Zeolites have unique pore structures of molecular dimensions and tunable compositions, making them ideal for shape selective catalysis and separation. However, targeted synthesis of zeolites with new pore structures and compositions remains a key challenge. Here, we propose an approach based on a unique 3D-3D topotactic transformation, which takes advantage of weak bonding in zeolites. This is inspired by the structure transformation of PST-5, a new aluminophosphate molecular sieve, to PST-6 by calcination. The structure of nano-sized PST-5 crystals is determined by 3D electron diffraction. We find that the 3D-3D topotactic transformation involves two types of building units where penta- or hexa-coordinated Al is present. We apply this approach to several other zeolite systems and predict a series of new zeolite structures that would be synthetically feasible. This method provides a concept for the synthesis of targeted zeolites, especially those which may not be feasible by conventional methods.
Zeolites and related microporous materials have been widely used as selective catalysts, absorbents, and ion-exchangers\textsuperscript{1–4}. Recently, zeolites have gained interest as catalysts for biomass conversion\textsuperscript{5–8} and automotive exhaust remediation\textsuperscript{9,10}. The unique properties of zeolites are associated with the well-defined pores and channels of molecular dimensions. Because of the close structure-property relationship, numerous efforts have been made to synthesize zeolitic materials with new pore structures and compositions and subsequently explore their properties and applications. Currently, only 252 zeolite framework topologies have been approved by the International Zeolite Association\textsuperscript{11}, even though millions of possible zeolite topologies have been identified by computational methods\textsuperscript{12,13}. The huge gap between experimental output and theoretical prediction indicates that our understanding of zeolite crystalization is still limited. It is, therefore, important to push the field by developing rational strategies to synthesize new zeolites. Besides the conventional hydro-/solvothermal synthesis, several new synthetic strategies have been proposed for targeted zeolites. These include bottom-up approaches that use, for example, pre-designed organic structure-directing agents (OSDAs)\textsuperscript{14–17} and promotion of specific cavitites as in the synthesis of embedded isoreticular zeolites\textsuperscript{18,19} and top-down approaches, i.e., toptotic transformations. The most common top-down approach to synthesize new zeolites has been two-dimensional–three-dimensional (2D–3D) layer condensation from a layered precursor\textsuperscript{20–23}. Recently, a new approach of using known zeolites as precursors to synthesize new zeolites via 3D–2D–3D toptotic transformation has gained significant attention\textsuperscript{24–27}. The most successful example of these methods is the assembly–disassembly–organization–reassembley (ADOR) synthesis route. The unique concept of the ADOR process is to exploit chemically selective weak T–O bonds (T = Al, Si, Ge, etc.), e.g., Ge–O bonds in germanosilicates, in known zeolites and subsequently promote the formation of new T–O bonds. The ADOR strategy provides a synthetic tool for the generation of zeolites with novel topologies and compositions that cannot be synthesized by conventional hydrothermal methods. To the best of our knowledge, these targeted synthesis strategies have only been demonstrated on silica-based zeolite syntheses (e.g., silicates, aluminosilicates and germanosilicates), but never on phosphate-based frameworks.

Aluminophosphates (known as AlPO\textsubscript{4–n}), first synthesized by Union Carbide\textsuperscript{28}, are an important class of zeolitic molecular sieves with wide applications. AlPO\textsubscript{4} molecular sieves have a unique feature, which allows substitution of the Al or P by main block elements (e.g., Si) or transition metals to engender specific properties in the material\textsuperscript{29,30}. For example, silicoaluminophosphate SAPO-34 (framework type CHA) is one of the most widely used solid catalysts for biomass conversion, with a global market of tens of billions of US dollars per year. Currently, only 20\% of the zeolite framework types can be synthesized as AlPO\textsubscript{4} phases\textsuperscript{11}, and examples of rational synthesis of AlPO\textsubscript{4} materials are very rare\textsuperscript{31}. In AlPO\textsubscript{4} frameworks, the AlO\textsubscript{4} and PO\textsubscript{4} tetrahedra always alternate according to Löwenstein's rule\textsuperscript{32}. Nevertheless, Al can adopt pentacoordination with a pair of Al atoms bridged by a hydroxyl group as found in AlPO\textsubscript{4–21} (AWO)\textsuperscript{33} and hexa-coordination with two additional water molecules, for example, in AlPO\textsubscript{4–C} (APC)\textsuperscript{34}. Upon the loss of water molecules by heating, AlPO\textsubscript{4–21} and AlPO\textsubscript{4–C} transform into new fully tetrahedrally coordinated zeolite frameworks AlPO\textsubscript{4–25} (ATV) and AlPO\textsubscript{4–D} (APD), respectively. In 2014, we reported the synthesis and structure of a new AlPO\textsubscript{4} zeolite denoted PST-6\textsuperscript{35}. The framework of PST-6 (PSI) has a one-dimensional (1D) pore system containing parallel 8- and 10-ring channels. PST-6 was obtained by calcination of PST-5 at 500 °C, which was synthesized using diethylamine (DEA) as an OSDA.

