



Recent progress in chemical modification of polyaniline



Mehdi Jaymand*

Research Center for Pharmaceutical Nanotechnology, Tabriz University of Medical Sciences, Tabriz 5158654778, Iran

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ABSTRACT

Polyaniline (PANI) has been the subject of considerable recent interest because of their unique electrical behavior, good environmental stability in doped and neutral states, ease of synthesis and wide applications in different fields. However, the main drawback of PANI is lack of solubility, which explains its limited processability due to a rigid backbone. Various procedures have been adapted to improve its processability. The major part of this review is a discussion of some of the methods employed for chemical modification of PANI, including doping via Acid–Base chemistry, sulfonation of PANI or copolymerization of aniline with sulfonated aniline derivatives, synthesis of PANI composites with processable polymers, copolymerization of aniline with substituted corresponding monomers, incorporation of polymeric chains or long and flexible alkyl chains in the PANI backbone, and enzymatic polymerization of aniline during the last decade.

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Abbreviations: PANI, polyaniline; PTh, polythiophene; PPY, polypyrrole; NMP, *N*-methylpyrrolidone; DMSO, dimethylsulfoxide; CSA, camphor-sulphonic acid; DBSA, dodecylbenzenesulfonic acid; *p*-TSA, *p*-toluenesulfonic acid; SPS, sulfonated polystyrene; PAPBA, poly(3-aminophenylboronic acid); EB, emeraldine base; LEB, leucoemeraldine base; PAB, pernigraniline base; PAPBA, poly(3-aminophenylboronic acid); CuClO₄·4BN, copper perchlorate tetrabenzonitrile; NaSIPA, sodio-5-sulfo-isophthalic acid; [P(MMA-*co*-StSA)], poly(methyl methacrylate-*co*-*p*-styrenesulfonic acid); [P(St-*co*-StSA)], poly(styrene-*co*-*p*-styrenesulfonic acid); [P(MMA-*co*-AMPS)], poly(methyl methacrylate-*co*-2-acrylamido-2-methyl-1-propanesulfonic acid); ELBL, electrostatic layer-by-layer; PDAC, poly(dimethyl diallylammonium chloride); VM, vinyl-coumarin; AMPS, 2-acrylamido-2-methyl-1-propanesulfonic acid; SPANI, sulfonated polyaniline; MA, metanilic acid; HOS, human osteosarcoma; LSC, laser scanning cytometry; D, dispersity index M_w/M_n ; CRP, controlled/living radical polymerization; NMP, nitroxide-mediated polymerization; ATRP, atom transfer radical polymerization; RAFT, reversible addition–fragmentation chain transfer polymerization; PMDETA, *N,N,N',N',N''*-pentamethyldiethylenetriamine; DMF, *N,N*-dimethylformamide; APS, ammonium peroxydisulfate; PSDA, polysulfonic diphenyl aniline; PEO, poly(ethylene oxide); Tos, tosylate; PDMS, polydihydroxydimethylsiloxane; BMF, butylated melamine formaldehyde; XPS, X-ray photoelectron spectroscopy; EMI, electromagnetic interference; HIL, hole injection layer; PLED, light-emitting diode; GMA, glycidyl methacrylate; AAc, acrylic acid; SSAC, 4-styrenesulfonic acid; DMAPS, amphoteric *N,N'*-dimethyl(methacryloylethyl)ammonium propanesulfonate; PMAS, poly(2-methoxyaniline-5-sulfonic acid); SSDP, self-stabilized dispersion polymerization; PEG, poly(ethylene glycol); PEA, poly(ethyl acrylate); SEM, scanning electron microscopy; PDGEB, poly(diglycidyl ether bisphenol A); NBS, *N*-bromosuccinimide; PSt-*co*-PMSt, poly(styrene-*co*-*p*-methylstyrene); SBP, soybean peroxidase; HRP, horseradish peroxidase; DNA, deoxyribonucleic acid; PSC, pleurotus sajor-caju; DODD, sodium dodecyl diphenyloxide disulphonate; TPYP, tetrapyrrolylporphyrin; (TPYP)FeIII–OOH, iron (III) hydrogen peroxide porphyrin; SDBS, sodium dodecylbenzenesulfonate; PAMPS, poly(2-acrylamido-3-methyl-1-propanesulfonic acid).

* Tel.: +98 411 3367914; fax: +98 411 3367929.

