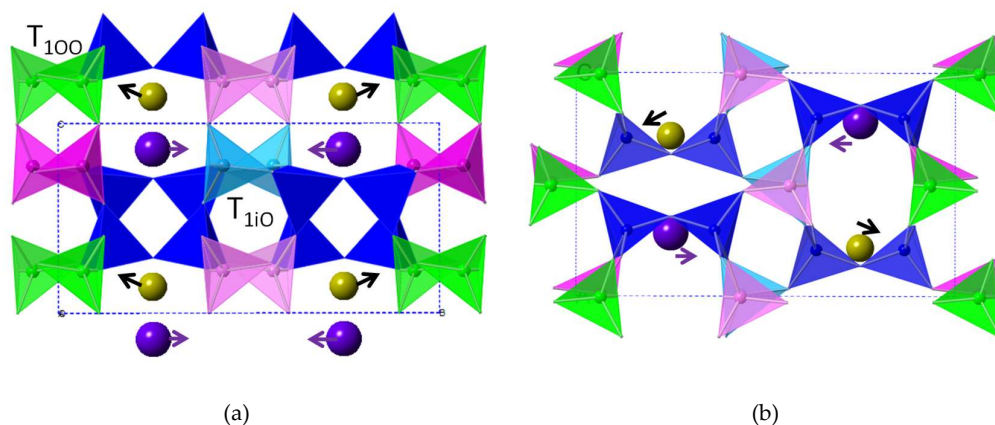
**Table S3.** Fractional coordinates for the nano-phase alkali feldspar with *Pa* symmetry (100K).

Label	Atom	Occ	x	y	z	U <sub>eq</sub>
Mo	Na	1	0.2789(13)	0.2494(4)	0.1413(13)	0.034(4)
M2	K	1	0.2124(10)	0.7498(2)	0.8648(10)	0.059(3)
T1oo	Si	0.5763	0.5095(10)	0.9335(5)	0.2272(9)	0.048(3)
	Al	0.4237				
T1io	Si	0.4032	0.9959(13)	0.4364(7)	0.7834(10)	0.057(4)
	Al	0.5968				
T1o2	Si	0.5763	0.4924(10)	0.9364(5)	0.7830(8)	0.045(3)
	Al	0.4237				
T1i2	Si	0.5763	0.0108(11)	0.4336(5)	0.2280(10)	0.055(3)
	Al	0.4237				
T2oo	Si	0.9237	0.8002(9)	0.8659(4)	0.6623(8)	0.047(3)
	Al	0.0763				
T2io	Si	0.9237	0.7063(10)	0.3675(5)	0.3473(8)	0.057(3)
	Al	0.0763				
T2o2	Si	0.9237	0.2075(8)	0.8678(4)	0.3479(7)	0.044(2)
	Al	0.0763				
T2i2	Si	0.9237	0.3002(11)	0.3648(5)	0.6624(9)	0.057(3)
	Al	0.0763				
Oa1_1	O	1	0.505(3)	0.8942(9)	0.001(2)	0.055(7)
Oa1_2	O	1	0.004(3)	0.3963(10)	0.999(3)	0.059(8)
Oa2o	O	1	0.632(3)	0.2495(7)	0.295(3)	0.041(7)
Oa22	O	1	0.904(6)	0.7497(7)	0.735(4)	0.120(18)
Oboo	O	1	0.664(2)	0.8978(11)	0.772(2)	0.054(7)
Obio	O	1	0.825(3)	0.4010(10)	0.235(2)	0.055(8)
Obo2	O	1	0.325(2)	0.8998(11)	0.235(2)	0.051(6)
Obi2	O	1	0.164(2)	0.3981(13)	0.771(2)	0.057(8)
Ocoo	O	1	0.950(2)	0.9394(13)	0.7454(17)	0.048(6)
Ocio	O	1	0.509(2)	0.4371(11)	0.259(2)	0.050(8)
Oco2	O	1	0.009(2)	0.9376(11)	0.258(2)	0.052(8)
Oci2	O	1	0.449(2)	0.4382(14)	0.746(2)	0.053(7)
Odoo	O	1	0.675(2)	0.8726(9)	0.4005(16)	0.039(5)
Odio	O	1	0.811(2)	0.3856(10)	0.573(3)	0.062(9)
Odo2	O	1	0.310(2)	0.8837(7)	0.571(3)	0.061(10)
Odi2	O	1	0.174(2)	0.3709(11)	0.397(2)	0.048(6)

