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Hybrid Molybdenum-Tungsten Oxide as Novel Dual-Band, VIS-NIR Selective Electrochromic Material in Advanced Smart Windows

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The energy efficiency of smart fenestration devices used in modern buildings could be greatly improved by the integration of advanced electrochromic materials. In particular, the ability to selectively control the amount of visible and near infrared (NIR) radiations transmitted through the windows could allow for independent control of light and heat supplies, offering a better suitability to respond to different climates, seasons, and users comfort. Such advanced smart windows typically involve electrochromic layers made of nanostructured highly doped metal oxides (tin-doped indium oxide ITO, aluminum-doped zinc oxide AZO), in which the selective modulation of NIR independently from the VIS behavior can be supported through localized surface plasmon resonance (LSPR) effects. In that configuration, the electrochromic material is only capacitively charged and discharged through electrochemical bias, with no redox reactions and cation insertion / extraction occurring like in conventional electrochromic metal oxides.

Therefore, there is a strong need to develop novel formulations and/or structures of electrochromic materials coupling conventional and plasmonic behaviors, so to reach a selective, dual-band modulation ability of VIS and NIR. In this context, specific formulations of nanostructured tungsten oxide (Cs-doped WO₃, WO_{3-x}) have already been highlighted as promising. Moreover, the plasmonic optical features of hybrid molybdenum – tungsten oxide have been shown to be strongly enhanced in comparison to the parent oxides, displaying an intense absorption peak around 700 nm. This resonance frequency, coupled with the stronger intensity of the signal, makes this "MoWOx" hybrid oxide a very interesting candidate as dual-band VIS/NIR-selective electrochromic material.

In this work, hybrid Mo-W oxide is successfully synthesized through a one-step hydrothermal route, with the recovered powder being dispersed in low toxic solvent and coated onto glass substrates by spin or bar coating. The morphological, structural and optoelectronic properties of the powders and

films are investigated through SEM, XRD, XPS, EPR, UV-VIS-NIR spectrophotometry and electrochemistry. Following the developed protocols, a "parent", non-hybridized WO_{3-x} formulation testifies for a "traditional" dual-band signature between "clear" (62% transmittance in VIS, 82% in NIR), "cool" (61% in VIS, 44% in NIR) and "dark" (42% in VIS, 18% in NIR) states. On the other hand, some of our selected "MoWOx" formulations - typically obtained with initial Mo/W atomic ratios of 2/1 - offers the possibility of a "warm" dual-band commutation mode that implies a darker state in VIS (modulable from 43 to 11% transmittance) and a fully-tunable NIR transmittance (from 76 to 8%). Such behavior can be directly corelated to the ratio of oxygen vacancies and the stoichiometry of metallic elements specifically observed in hybrid Mo-W formulations in comparison with the parent oxides. Preliminary cycling assessments also indicate an improved electrochemical reversibility and durability of the "MoWOx" compounds.

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