Supporting information

Atomic Models and Molecular Dynamics Simulations

The minimized atomic structures of Psl repeat unit, which is a pentasaccharide of D-mannose, D-glucose and L-rhamnose, and its polysaccharide chain containing at least two repeat units [→3)-*α*-L-RhaP-(1→3)-*β*-D-Glcp-(1→3)-[*α*-D-Manp-(1→2)]-*β*-D-Manp-(1→3)-*β*-D-Manp-(1→]$_2$ was generated by the GLCAM biomolecule builder (Woods Group. (2005-2016) GLYCAM Web. Complex Carbohydrate Research Center, University of Georgia, Athens, GA. (http://glycam.org)). The molecular model of cationic antimicrobial peptide LG21, with acetylated at C-termini and amidated at N-termini (Ac-LLPIVGNLLKSLLGWKRKRFG-NH_2) shown in Figure S2 (a) was built by using tLeap module of AmberTools [1]. The simulation system to study the interaction of LG21 with Psl chain was generated by placing one Psl chain shown in Figure 2(b) and one LG21 peptide in a simulation box with charge balancing counterions. The Psl polysaccharide was modeled using the standard carbohydrate specific forcefield GLYCAM06 [2], whereas the peptide and ions were modeled using amber94SB forcefield [3]. As the atom types of GLYCAM06 are AMBER-consistent, we use this combination of two different forcefields to describe the interactions of LG21 with Psl. Water molecules were represented with SPC/E model [4]. All simulations were carried out using GROMACS 4.6.5 simulation package [5]. First, energy of the system was minimized using steepest descent method. Equilibration of the systems was performed in two steps with position restraint on the LG21 and Psl chain. First, according to the Maxwell-Boltzmann distribution at 300 K initial velocities were assigned to the system and a 100 ps equilibrium simulation was performed in a NVT ensemble at 300 K. A subsequent 200 ps equilibration was performed in an NPT ensemble. Finally, a longer production run without any position restraints was carried out for 200 ns to achieve the enough sampling. For all the simulations 2 fs time step was used. The simulation temperature and pressure were controlled by a V-rescale thermostat and Parrinello-Rahman barostat, respectively [6,7]. The particle-mesh Ewald method was applied to calculate the electrostatic interactions, and a Lennard-Jones potential was used to describe the dispersive interactions.[8,9] Atomic coordinates were collected every 20 ps from the total production run. For comparison of peptide secondary structure evolution, a replica exchange molecular dynamics (REMD) was carried out as a control on a

similar simulation system as above but containing only LG21 peptide in the solution. The temperatures in REMD simulations were distributed from 300 to 470 K among 80 replicas, and predicted using the temperature generator tool to achieve the exchange probability of ≈ 0.25 [10].

Figure S1. (a) Initial positions of the Psl chain and LG21 in the simulation box for MD simulations. **(b)** Minimum distance between Psl chain and LG21 peptide during the 200 ns MD simulation.

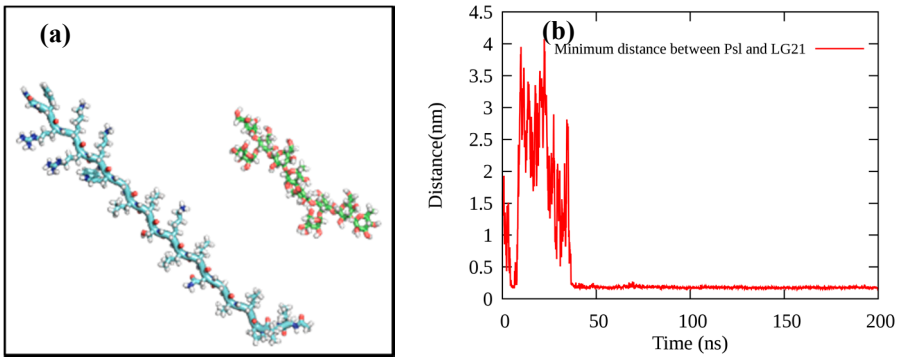
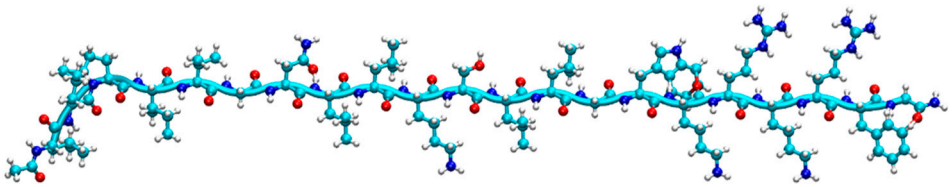


Figure S2. Atomic models of (a) LG21 peptide and (b) Psl polysaccharide chain. Color Code : C, cyan ; N, blue ; O, red; H, white.

