

ESI

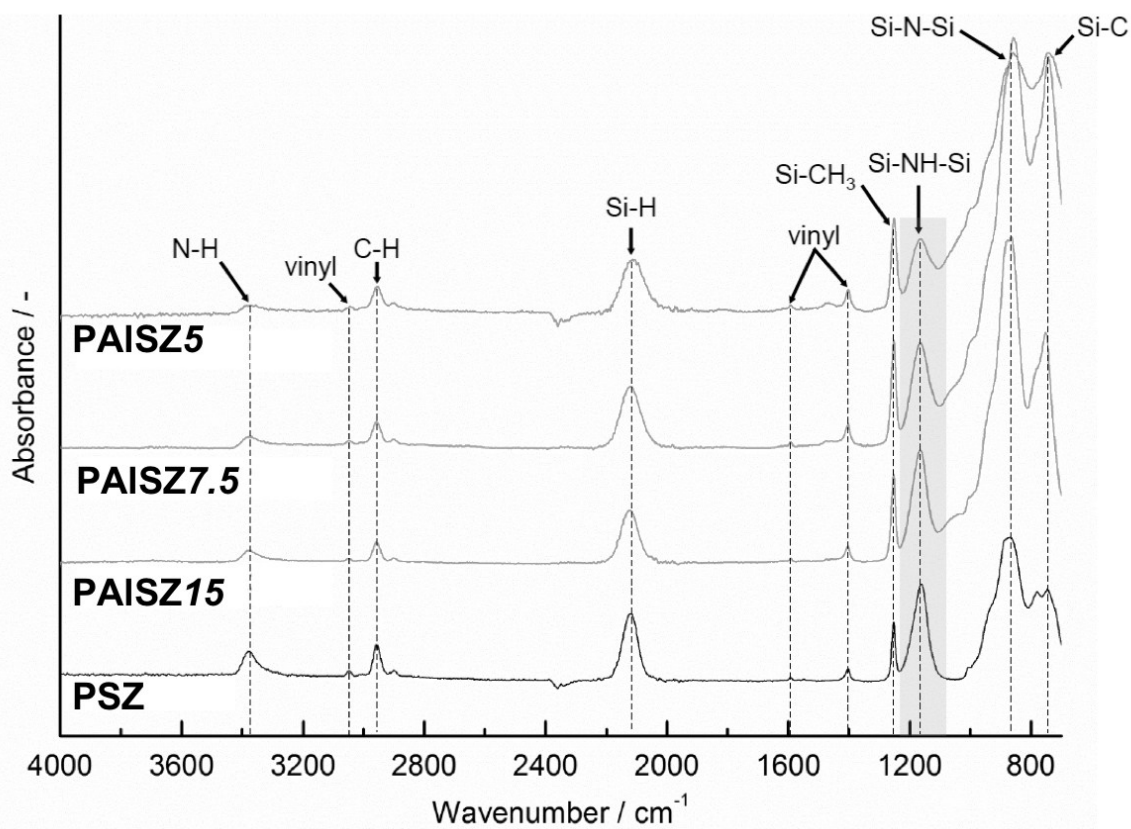
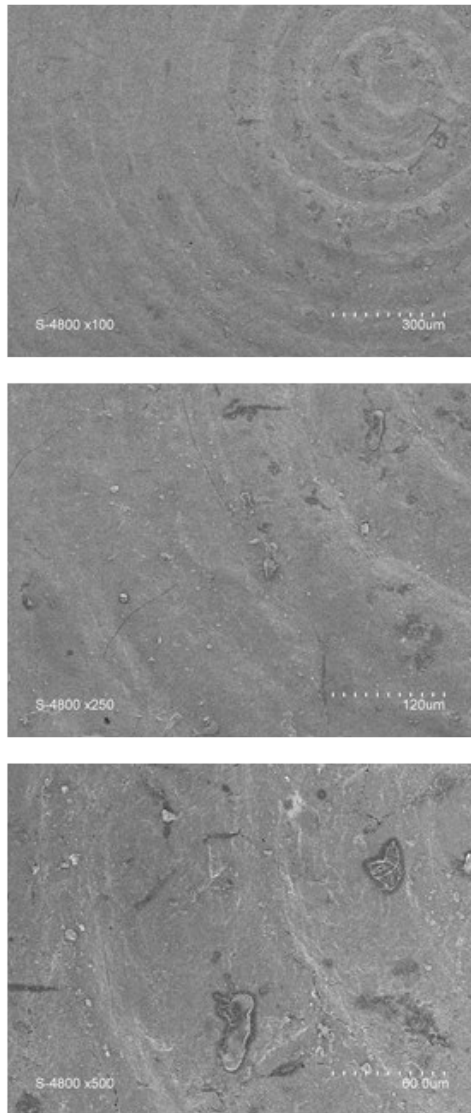


Fig. 1SI. FTIR spectra of PSZ and PAISZX ( $X = 15 \rightarrow 5$ ) samples.

**Table 1S1.** Calculated  $^{27}\text{Al}$  NMR parameters for two crystalline structures and for the models presented in Figure 5.

	$\delta_{\text{iso}}$ (ppm)	$C_Q$ (MHz)	$\eta$
<b>Crystalline structures</b>			
<b>Tris(aluminium hydride dimethylamine)-ICSD 290274</b>			
Al1 (AlN <sub>2</sub> H <sub>2</sub> )	157.8	13.1	0.4
Al2 (AlN <sub>2</sub> H <sub>2</sub> )	160.0	12.3	0.5
Al3 (AlN <sub>2</sub> H <sub>2</sub> )	159.4	12.0	0.5
<b>Tris(dimethylaluminium) hexakis(methylhydrazido)aluminate-ICSD 173483</b>			
Al1 (AlN <sub>6</sub> )	27.5	1.0	0.7
Al2 (AlN <sub>2</sub> C <sub>2</sub> )	180.1	15.9	0.8
Al3 (AlN <sub>2</sub> C <sub>2</sub> )	175.9	13.7	1.0
<b>Models</b>			
<b>Model I (coord 3)</b>			
Al1 (AlN <sub>3</sub> )	164.0	30.2	0.2
Al2 (AlN <sub>2</sub> H)	186.3	32.3	0.1
Al3 (AlNH <sub>2</sub> )	227.4	37.1	0.1
<b>Model II (coord 4)</b>			
Al1 (AlN <sub>3</sub> H)	133.6	14.6	0.0
Al2 (AlN <sub>3</sub> C)	131.9	13.9	1.0
<b>Model III (coord 4)</b>			
Al1 (AlN <sub>4</sub> )	119.7	16.8	0.1
<b>Model IV (coord 5)</b>			
Al1 (AlN <sub>4</sub> C)	85.8	4.3	0.8
Al2 (AlN <sub>5</sub> )	67.8	4.4	0.6
<b>Model V (coord 6)</b>			
Al1 (AlN <sub>4</sub> C <sub>2</sub> )	67.5	9.9	0.3
<b>Model VI (coord 6)</b>			
Al1 (AlN <sub>5</sub> C)	53.8	12.9	0.3



**Fig. 2SI.** SEM images of ceramics prepared at 1000 °C and derived from the **PAISZ3.75** sample warm-pressed at 80 °C