An Unicursal method to give hydrogen atom placement of Hydrogen Hydrate Clathrate

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We show a method to give an initial point of Hydrogen Atom (HA) needed by a molecule dynamics calculation of water molecules [1, 2]. According to the X-ray scattering experiment of D_2O, around one oxygen atom, there are four points of HA which occupy half hydrogen from the view of quantum mechanics. There are two points of HA between nearest point of Oxygen Atoms (OA). A point of OA is surrounded by four neighbour point of HA. However those points of atoms can not be applied directly to the calculations of molecule dynamics simulations, because actual water molecule consists of one oxygen atom and two hydrogen atoms.

One of two points of HA between oxygen atoms must be removed. In the remaining point of HA there is a hydrogen atom of water molecule. The choice must be performed with consistency, because each oxygen atom is surrounded by two hydrogen atoms. However, it is difficult to expect appropriate choice. The simulation that used a first point of HA without consistency does not give a good result.

To stand off this trouble, typically points of HA are put on the center of hydrogen bond. However, this method makes the numerical simulation complicate. It takes long time to calculate molecule dynamics simulations and the structure of Clathrate collapses easy. Our new method provides consistency, and leads initial points of HA without trouble.

A method introduced in this report keeps certainly consistency, each oxygen atom have two hydrogen atoms. Our method consists of two steps. At the first step, we find the points of four OA, which bonds by hydrogen bond for each oxygen atoms. For example, we consider a structure sII which has Fd3m symmetry. It is made from two kinds of cages. The small cage consists of 20 water molecules, has 12 pentagons and 30 sides, and 30 hydrogen atoms exist on the line forming hydrogen bond. Similarly the large cage consists of 28 hydrogen molecules, 12 pentagons and 4 hexagons, and 42 lines, and 42 hydrogen atoms exist on the lines forming hydrogen bound. The sII is one of structure which built up Crathrate hydrate, and then consists of two kinds of cages, the Small cage and the Large cage. The points of all oxygen in Clathrate hydrate are determined by Fd3m Symmetry and by X-ray scattering experiment of D_2O. At the next step, we draw arrows from point of OA to near point of OA. The starting (end-) point of arrow corresponds to coordinate (hydrogen) bond. We take arbitrary point of OA, and then draw an arrow from it to one of four neighbour points of OA. From end-point of the arrow, a next arrow is draw to one of three near point of OA. We repeat his step of procedure until that the end-point of arrow arrives in first point of OA twice. If the point of OA doesn't have two end-points of arrow, one of them is choice as "next arbitrary point of OA". We repeat this procedure until all points of OA have two end-points of arrow.

We call this method "unicursal method". Based on Euler path in topology, the number of hydrogen bond corresponds to order for a point of oxygen atom. This order is an even number. Therefore, the appropriate choice leads a unicursal path.

Reference: [1] Masahiro Nakano, Hajime Endou, Hiroyuki Matsuura, Ken-ichi Makino, Tadahiro Kin, Kazuharu Koide and Nobuo Noda: Clathrate hydrates as a phase of H_2O molecules, Biomedical Soft Computing and Human Sciences, Vol.11 No.1, pp.51—55, 2006.
[2] Masahiro Nakano, Hiroyuki Matsuura, Hajime Endou, Tadahiro Kin, Ken-ichi Makino, Kazuharu Koide and Nobuo Noda: Molecular Dynamics Simulations of Hydrogen Storing in Clathrate Hydrates, INFORMATION, Vol.10. No.2. pp.243-251, 2007