(a)



(b)

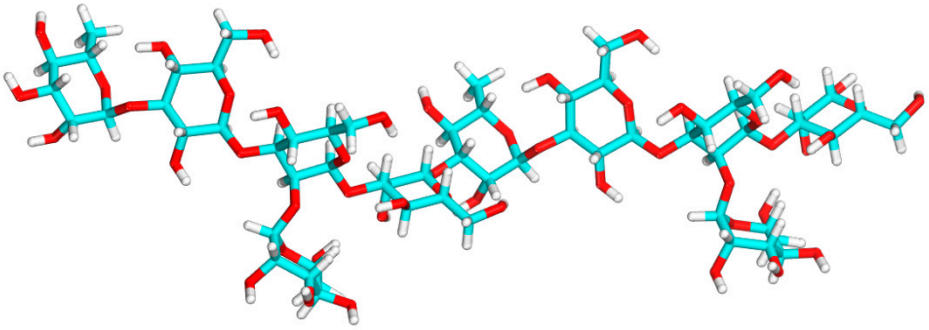


Figure S3. Interaction energy between Psl chain and each residue in LG21.

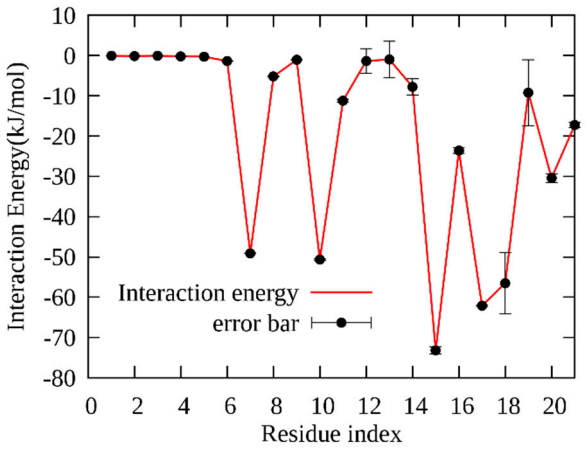
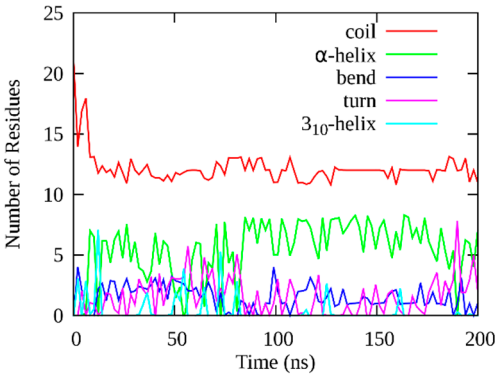
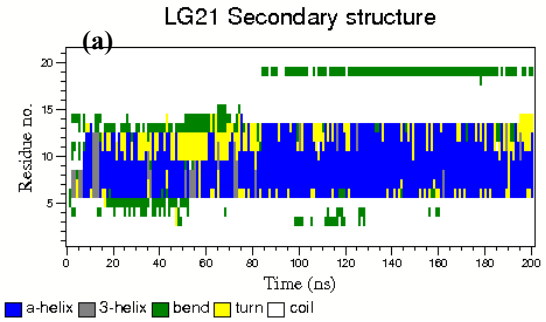


Figure S4. (a) Total number of residues possessing a secondary-structure are plotted along the simulation time. At the starting of the simulation, all the 21 residues adopt a random-coil structure. As the Psl comes into contact with the LG21 peptide, some residues take a helical conformation which can be seen from the decrease in residue number for random-coil structure and increase in the number of residues representing helical conformation (b) Structural evolution of each residue of LG21 peptide while interacting with Psl chain.





(b)

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