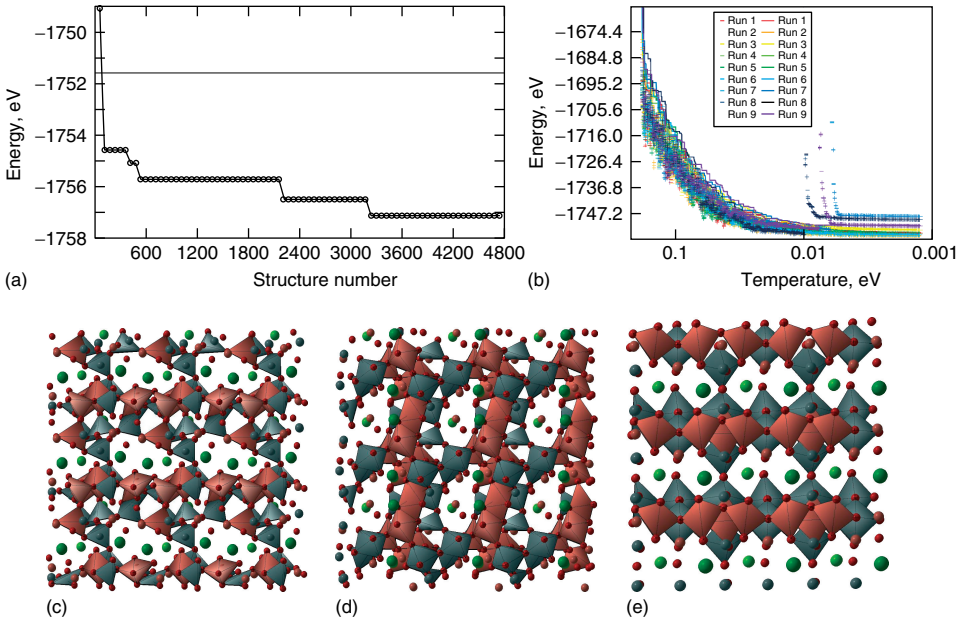

Figure A.2 Test #2 ($Ba_2Mg_2Al_8Si_8O_{32}$, with fixed cell): (a) Variation of the lowest energy during the evolutionary USPEX run, (b) Summary of simulated annealing runs, (c–e) Lowest-energy structures obtained by random sampling, simulated annealing and USPEX, respectively. Shown are Al–O and Si–O polyhedra and Mg, Ba and O atoms are denoted as spheres of different sizes. Thin horizontal line in (a) shows the lowest energy found in 14102 random sampling attempts.

unlikely to produce a similar ''best'' structure so far found by the other methods. Random search can also be viewed as the first (and energetically the poorest) generation of an evolutionary search, or an evolutionary search without selection and without any learning mechanism. Figures A.1a, A.2a, A.3a and A.4a show that in all cases, even short evolutionary runs outperform long and expensive random sampling runs. For Test #1, after 450 structure relaxations USPEX produces better structures than 14794 random sampling attempts. For Test #2, just 120 structure relaxations in USPEX produce better structures than 14102 relaxations in random sampling. For Test #3, within 60 structure relaxations we find better structures than after 13029 relaxations in random sampling. For Test #4, USPEX again wins – in 690 structure relaxations it found better structures than 978 random structure relaxations).

Structures produced by all methods were highly disordered (*cf* their low degrees of order and high quasientropies in Table A.1), which reflects the highly complex chemical compositions that probably do not correspond to chemical compounds that are stable against decomposition. In many ways, these optimization problems are reminiscent of trying to find the crystalline ground state of a system that exhibits a strong tendency to form amorphous structures (c.f. e.g. amorphous silicon boron