Biodegradation Mechanism of Polycaprolactone by a Novel Esterase MGS0156: A QM/MM approach

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Method details

MM-PBSA calculation.

In the MM-PBSA method, the free energy of binding was deduced from the following equation:

$$\Delta G_{bind} = G_{PL} - G_P - G_L$$

where the free energy of a state (P denotes the protein and L denotes the ligand) was speculated from processing the receptor-ligand conformations which were generated from 25 ns MD simulations on the basis of Amber18 software [1]. The binding of eight polyesters (e.g. PCL2, PCL3, PCL4, PCL5, PCL6, PCL8, PCL10, PCL12) and PCL monomer with hydrolase MGS0156 were investigated.

In order to get insights of the binding of enzyme and ligand, MM-PBSA calculations were performed using the MMPBSA.py.MPI scripts in the parallel version of AMBER18 [2]. For each system, totally 2500 frames obtained from the MD simulations were used to calculate the average binding free energy. Single trajectory protocol (STP) was selected to run the MD simulation for MM-PBSA rather than the multiple trajectory protocol mainly due to the following reasons: (i) ligand is not large enough to change the structure of enzyme; (ii) the interaction between the ligand and protein is mainly from the residues of the enzyme active site environment. The other settings were set as default. Energy decomposition was also calculated [3].

1. The binding of nine independent oligomers to hydrolase MGS0156

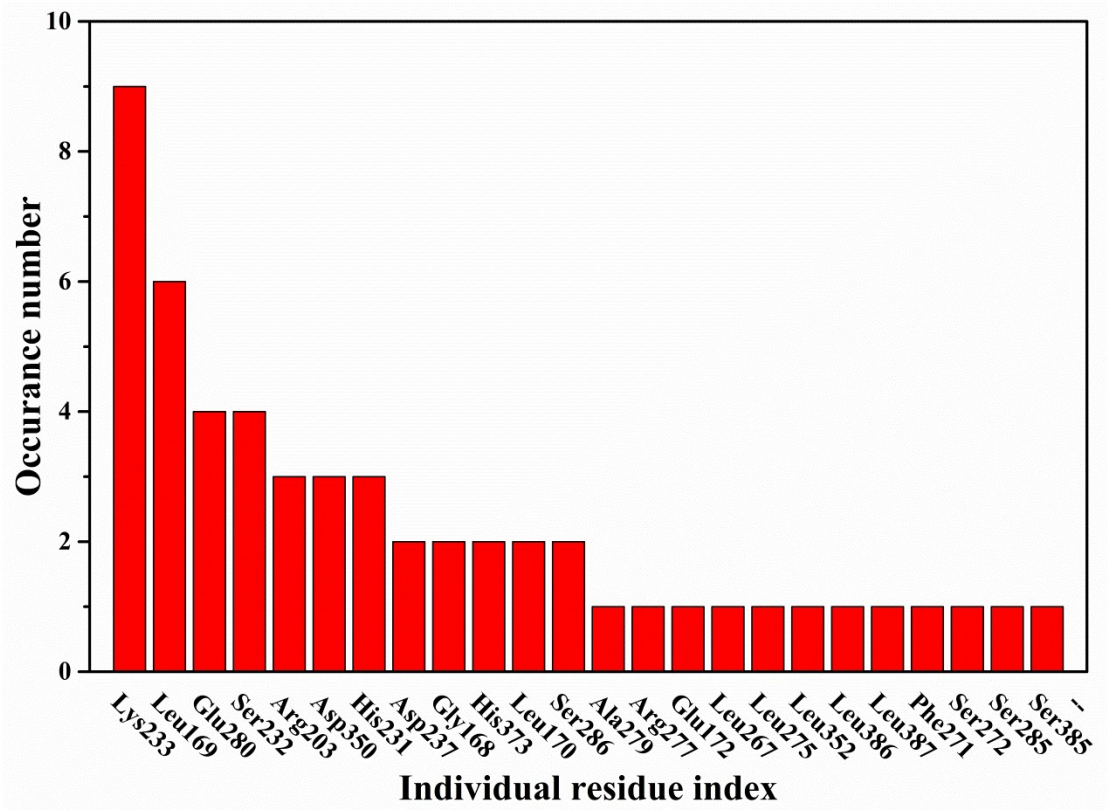

Figure S1. Occurrence number of amino acids that contribute most to the substrate binding.