E-mail addresses: m.jaymand@yahoo.com, m.jaymand@gmail.com

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1. Introduction

In recent decades, there has been growing interest in the development of organic materials for both optical and electronic applications [1–4]. The excellent structural versatility that may be achieved with organic compounds by both high-precision molecular design and an appropriate synthetic method is one of the main reasons for this fast-growing interest. Among the wide range of materials that may be suitable for the aforementioned applications, conjugated polymers such as polyaniline (PANI), polythiophene (PTh), and polypyrrole (PPy), are of particular interest. Because of their potential applications, such as electromagnetic interference (EMI) shielding [5–7], rechargeable battery [8,9], chemical sensor [10–13], photovoltaic cell [14,15], gas separation membranes [16–18], enzyme immobilization matrices [19,20], corrosion devices and microwave absorption [21–23]. In addition, conducting polymers can be used as conducting fillers in insulating polymer matrices in preparation of electrically conducting composites. These composites offer potentials in electromagnetic interference (EMI) shields, electronic packaging, display devices and electrodes [24–29].

Conductive polymers can be prepared by electrochemical, or chemical oxidation of corresponding monomers in various organic solvents and/or in aqueous media [30–41]. The electrochemical process is more advantageous since film properties such as thickness and conductivity can be controlled by the synthesis parameters, including the current density, substrate, pH, nature and concentration of electrolyte.

The main drawback of unsubstituted π -conjugated polymers is its lack of solubility, which explains its limited processability due to the rigid backbone. To improve their processability, various procedures have been adapted. This review highlights the major developments in chemical modifications of polyaniline (PANI) during the last decade.

2. Polyaniline (PANI)

Since the discovery of doped polyacetylene as a conducting polymer in 1977 [42] it has been found that only a few polymers are stable enough under normal processing conditions to be incorporated in practical applications. Among leading candidates are polythiophene, polypyrrole, and polyaniline. In these important conducting polymers, polyaniline has many advantages over other conducting polymers such as polythiophene and polypyrrole. Polyaniline is thermally stable up to 250 °C and can be easily synthesized chemically and electrochemically via

oxidative polymerization in various organic solvents and/or in aqueous media [43]. Moreover, polyaniline has attracted attention due to its low cost, good environmental stability [44,45], adequate level of electrical conductivity [46–49], and wide range of commercial and technological applications, such as secondary batteries [8,50], electromagnetic interference (EMI) shielding [5,6], solar cells [9,51], bio/chemical sensors [52–54], corrosion devices [21,22], organic light emitting diodes (OLEDs) [55], and electrorheological (ER) materials [56–58].

In addition, a novel achievement with PANI and its derivatives is their electrical memory performance [59–70], because they exhibit significant advantages over inorganic silicon- and metal-oxide-based memory materials in the fabrication of volatile and non-volatile memory devices: their dimensions can easily be miniaturized, and their properties can easily be tailored through chemical synthesis [64]. For example, Lee et al. [67] investigated the effect of the electrode material on the electrical-switching characteristics (i.e., electrical-switching behavior, switching voltage, and on/off current ratio) of a non-volatile resistive-memory device based on an active poly(*o*-anthranilic acid) thin film. They found that the switching characteristics of the active polymer layer depended strongly on the bottom-electrode (BE) material.

Three different types of basic PANI, i.e., fully reduced leucoemeraldine (LEB) ($x=0$), half oxidized emeraldine (EB) ($x=0.5$), and fully oxidized pernigraniline (PAB) ($x=1$) are known (Scheme 1).

All conducting polymers (and most of their derivatives), can be transformed to conducting materials from their insulating state through several doping techniques. Selected examples of the different types of doping are: (1) chemical doping by charge transfer groups, (2) electrochemical doping, (3) doping by Acid–Base chemistry, (4) photodoping and (5) doping by charge injection at a metal/semiconducting polymer interface [71–74].

However, similar to other π -conjugated polymers, the application range of unsubstituted polyaniline, is limited due to its insolubility and infusibility [40,75]. Lack of solubility of polyaniline may result from the stiffness of its main chain and the existence of a strongly conjugated π electron system. Therefore, in order to improve the solubility and induce fusibility of the stiff chain of this polymer, various procedures have been adapted: (1) doping induced processibility of PANI, suggesting the use of the so-called functionalized protonic acids (such as dodecylbenzenesulfonic and camphorsulfonic acid) in the protonation of PANI, resulting in enhanced solubility [76–79]; (2) preparation of PANI composites with thermoplastic polymers

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