Both PST-5 and PST-6 are obtained as nano-size plate-like crystals (Supplementary Fig. 1). The structure of PST-6 was successfully solved by combining electron diffraction and powder X-ray diffraction (PXRD). However, the poorer stability of PST-5 under the electron beam prevented its structure determination, and therefore the understanding of its structure transformation to PST-6. Solid-state nuclear magnetic resonance (NMR) spectroscopy indicated the presence of bridging hydroxyl groups in PST-5.

Recent developments of fast 3D electron diffraction (3DED) or microcrystal electron diffraction (MicroED) data collection techniques (<5 min) using continuous rotation (denoted cRED here) have made it possible for studying beam sensitive nano- and micrometer-sized crystals\textsuperscript{36–49}. Here, we demonstrate the ab initio structure determination of PST-5 using cRED data collected under low-dose condition. By detailed investigations of the structure transformation of PST-5 to PST-6, we propose an approach to generate novel zeolite structures from known zeolites via 3D–3D toptotic transformation. The transformation is initiated via the weak Al–O bonding in penta-coordinated AlO\textsubscript{4}(OH) pairs or hexa-coordinated AlO\textsubscript{4}(OH)\textsubscript{2} units. By adopting this approach, we have, in addition, predicted a series of targeted hypothetical zeolite structures.

**Results**

**Structure of PST-5.** A high-resolution cRED dataset of PST-5 (with a resolution better than 0.85 Å) was collected from a submicrometer-sized crystal using low dose and fast data collection (in total 15 e Å\textsuperscript{–2} and 3 min) on a JEOL JEM2100 transmission electron microscope (Fig. 1a and Supplementary Fig. 2). PST-5 crystallizes in an orthorhombic space group P\textit{2}_{1}b\textit{2}_{1}b (No. 27), with the unit cell parameters \(a = 36.8956(2)\) Å, \(b = 21.80273(9)\) Å, \(c = 10.26929(4)\) Å (Supplementary Tables 1 and 2). Its framework structure was first solved ab initio by direct methods. To confirm the model and obtain more accurate positions and atomic displacement parameters (ADPs) of individual atoms, the model was further refined against the cRED data (Supplementary Table 2). Details of structure determination are given in the Supplementary Information. The framework of PST-5 exhibits a totally new topology (Supplementary Figs. 4 and 5, see Supplementary Information for more details) with a fully four-connected 3D net and a 2D pore system containing parallel 8-ring (5.5 × 2.6 Å) and 10-ring (5.8 × 4.4 Å) channels along the c-axis, and another 8-ring (3.3 × 3.3 Å) channel along the b-axis (Fig. 2). PST-5 is one of the most complex zeolite structures, with 18 Al, 18 P, and 76 O atoms in the asymmetric unit. Four of the O atoms belong to hydroxyl groups and act as bridging atoms to form Al–OH–Al linkages. All 18 P atoms and 10 out of the 18 Al atoms are tetrahedrally coordinated with O atom to form TO\textsubscript{4} units (T = Al, P). The remaining eight Al atoms have trigonal bipyramidal-coordination and form four symmetry-independent AlO\textsubscript{4}–OH–AlO\textsubscript{4} pairs in the structure. Each AlO\textsubscript{4}–OH–AlO\textsubscript{4} pair becomes part of a 3- and a 5-ring (Supplementary Fig. 6). Solid-state NMR spectroscopy confirmed the presence of Al–O–Al linkages (Supplementary Figs. 7–9), and a sharp Infrared (IR) band appearing at 3398 cm\textsuperscript{–1} indicates the presence of bridging OH groups (Supplementary Fig. 10). The structure model obtained by Rietveld refinement against synchrotron PXRD data agrees well with that obtained from cRED data, which further confirms the framework structure of PST-5. In addition, the OSDA positions in the channels could be located from both cRED and PXRD data, and subsequently refined by Rietveld refinement. The final refined unit cell composition is [(C\textsubscript{2}H\textsubscript{5})\textsubscript{2}NH\textsubscript{2}]\textsubscript{16} [Al\textsubscript{2}Z\textsubscript{2}P\textsubscript{2}O\textsubscript{28}(OH\textsuperscript{–})\textsubscript{16}] (Fig. 1b, Supplementary Figs. 11 and 12 and Supplementary Table 3, 20200113762 | https://doi.org/10.1038/s41467-020-17586-7 | www.nature.com/naturecommunications
see Supplementary Information for more details). Raman spectrum shows that DEA molecules are protonated during PST-5 crystallization. Therefore, the negative framework charges created by the bridging Al–OH–Al groups are balanced by the protonated form of the occluded OSDA molecules (DEA∙H⁺), as reflected by quite similar numbers of OSDA molecules and bridging O atoms.

