



# Three-dimensional hierarchical porous platinum–copper alloy networks with enhanced catalytic activity towards methanol and ethanol electro-oxidation



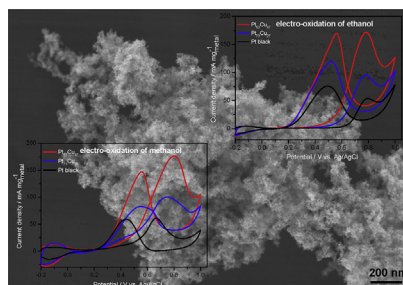
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## HIGHLIGHTS

- Three-dimensional hierarchical porous Pt–Cu alloy networks were prepared.
- Pt–Cu alloy networks were assembled by interconnected nanodendrites.
- Pt–Cu alloy networks possess high specific surface area and concave surface topology.
- Pt–Cu alloy networks exhibit enhanced electrocatalytic activity toward MOR and EOR.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Porous Pt–Cu alloy networks are synthesized through a one-pot hydrothermal process, with ethylene glycol as the reducing agent and the block copolymer Pluronic F127 as structure-directing agent. The structure, porosity and surface chemical state of as-prepared Pt–Cu alloy with different composition are characterized. The formation mechanism of the porous structure is investigated by time sequential experiments. The obtained Pt<sub>53</sub>Cu<sub>47</sub> alloy possesses a unique 3D hierarchical porous network structure assembled by interconnected nanodendrites as building blocks. Because of the high surface area, concave surface topology and open porous structure, the Pt<sub>53</sub>Cu<sub>47</sub> alloy catalyst exhibits enhanced catalytic activity towards methanol and ethanol electro-oxidation in comparison with commercial Pt black and the Pt<sub>73</sub>Cu<sub>27</sub> alloy synthesized following the same process as Pt<sub>53</sub>Cu<sub>47</sub>.

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## 1. Introduction

The development of Pt-based electrocatalysts with high specific activity, durability and reduced Pt loading is of particular significance for commercialization of low-temperature fuel cells. Among

various strategies towards this target, the synthesis of porous Pt nanostructures with large surface area, controlled porosity, morphology and shape has attracted increasing attentions [1–10]. As well known, porous nanostructures could enable increasing surface-to-volume ratio and thus increasing numbers of catalytically active sites per mass, which in turn promote Pt utilization. In addition, a three-dimensional (3D) porous nanostructure is ideal for high-performance electrode material which can not only provide continuous pathway for fast electron transport, but also

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facilitate mass transport by shortening diffusion distances. Furthermore, the partitions between nanoparticles by the porous structures greatly inhibit particle migration, Ostwald ripening and aggregation, which is of great importance for the stability and durability of electrocatalysts [1]. Recently, several methods have been explored for the synthesis of porous Pt nanostructures. For instance, Ryoo and Hyeon et al. first proposed the hard-template method, by which various nanostructured metals, including 3D networks, nanoparticles and nanowires have been created with mesoporous silica (e.g., FSM-16, MCM-41, MCM-48, and SBA-15) as hard templates [11,12]. The use of lyotropic liquid crystals (LLCs) as soft template, initially proposed by Attard et al. and later adopted by Yamauchi et al., allow formation of highly ordered mesoporous metals with lamellar, 2D hexagonal, 3D cubic and other nanostructures [4,13,14]. Dealloying is another widely used method for the fabrication of nanoporous metals, in which a less noble metal is selectively dissolved from an alloy precursor, resulting in a self-supported nanoporous structure of a noble component [7,15–17].

Recent investigations have demonstrated that Pt-based nanocrystals with highly branched morphologies possess large specific surface area and high specific activity owing to high densities of edges, corners and stepped atoms present on their branches [18]. Moreover, porous structure can be derived from the interspaces among the spatially and locally separated branches, which can provide sufficient accessible catalytically active sites at both interior and exterior surfaces [19–22]. Among various branched Pt nanostructures, Pt-based bimetallic or alloy nanodendrites are of particular interest. Seed-mediated growth was commonly used for the synthesis of Pt-based bimetallic nanodendrites. This strategy involves the synthesis of seeds nanoparticles of one metal and subsequent nucleation and growth of another metal [18]. Recently, Yamauchi and co-workers reported one-step synthesis of a series of Pt-based bimetallic dendritic nanoparticles by using block copolymer (e.g. Pluronic F127 and P123) as structure-directing agent [19–21]. Owing to the amphiphilic nature of block polymer, it could effectively mediate the growth of dendritic nanostructures through

