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New life of a forgotten method: Electrochemical route toward highly efficient Pt/C catalysts for low-temperature fuel cells

Igor Leontyev[a,b,*], Aleksandra Kuriganova[c], Yuri Kudryavtsev[c], Brahim Dkhil[a], Nina Smirnova[c]

[a] Laboratoire Structures, Propriétés et Modélisation des Solides, Ecole Centrale Paris, CNRS-UMR8580, Grande Voie des Vignes, 92295 Chatenay-Malabry Cedex, France
[b] Physics Department, Southern Federal University, 5 Zorge St., Rostov-on-Don 344090, Russia
[c] South-Russian State Technical University, 132 Prosveschenia St., Novocherkassk 346428, Russia

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ABSTRACT

Driven by global environmental concerns, great efforts are currently made to develop novel, cheap and practical means for producing highly efficient electrocatalysts, specifically for the low-temperature fuel-cell applications. Employing an old but yet unexplored method based on electrochemical dispergation of platinum by the alternating current, we show that the freshly prepared Pt/C catalysts exhibit greatly enhanced catalytic activity (up to 350% for oxygen reduction and up to 200% for ethylene glycol oxidation) as compared with that of a commercially available counterpart (E-TEK). A key role in these reactions is attributed to a specific nanoparticle morphology. These findings should provide a revival of an ancient but simple and efficient electrochemical process and motivate further researches in the field.

1. Introduction

Fuel cells represent a promising power source for use in a variety of systems, ranging from automotive to portable electronic devices [1]. Currently, the most promising electrocatalysts for fuel cells are those employing nanoparticles of pure or alloyed Pt deposited onto a conductive support [2]. However, the high cost of platinum has erected a barrier to wide commercialization of Pt-containing fuel cells. Accordingly, extensive efforts have been directed toward the search for either non-platinum catalysts or ways to most efficient utilization of platinum, i.e., mass production of Pt/C catalysts featuring elevated catalytic activity for oxygen reduction and fuel oxidation, CO tolerance and high aggregation stability of the constituent particles.

Apart from the composition and particle size, a decisive role in electrocatalysis involving nanoparticles is played by the particle shape [3] and crystallographic orientation [4]. As known, owing to an increased concentration of the active sites, the high-index planes of bulk Pt single crystals exhibit much higher activity than do the more common stable planes, such as {1 1 1}, {1 0 0}, and even {1 1 0} ones [5]. A similar trend was observed with Pt nanoparticles [6]. On the other hand, the high-index planes demonstrate less resistance to CO-poisoning compared to the low-index ones (which leads to a lower catalytic activity) [7]. Therefore, nanosized Pt catalysts with optimized morphology providing an increase in both the catalytic activity and resistance to CO-poisoning are obviously required for future device applications.

One of the major approaches to improving the activity of Pt-based catalysts is optimization of the synthesis procedure. Currently, various physical and chemical techniques are employed for producing Pt/C catalysts. Dozens of methods developed so far have been mostly of the condensation (bottom-up) type. The formation of the nanoparticles here proceeds via chemical reduction of the relevant precursors by different reducing agents, such as hydrogen, $NaBH_4$, or ethylene glycol which may simultaneously serve as a solvent (the "polyol" process) [8]. Other group of methods such as the atomic layer deposition [9], high-energy ball-milling [10], etc., have been used less frequently. However, all these methods present certain limitations. In fact, in the ball-milling process, it is impossible to control the shape of the nanoparticles and prevent their agglomeration, on the other hand, the reduction methods are multi-staged and highly sensitivite to extrinsic factors. In addition, the capping agents used for the shape control in the reduction methods [11] can deactivate the catalytic sites [12].

Therefore, the development of a simple and accessible method for producing catalysts with desired nanoparticle shapes and sizes has remained a challenge. In this study, we illustrate a low-cost, highly reproducible, one-step, capping agents-free, nontoxic

* Corresponding author. Tel.: +7 9185524024; fax: +7 8632975120.
E-mail address: i.leontiev@rambler.ru (I. Leontyev).

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