3D–3D topotactic transformation of PST-5 to PST-6. In situ PXRD patterns indicate that the structural transformation of PST-5 to PST-6 starts at 200 °C, and finishes at 500 °C (Supplementary Fig. 13), which is accompanied by notable changes in the PXRD pattern. The observed pattern after calcination of PST-5 at 500 °C agrees very well with the simulated pattern from the PST-6 model, confirming the calcined PST-5 (i.e., PST-6) has the PSI framework (Supplementary Fig. 14). In situ IR studies show that dehydroxylation of bridging OH groups in PST-5 starts above 200 °C and finishes at 300 °C (Supplementary Fig. 10, see SI for more details). Both structures are built from a similar building layer containing 4-, 6-, 8-, and 10-rings (if the OH groups, shown in red are not considered). The difference is the orientations of the (Al,P)O₄ tetrahedra in the two structures. The red atoms in (a) correspond to the OH groups bridging two Al atoms in PST-5. These Al atoms are five-coordinated. Structural models of PST-5 (a) and PST-6 (b) viewed along the c-axis, showing the transforms of double-crankshaft chains (dcc) and double-4-rings (d4r) in PST-5 (a) to narsarsukite-type chains (nsc) in PST-6 (indicated by purple boxes). c Building units in PST-5 (dcc and d4r) and PST-6 (nsc).
Oxygen atoms associated with OH or bond-breaking in the chains are types of building units; double-crankshaft chain (dcc) in PST-5 to PST-6 can be described by the dcc transformation to nsc in PST-6. Green, penta-coordinated (and penta-coordinated derived) AlO4 tetrahedra; yellow, tetra-coordinated AlO4; blue, PO4. Oxygen atoms associated with OH or bond-breaking in the chains are shown in white and the rest O in red.

New zeolite generation. Inspired by the 3D–3D topotactic transformation in PST-5/PST-6, we investigated the existing structures in the Zeolite Structure Database. 12 framework types are found to contain infinite dcc, and they can be divided into three groups based on the structural complexity (Table 1): I) built solely from dcc that are connected directly (APC, GIS, MER, PHI, SIL, GME, Supplementary Fig. 18); II) built from dcc that are bridged by isolated 4-rings (ATT, AWO, UEL, Supplementary Figs. 19 and 20); III) built by dcc and other building units (PST-5, DON, STO, Supplementary Fig. 21). Interestingly, regardless of the complexity, the dcc-nsc structural transformation has been observed in all groups, from AlPO4-C (APC) to AlPO4-D (APD) in group I, AlPO4-21 (AWO) to AlPO4-25 (ATV) in group II, and PST-5 to PST-6 (PSI) in group III (Table 1 and Supplementary Table 4). The three parent frameworks all contain stacking faults, due to changes in both orientation and tetrahedral conformation. There are two symmetry-independent dcc chains in PST-5 with different orientations. After being transformed to PST-6, they orient in the same way. The reconstructed PST-6 (PSI) material was found to contain stacking faults, due to changes in both orientation and tetrahedral conformation. There are eight unique penta-coordinated Al atoms that can initiate the topotactic transformation. When the transformation starts at different positions, it will also generate disorder after the reconstruction.
Table 1 Observed (in bold) and predicted 3D-3D topotactic transformations of zeolites through chain transformation from dcc to nsc and d4r to nsc.