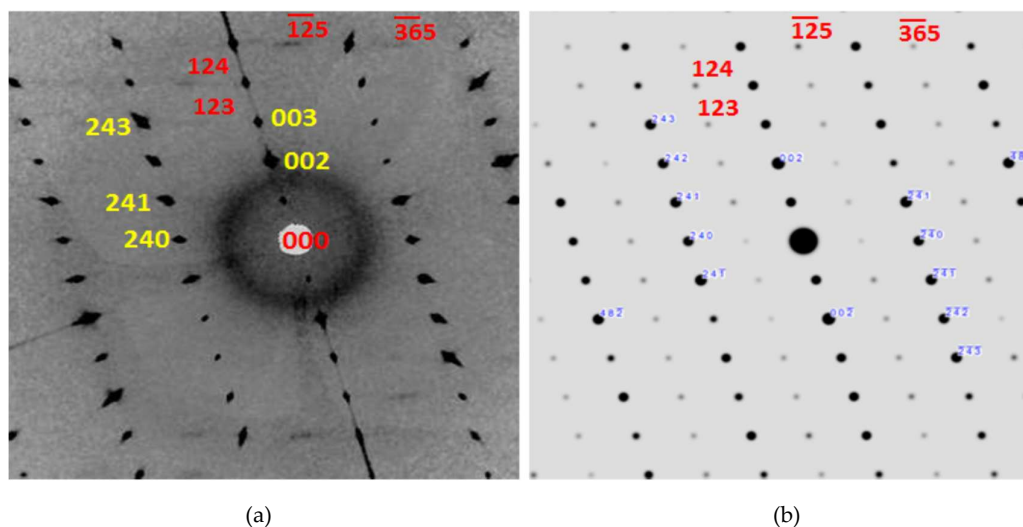
Note: See Supplementary cif file for the electronic file.

**Table S4.** Bond distances for the T sites.

	O1	O2	O3	O4	average
T1oo	1.6104	1.6622	1.6940	1.6955	1.6655
T1io	1.5815	1.6215	1.6496	1.7701	1.6557
T1o2	1.6211	1.6578	1.6880	1.6916	1.6646
T1i2	1.5567	1.6104	1.6695	1.7662	1.6507
T2oo	1.4865	1.6909	1.7165	1.7355	1.6574
T2io	1.4681	1.5982	1.6317	1.7730	1.6178
T2o2	1.4872	1.7104	1.7133	1.7180	1.6572
T2i2	1.4901	1.6086	1.6369	1.7642	1.6250
Bond distance in host <i>C2/m</i> structure					
T <sub>1</sub>	1.6579	1.6629	1.6636	1.6722	1.66415
T <sub>2</sub>	1.6163	1.6184	1.6275	1.6339	1.624025



**Figure S3.** Structural model of the nano-phase with  $Pa$  symmetry along  $a$ -axis (a) and  $c$ -axis (b), respectively. Arrows indicate shift directions of Na and K atoms with respect to the special position (i.e., K in  $C2/m$  structure). All  $T_2$  sites are in blue color.



**Figure S4.** Fig. [2-10] zone-axis precession image (a) and calculated SAED pattern (b) showing consistent intensities for the diffraction spots violating  $C$ -centering.

Note: Procedures for structure refinement

JANA2006 software is capable of refining multiple structures that are sharing the main reflections in the diffraction data ([2]). A .cif file was exported from the  $C2/m$  refinement with only K assigned to the M site. The symmetry of the .cif file was modified to  $Pa$  with VESTA software, quadrupling the number of tetrahedral sites and oxygen atoms while leaving the structure exactly the same as in  $C2/m$  structure. One of the M site was changed to Na atom. A new phase with stoichiometry of  $KNa(Si_6Al_2)O_{16}$  was added to the refinement in JANA2006 program by importing the modified .cif file. The proportion of the new phase is initially fixed to 5%. Only atom positions of the new phase were refined, with the occupancies of the tetrahedral sites kept the same as the  $C2/m$  phase. After the refinement converged, the proportion of the second phase was increased to 10%. The same process was repeated several times until the proportion of the second phase reaches 20%. Because the reflections from the second phase were too weak and diffuse, it took several attempts to make the refinement reach a stable final structure. Some parameters had to be fixed during the refinement that were relaxed at a later step. Detail parameters and conditions are listed in Table S1.

**Reference:**

1. Medaris Jr, L.G.; Driese, S.G.; Stinchcomb, G.E.; Fournelle, J.H.; Lee, S.; Xu, H.; DiPietro, L.; Gopon, P.; Stewart, E.K. Anatomy of a Sub-Cambrian Paleosol in Wisconsin: Mass fluxes of chemical weathering and climatic conditions in North America during Formation of the Cambrian Great Unconformity. *J. Geol.* **2018**, *126*, 261–283.
1. Topa, D.; Petříček, V.; Dušek, M.; Makovicky, E.; Balić-Zunić, T. Simultaneous refinement of two components of an exsolution intergrowth: Crystal structures of the lindströmite–krupkaite pair. *Can. Mineral.* **2008**, *46*, 525–539.