

Catalytic activity of polypyrrole nanotubes decorated with noble-metal nanoparticles and their conversion to carbonized analogues



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ABSTRACT

Polypyrrole nanotubes were prepared by the oxidation of pyrrole with iron(III) chloride in the presence of methyl orange. They were subsequently used as a substrate for the reductive deposition of noble metal particles. Polypyrrole nanotubes decorated with palladium, platinum, rhodium, or ruthenium nanoparticles were characterized by electron microscopy, conductivity, energy dispersive X-ray analysis, and FTIR and Raman spectroscopies. A typical metal content varied between 15 and 20 wt.%. The catalytic activity of composites was illustrated on the reduction of 4-nitrophenol to 4-aminophenol. The carbonization of composites has been followed by thermogravimetric analysis in nitrogen atmosphere. The nanotubular morphology of polypyrrole was retained after carbonization up to 830 °C. The noble-metal nanoparticles, nanometres in size, fused to clusters during this process, except for ruthenium. Polypyrrole nanotubes were converted to a nitrogen-containing carbon and platinum nanoparticles still preserved during carbonization at 400–500 °C.

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1. Introduction

Heterogeneous catalysis plays an important role in the organic synthesis. It replaces homogeneous catalysis due to better control of reaction process and repeated use of the catalyst. As a rule, the catalyst consists of a support and active element, which is deposited as nanoparticles over the support surface. The nature of the support is important as it can affect electron configuration of active metal and even to participate in the various phases of the catalysis. The theory of the catalysis has not yet been sufficiently developed, and the search for efficient catalysts takes place in extensive experimental manner by testing various systems for specific purposes.

Conducting polymers have recently become a part of catalysts systems [1–4]. Especially electrocatalysis using conducting polymers has successfully been exploited in many devices, such as sensors, batteries of metal–air type, low-temperature fuel cells or

electrochemical sensors [5–13]. The supports for electrocatalysis require a high conductivity, large specific surface area, and good environmental and chemical stability. These features are satisfied by conducting polymers, such as polyaniline or polypyrrole, and especially by their nanostructured forms, such as nanotubes or nanofibres [14,15]. In addition, conducting polymers alone display catalytic activity [3,16–18]. This has been demonstrated in the oxidation of ascorbic acid [19], formic acid [20], hydrocarbons [21], hydroquinone [22], or in hydrogen evolution reaction [23]. Along with electronic conductivity, these polymers possess ionic conductivity, and can provide the transport of ions as charge carriers. The hydrophilicity/hydrophobicity of conducting polymers can be varied [24] and allows for the control of adsorption of both reactants and products. In addition, the catalytic synergetic effect of conducting polymers and noble metals is of promise.

Various carbons have often been used as supports for conducting polymers [20,25,26] due to their chemical stability and high surface area. The nitrogen-containing carbons represent the crossroads between conducting polymers and carbons, which have recently also become popular catalyst supports. Nitrogen-containing carbons are easily obtained by the carbonization of

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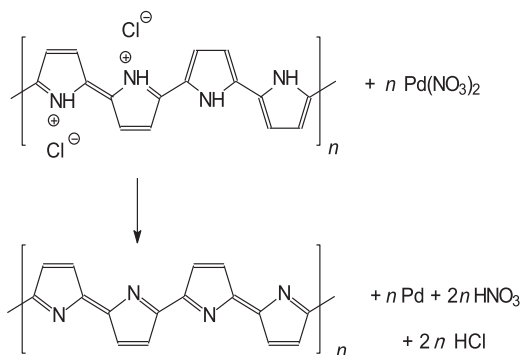


Fig. 1. Polypyrrole salt (hydrochloride) reduces noble-metal compound, here illustrated on palladium(II) nitrate, to a corresponding metal.

conducting polymers in inert atmosphere [27,28]. During such carbonization, the polymers do not melt and, consequently, their morphology is preserved [29,30], except for some shrinkage. Such materials alone have catalytic properties [31–34], and may compete with traditional platinum–carbon catalysts, especially in the oxygen reduction reaction. In the combination with platinum, they have been tested in low-temperature fuel cells [35]. The increase in the catalytic activity and improved stability has been observed in comparison with systems using plain carbons. The catalyst poisoning was reduced as well as the

