論 文 の 内 容 の 要 旨

論文題目　Antioxidant and adhesive properties of bio-inspired gallol polymers
（ガロール基をもつ生体模倣高分子の抗酸化性と接着性評価）

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Polyphenols, which are the plant-derived chemicals with two or more phenolic groups, have antioxidant, adsorption, and anti-inflammatory properties. Most polyphenols include gallol groups in their chemical structures, which have inspired me to synthesize gallol polymers. In my research, I synthesized new gallol polymers and studied their antioxidant, adsorption, and adhesive properties.

My PhD thesis consists of four chapters. The first chapter is the general introduction of polyphenols, antioxidants, and adhesives.

The second chapter is my research on antioxidant and adsorption properties of gallol polymers. In this chapter, I carried out the reversible addition-fragmentation chain transfer polymerization to obtain polymers with a wide range of molecular weight and low polydispersity. These newly synthesized gallol polymers (polyvinylgallol, PVGal) exhibited greater antioxidant activities than catechol polymers (polyvinylcatechol, PVCat) as revealed by radical scavenging ability measurements using antioxidant assays. Also, I investigated the adsorption properties of PVGal by quartz crystal microbalance measurements. I found that the adsorbed amount of PVGal was higher than that of other phenolic polymers on silicon dioxide, gold, nickel and aluminum substrates, suggesting that gallol polymers are a candidate for high-performance adhesives.

The third chapter is the research on underwater adhesive properties of gallol copolymers. Inspired from the tunicate adhesive protein which contains gallol groups, I first tried to investigate the adhesive properties of PVGal. However, the PVGal were too fragile to be used as adhesives.

Therefore, I synthesized softer gallol copolymers [poly(vinylgallol-*co*-*n*-butyl acrylate), P(VGal-*co*-BA)] containing soft butyl acrylate moieties by means of the methoxymethyl protecting/deprotecting (MOM) method. I evaluated the bonding strength of P(VGal-*co*-BA) based on the lap shear measurements in air, water, seawater, and phosphate-buffered saline solution. I found that the bonding strength of P(VGal-*co*-BA) was much better than that of catechol copolymers [poly(vinylcatechol-*co*-*n*-butyl acrylate), P(VCat-*co*-BA)] in all tested conditions. Then I analyzed the mechanism of bonding strength. There were two contributions to bonding strength; the interfacial interaction and cohesive interaction. In my case, the stronger bonding strength of P(VGal-*co*-BA) was attributed to the stronger cohesive interaction inside P(VGal-*co*-BA). Additionally, I found that the P(VGal-*co*-BA) could be used as adhesives on many substrates including polytetrafluoroethylene, polyvinylchloride, aluminum, and pig skin. Moreover, the bonding strength of P(VGal-*co*-BA) was greater than a commercial water-proof glue in seawater condition.

The fourth chapter is the conclusion of my works and perspective.

In summary, I synthesized polyphenol-inspired polymers containing gallol groups. The antioxidant activity of the newly developed PVGal was higher than that of PVCat. The adsorption property of PVGal on various substrates was higher than the other phenolic polymers. Moreover, the P(VGal-*co*-BA) which were synthesized by the MOM protecting/deprotecting method showed greater adhesive performance than the other phenolic copolymers under all tested conditions. Thus, the gallol polymers were a promising candidate for high-performance antioxidants and adhesives. The present study will shed light on the possibility of applying gallol polymers for various applications such as antioxidant coating, surface modification, self-healing, and adhesive.