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Facile scalable synthesis of MoO₂ nanoparticles by new solvothermal cracking process and their application to hole transporting layer for CH₃NH₃PbI₃ planar perovskite solar cells



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HIGHLIGHTS

- MoO₂ nanoparticles were synthesized via scalable solvothermal cracking process.
- The mechanism of solvothermal cracking process was systematically studied.
- The MoO₂ nanoparticles could be deposited on CH₃NH₃PbI₃ layer by orthogonal process.
- The MoO₂ nanoparticles used as dopant-free hole transporting materials in the perovskite solar cells.

$A\ R\ T\ I\ C\ L\ E\quad I\ N\ F\ O$

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ABSTRACT

 \sim 10 nm sized MoO₂ nanoparticles with high BET surface area of 170.14 m²/g were synthesized via scalable new process of combining the ultrasonic spray pyrolysis and solvothermal cracking process. Initially, we synthesized polycrystalline MoO₃ microparticles by the ultrasonic spray pyrolysis at 500 °C or 600 °C. Then the MoO₃ microparticles were disassembled into crystalline grains and the grains were subsequently shattered to small MoO₂ nanoparticles through the solvothermal polyol reduction process because the polycrystalline MoO₃ microparticles are disassembled by thermal expansion and the crystalline MoO₃ grains are shattered by volume shrinkage due to phase transition from MoO₃ to MoO₂. Finally, we applied the MoO₂ nanoparticles/toluene solution to new inorganic hole transporting material for planar CH₃NH₃PbI₃ perovskite solar cells. The planar type CH₃NH₃PbI₃ perovskite solar cells exhibited stable efficiency of 14.8% for forward scan condition and 15.5% for reverse scan condition under illumination of 1 Sun (100 mW/cm²).

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1. Introduction

Molybdenum has been well known as an alloying element in various steels to improve corrosion resistance, as well as a refractory metal used for high temperature applications [1]. Although it has oxidation states from +2 to +6, most of molybdenum oxides exist in two forms: molybdenum trioxide (MoO_3) and molybdenum dioxide (MoO_2). MoO_3 is industrially produced by roasting molybdenum disulfide (MoS_2) which is the chief ore of molybdenum [2], and has been used in various applications such as a semi-

conductor [3], a field-emitter [4], an electrochromic material [5] and a gas sensor [6,7]. Recently, MoO₂ has attracted a lot of attentions due to its unusual metallic electrical conductivity, which is associated with its mixed interatomic bonding and a relatively high density states at the Fermi level [8]. The existence of these free electrons may enhance the catalytic activity of Mo⁺⁴ in MoO₂, unlike Mo⁺⁶ in MoO₃, where all the valence electrons of the metal are bonded to neighboring oxygen atoms [9]. Due to its electrical and catalytic properties, MoO₂ has been of great interest as a fuel reforming catalyst [8–10], alternative anode material for solid oxide fuel cells [11,12], anode material for secondary lithium ion batteries [13,14], hole injection or transporting layer for organic light emitting diodes (OLEDs) [15], and hole transporting layer for solar cells (SCs) [16].

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