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# Highly efficient mussel-like inspired modification of aramid fibers by UV-accelerated catechol/polyamine deposition followed chemical grafting for high-performance polymer composites

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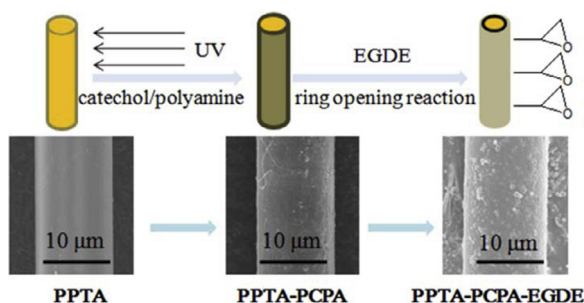
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## HIGHLIGHTS

- Aramid fibers were modified by catechol/polyamine deposition and EGDE grafting.
- The deposition process of catechol/polyamine was accelerated by UV irradiation.
- The modified aramid fibers show an improved interfacial adhesion property with rubber matrix.

## GRAPHICAL ABSTRACT



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## ABSTRACT

A facile and efficient mussel-like inspired modification method was developed to improve the interfacial adhesion of aramid fibers/rubber composites. The surfaces of aramid fibers were pre-deposited by poly (catechol/polyamine) (PCPA) layer, which was formed via the oxidation polymerization of catechol/polyamine (CPA) under UV irradiation. Then, the PCPA-coated fibers were further grafted with ethylene glycol diglycidyl ether (EGDE) to introduce epoxy groups onto aramid fibers surface. The kinetics of catechol oxidation was investigated by UV–vis measurements, suggesting that UV irradiation accelerated the polymerization process of CPA. The effects of EGDE concentration and grafting time on the adhesion force between aramid fibers and rubber matrix were evaluated by pull-out test, and a maximum increase of 85.6% in adhesion force was achieved, even better than that by dopamine in our previous study (a 67.5% improvement). Compared with the method based on dopamine chemistry, this method has advantages of lower cost (less than 1% of the price of dopamine) and shorter reaction time (3 h less in pre-deposition time). This study provides a new efficient strategy for fibers surface functionalization, which has promising application in rubber industry.

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## 1. Introduction

Aramid fiber has attracted tremendous interests owing to its unique properties including low specific density, high strength and modulus, and excellent resistance to heat and chemicals

[1–3]. It has become one of the most ideal reinforcement agents in high-performance composites fields [4–6]. To meet the certain requirements, a superior interfacial bonding strength between fibers and composites matrix is of great practical significance. However, due to its chemical inert and smooth surface, aramid fiber suffers from a poor interfacial interaction with polymer matrix. Over the past decades, enormous chemical and physical methods have been exploited to improve its surface activity, including wet oxidation [7], monomers grafting [8], plasma treatment [9–11], and  $\gamma$ -rays irradiation [12]. However, chemical treatments lead to the decrease in fiber strength to some extent, and physical treatments are expensive and usually require rigorous conditions.

Since 2007, polydopamine (PDA) has attracted considerable attention owing to its outstanding adhesion ability, simple modification process, and versatile potential for secondary functionalization [13]. With these advantages, PDA has been widely applied throughout the chemical, biological, medical, and material fields [14–22]. Recently, some studies have used PDA to enhance the interface interaction between fibers and composites matrix [23–27]. Chen et al. [27] developed an efficient route to functionalize carbon fibers based on dopamine chemistry and the functionalized fibers showed great reinforcing effect with epoxy resin compared with untreated ones. The interfacial adhesion between carbon fibers and polymer matrix was highly improved by 35%. In our previous work [23], the combination of PDA deposition and silane grafting on PPTA fiber surfaces was introduced. The result showed that the interfacial adhesion force of the fiber/rubber composites was improved by 67.5%.

However, the high price of dopamine greatly limits its application in industry fields. Fortunately, it has been confirmed that the catechol group and the crosslinked network formed via oxidative polymerization are the major reasons for the superior adhesion ability of PDA [28]. Wang et al. found that low-cost catechol and polyamine self-polymerized and co-deposited on the surface of polypropylene separator, which highly improved the electrochemical performances of separator. However, the tedious reaction time (24 h) was inefficient for quantity modification [29]. Meanwhile, Du et al. reported that the kinetics of dopamine polymerization was accelerated by UV irradiation due to the introduction of reactive oxygen species, which promoted the dopamine-quinone oxidation [30]. Considering that catechol/polyamine (CPA) possesses the similar chemical structure with dopamine, we hypothesize that the polymerization and deposition process of CPA can also be accelerated by UV irradiation. Moreover, owing to the possession of abundant hydroxyl and amine groups, the formed poly(catechol/polyamine) (PCPA) layer can act as a versatile platform for further functionalization via the reaction with functional monomers. It has been noted that the epoxy groups can react with amine groups on PCPA layer, and can also join the rubber vulcanization [23,25]. Therefore, ethylene glycol diglycidyl ether (EGDE) which containing epoxy groups were further grafted

on PCPA-coated aramid fibers to improve the interfacial interaction of the aramid fiber/rubber composites.