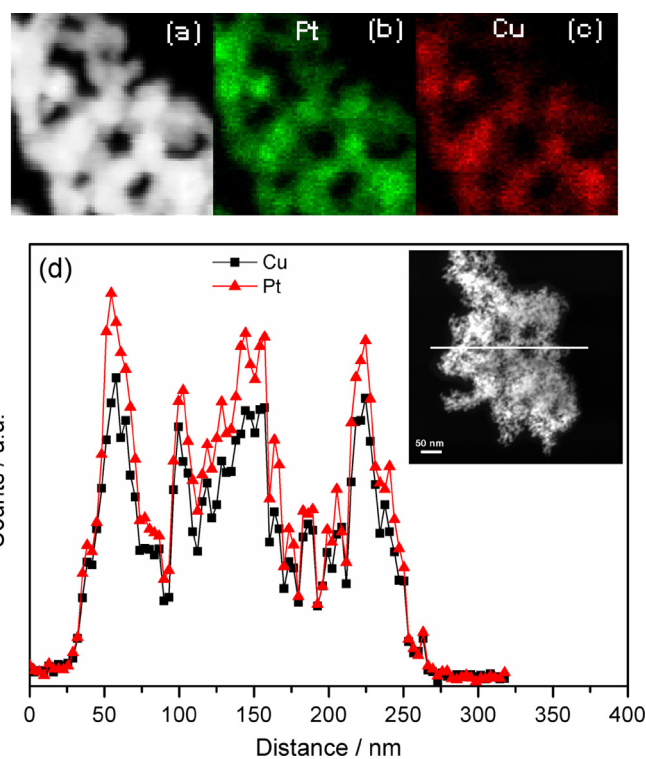


Fig. 2. (a) HAADF-STEM image, (b and c) HAADF-STEM-EDS mapping images, (d) the cross-sectional compositional line profiles of Pt<sub>53</sub>Cu<sub>47</sub>.

physical and chemical interactions with Pt precursors.

Herein, we report a one-pot hydrothermal synthesis of porous Pt–Cu alloy networks by using the block copolymer Pluronic F127 as structure-directing agent. The as-prepared Pt–Cu alloy exhibit a unique 3D hierarchical porous network structure with numerous

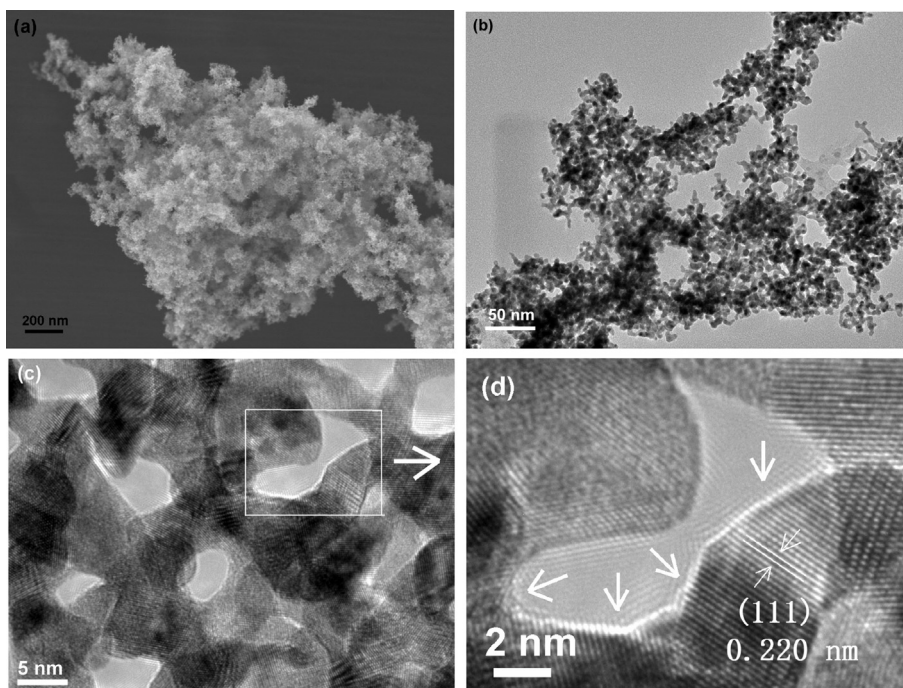


Fig. 1. (a) SEM, (b) TEM and (c and d) HRTEM images of Pt<sub>53</sub>Cu<sub>47</sub>.

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