Poster Presentations

[MS04-P04] Verification and evaluation of site occupancies using Boolean satisfiability techniques. Reinhard X. Fischer[a], Werner H. Baur[b], Mathias Soeken[c], Rolf Drechsler[c]

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The maximum number of atoms on general and special positions is given by the multiplicities represented in the Wyckoff labels and determined by the site symmetries. The number of combinations to distribute different atoms on crystallographic positions can be calculated using the recursion algorithm given in [1,2]. Site occupancies of mixed atom positions, split positions, or not fully occupied positions are usually determined in least squares refinements of crystal structures based on X-ray or neutron diffraction data. Especially in those cases where two atom positions are too close to each other to be simultaneously occupied by atoms the plausibility of the refinement results has to be evaluated. That means, it has to be verified that there does exist a distribution of atoms avoiding such close contacts to next neighbors. This is trivial when there are just two positions being too close to each other. In that case it has to be obeyed that the total occupancy of the two positions must not exceed 100%. If a position has more than one distance to next neighbors being too short for simultaneous occupancies the situation becomes more complex.
If more than one set of atom positions is present in the unit cell with conflicting distances to nearest neighbors it is impossible to verify possible atom distributions by simple methods.
Here we present a method for the evaluation of site occupancy factors using Boolean satisfiability (SAT) techniques which are, e.g., successfully employed in the field of circuit and system design
[3]. A Boolean variable is assigned to each atom position in the unit cell being true if it is occupied and false if it is vacant. Restrictions due to site occupancies are encoded as Boolean expressions which are combined and passed to the SAT algorithm. The optimization yields the highest possible occupancy of one site and/or the validation of the occupancy factors from crystal structure refinements. That way it could be shown that the occupancy factors of many published structures with a statistical distribution of atoms are not correct. That means, it is impossible in these cases to find a possible distribution of atoms avoiding any distance too close for simultaneous occupancy.
Thus, this approach could be used to routinely evaluate the results of occupancy refinements for plausible distributions of atoms. The approach is demonstrated as applied to more
than 60 zeolite structures of the HEU (heulandite) type having partial and mixed occupancies in nonframework sites with a high number of wrong determinations.

[1] Müller, U. (1988). Z. Kristallogr. 182, 189.
[2] Müller, U. (2013). Symmetry relationships between crystal structures. Oxford University Press, pp 352.
[3] Drechsler, R., Eggersglüß, S., Fey, G., Glowatz, A., Hapke, F., Schlöffel, J. & Tille, D. (2008). IEEE Trans. on CAD (TCAD) 27, 1329-1333.

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