| Group | FCC (parent) | E\textsuperscript{b} | Channel dimensionality | FTC (transformed) | E\textsuperscript{b} | Channel dimensionality |
|-------|--------------|------------------|------------------------|-------------------|------------------|------------------------|
| Group I: dcc → nsc | | | | | | |
| APC  | 14.4         | 2D               | | APD               | 12.3           | 2D                   |
| GIS   | 15.0         | 3D               | | GIS\textsubscript{H1} | 10.3           | 1D                   |
| SIV   | 15.3         | 3D               | | SIV\textsubscript{H1} | 10.3           | 1D                   |
| MER   | 16.2         | 3D               | | MER\textsubscript{H1} | 12.3           | 2D                   |
| PHI   | 15.7         | 3D               | | PHI\textsubscript{H1} | 12.3           | 2D                   |
| GME   | 16.5         | 3D               | | AFI               | 11.0           | 1D                   |
| Group II: dcc + isolated 4-rings → nsc + scc\textsuperscript{c} | | | | | | |
| ATT   | 15.1         | 2D               | | ATT\textsubscript{H1} | 10.8           | 2D                   |
| AWO   | 14.0         | 2D               | | AWO\textsubscript{H1} | 12.2           | 1D                   |
| UEI   | 14.2         | 2D               | | UEI\textsubscript{H1} | 11.2           | 1D                   |
| ATT   | 15.1         | 2D               | | ATT\textsubscript{H1} | 10.8           | 2D                   |
| AWO   | 14.0         | 2D               | | AWO\textsubscript{H2} | 14.7           | 1D                   |
| UEI   | 14.2         | 2D               | | UEI\textsubscript{H2} | 11.3           | 1D                   |
| UEI   | 14.2         | 2D               | | UEI\textsubscript{H3} | 11.0           | 1D                   |
| UEI   | 14.2         | 2D               | | UEI\textsubscript{H4} | 11.3           | 1D                   |
| UEI   | 14.2         | 2D               | | UEI\textsubscript{H5} | 11.5           | 1D                   |
| Group III: dcc + other building units → nsc + other building units | | | | | | |
| PST-5 | 15.9         | 2D               | | PSI               | 9.2            | 1D                   |
| DON   | 10.2         | 1D               | | DON\textsubscript{H1} | 13.2           | 1D                   |
| STO   | 9.7          | 1D               | | STO\textsubscript{H1} | 10.6           | 1D                   |

\textsuperscript{a}Framework Type Code approved by International Zeolite Association.

\textsuperscript{b}Framework energy (kJ (mol Si))\textsuperscript{–1} relative to α-quartz.

\textsuperscript{c}Single crankshaft chain.

Table 6\textsuperscript{43}. Most of them were calculated to have much lower framework energies than the parent dcc-containing zeolites. It is worth mentioning that because each TO\textsubscript{4} tetrahedral unit not belonging to dcc can have two possible connectivities, either upwards or downwards, the number of hypothetical zeolites generated via this approach can be enormous for zeolites in group II and III (see Supplementary Information for more details).