In this work, an effective mussel-inspired approach to improve the interfacial activity of aramid fibers was developed. The aramid fibers were first modified by CPA under UV irradiation, and then the PCPA-coated aramid fibers were further functionalized with EGDE. The scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS) and thermogravimetric analysis (TGA) results demonstrated the formation of uniform coatings on fibers surface through UV-accelerated CPA deposition and EGDE grafting. The tensile strength of PPTA fibers was measured by an electronic tensile tester of single filament. Results showed that the process of polymerization and deposition of CPA on aramid fibers surface was accelerated by UV irradiation without much sacrificing the mechanical properties of aramid fibers. The influences of the EGDE concentration and grafting time on the structure and adhesion property of the modified aramid fibers were systematically investigated. The interfacial adhesion performance of PPTA fibers with rubber matrix was characterized by the pull-out test, and as a consequence, the modified aramid fibers exhibited a highly improved interfacial adhesion with rubber matrix due to the introduction of epoxy groups. With advantages of high efficiency and low-cost, this strategy shows great potential in the field of high performance fibers modification.

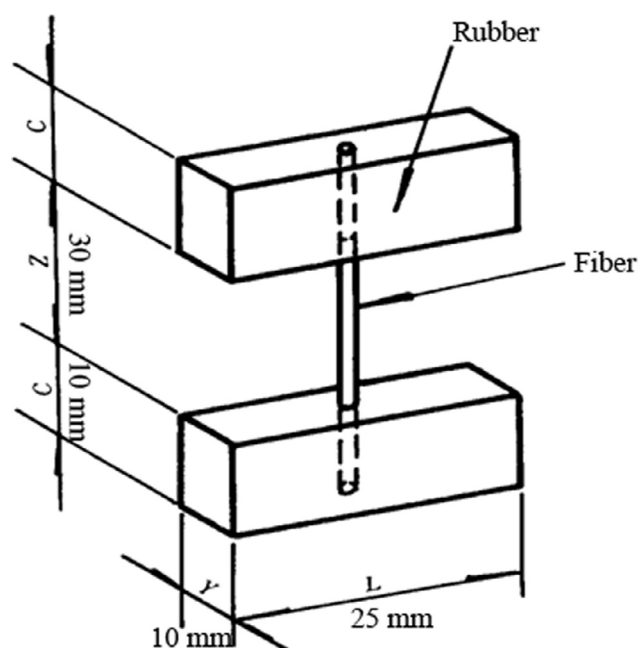


Fig. 2. The schematic structure of the PPTA fibers/rubber specimen.

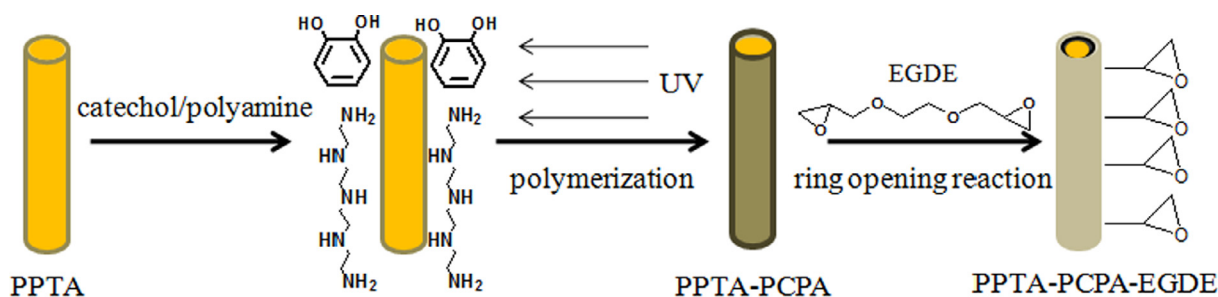


Fig. 1. Schematic description of surface modification procedure of PPTA fibers.

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