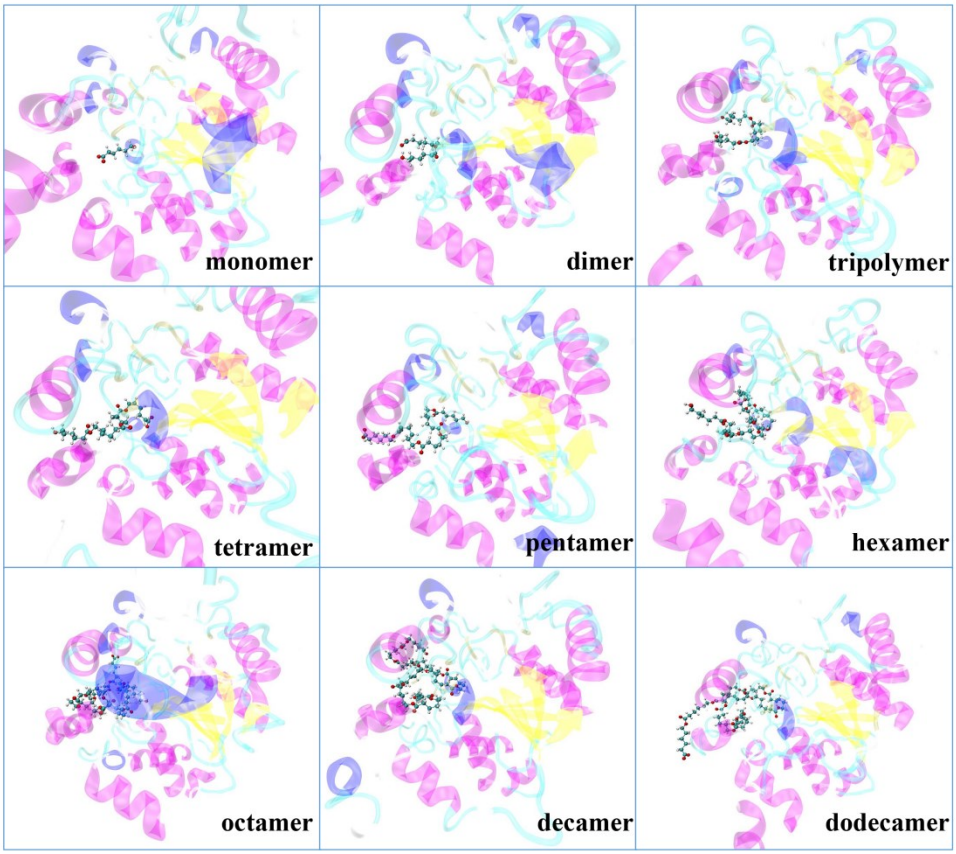

Figure S2. The three-dimensional structures of nine PCL oligomers with MGS0156 esterase after molecular dynamics.

2. the following detailed structural analysis

Table S1. Key structural data involved in the reaction state of 37.8 ns. Relative bond lengths are in Å.

	d(C^1-O^1)	**d(O^1-H^1)**	**d(H^1-N^1)**	**d(H^1-O^3)**	**d(C^1-O^3)**
R	2.71	1.01	1.66	3.04	1.33
TS1	1.78	1.29	1.19	2.72	1.39
IM	1.48	1.63	1.04	2.69	1.45
TS2	1.38	2.40	1.30	1.16	1.87
P	1.33	2.56	1.70	1.00	2.46

3. The ESP analysis

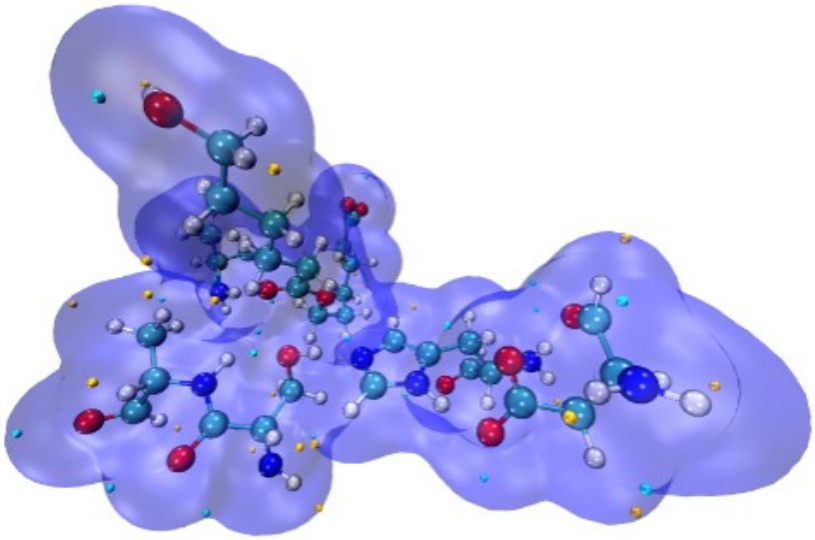

Figure S3. ESP mapped molecular vdW surface of PCL2 and two critical amino acids. The unit is in kcal/mol. Significant surface local minima and maxima of ESP are represented as blue and yellow spheres respectively.

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