We have demonstrated that many hypothetical zeolites can be predicted from known zeolite frameworks based on the dcc-nsc transformation. The 3D–3D topotactic transformation would provide a simple approach to generate novel zeolite frameworks, which may be inaccessible by conventional hydrothermal/ solvothermal synthesis. We have also shown by molecular dynamics simulations that the weakened bonding, such as penta- or hexa-coordinated Al atoms, is crucial to initiate the 3D–3D topotactic transformation. Therefore, to synthesize new structures by this approach, it is important to introduce penta- or hexa-coordinated Al atoms in dcc-containing zeolite frameworks, and additional exploratory work is needed. Previous studies showed that certain types of OSDAs, like easily protonatable amines, could promote the formation of bridging Al–OH–Al bonds in AlPO\textsubscript{4} molecular sieves, and penta- or hexa-coordinated Al atoms have been found in c.a. 25% of these materials, including AlPO\textsubscript{4}-17, AlPO\textsubscript{4}-41, AlPO\textsubscript{4}-EN3, AlPO-CJBI, VPI-5, IST-1, PST-13, etc.\textsuperscript{41,44–50} The 3D–3D topotactic transformation may also be applicable to other zeolite compositions. For example, the gallosphosphate Mu-18 (UEI) contains both dcc and penta-coordinated atoms (Fig. 4\textsuperscript{a},\textsuperscript{51}). We anticipate that with the discovery of many new zeolites, the approach described here will be more feasible.

Discussion

We have solved the structure of PST-5, a new 2D medium-pore AlPO\textsubscript{4} molecular sieve, using continuous rotation electron diffraction (cRED). Based on the investigations of the dcc to nsc transformations observed in PST-5/PST-6, AlPO\textsubscript{4}-C/AlPO\textsubscript{4}-D, and AlPO\textsubscript{4}-21/AlPO\textsubscript{4}-25 pairs, we propose an approach to generate novel zeolite structures, which enriches the structural diversity. The 3D–3D topotactic transformation relies on the chemical weakness in the dcc. PST-5 transforms dcc and d4r to nsc, changes its overall interlayer connectivities and finally generates PST-6. The penta-coordinated Al atoms, as observed in PST-5, could be built into a precursor material by using pre-designed OSDAs, which balance the framework energy. This approach via the 3D–3D topotactic transformation can be considered complementary to the existing strategies of synthesizing novel zeolite structures. By understanding the 3D–3D topotactic transformation, we anticipate that our approach can be generalized and provide new insights into the discovery of novel zeolitic materials, which may enable a completely new range of zeolite frameworks to be accessible and offer opportunities in technological applications.

Methods

Synthesis of PST-5. The reagents used for the synthesis of PST-5 included diethylamine (DEA, 99.5%+, Aldrich), aluminum isopropoxide (98%+, Aldrich), phosphoric acid (85%, Merck), and deionized water. PST-5 was synthesized from an AlPO\textsubscript{4} gel with the composition of 2.0DEA·1.0Al\textsubscript{2}O\textsubscript{3}·1.0P\textsubscript{2}O\textsubscript{5}·40H\textsubscript{2}O, where DEA is diethylamine. In a typical synthesis of PST-5, 3.84 g of DEA was added to a slurry of 6.95 g of aluminum isopropoxide (>98%, Aldrich) in 5.64 g of H\textsubscript{2}O. After stirring for 1 h, 2.45 g of DEA (99.5%, Aldrich) was added to the above synthesis mixture, which was stirred overnight at room temperature. Afterwards, the mixture was charged into a Teflon-lined 23-mL autoclave and heated under static conditions at 200 °C for 5 days. The resulting white powder was recovered by filtration, washed repeatedly with water and then dried overnight at room temperature.

Topotactic transformation of PST-5. The topotactic transformation of PST-5 to PST-6 was conducted by calcination of PST-5 at 550 °C for 8 h under air flow.

cRED data collection. PST-5 powder in its as-made form was dispersed in ethanol and a droplet of the suspension was transferred onto a carbon-coated copper grid and dried in air. cRED data was acquired in selected-area mode with a goniometer tilt speed of 0.45° s\textsuperscript{–1} and an exposure time of 0.4 s per frame (Supplementary Table 2). The total data collection time was <3 min in order to minimize the beam damage and maximize the data quality. The total tilt range was 92.6°.
Data availability
The crystallographic information files for PST-5 refined against the eRED and PXRD data have been deposited at the Cambridge Crystallographic Data Center (CCDC, free for charge at https://www.ccdc.cam.ac.uk) under deposition number CCDC 1948775 and 1944918, respectively. The cif containing all the hypothetical structures and Supplementary information are available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints.

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