Residue-Specific Dopa-Incorporated Engineered Mussel Bioadhesives

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Dopa, which is a hydroxylated form of tyrosine, has been suggested to be the key factor for rapid and strong underwater adhesion. Mussel adhesive protein (MAP) is one example where Dopa chemistry is the core of their underwater adhesion with extremely high Dopa contents of ~10-25 mol%. The biosynthesis of recombinant MAPs in an *E. coli* system was a good approach but lack of Dopa in recombinant MAPs critically limited the underwater adhesion. Here, we explore the *in vivo* residue-specific Dopa incorporation into recombinant MAPs. Misaminoacylation of Dopa to $tRNA^{Tyr}$ by tyrosyl-tRNA synthetase allowed the quantitative replacement of tyrosine residues with a yield of over ~90 %, to create engineered MAPs in *E. coli* with a very high Dopa content. The Dopa-incorporated MAPs exhibited a superior surface adhesion and water resistance ability by assistance of Dopa-mediated interactions, and furthermore, showed underwater adhesive properties comparable to those of natural MAPs. These results propose promising use of Dopa-incorporated engineered MAPs as bioglues or adhesive hydrogels for practical underwater applications.