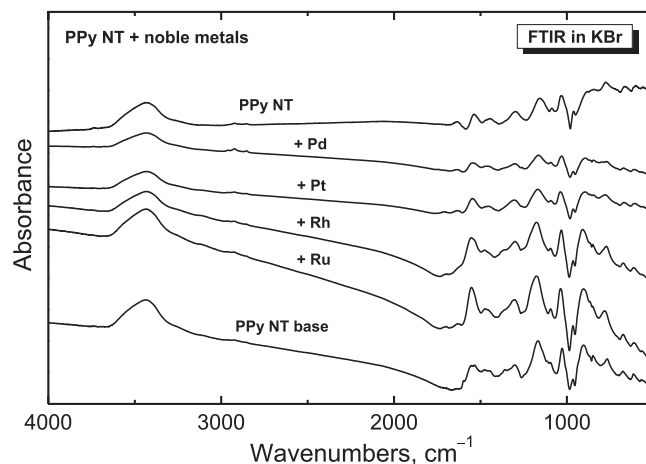


Fig. 3. FTIR spectra of original polypyrrole nanotubes (PPy NT), and of the nanotubes with deposited noble metals: palladium (+Pd), platinum (+Pt), rhodium (+Rh) and ruthenium (+Ru). The spectrum of deprotonated original polypyrrole nanotubes (PPy NT base) is shown for comparison.

coalescence of metal nanoparticles. The adherence of metal particles to the nitrogen-containing carbon support was also improved.

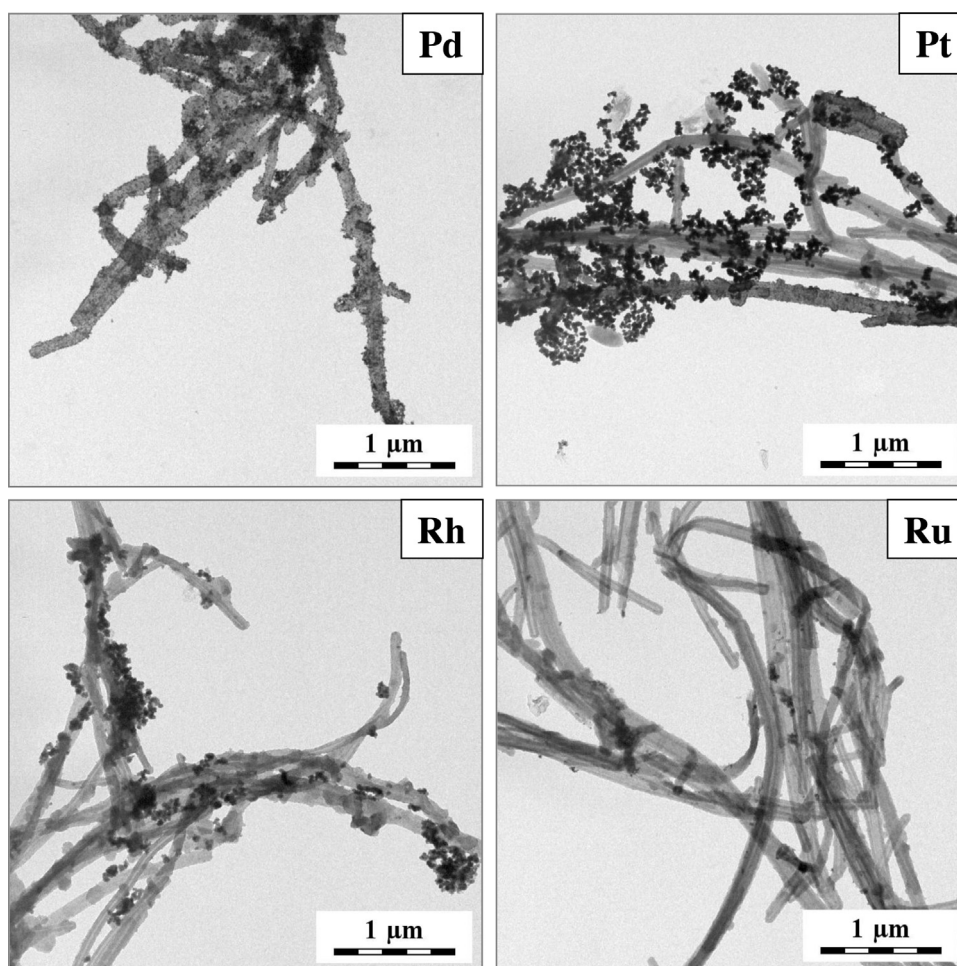


Fig. 2. Polypyrrole nanotubes decorated with palladium, platinum, rhodium, and ruthenium nanoparticles.

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