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Bias-switchable permselectivity and redox catalytic activity of a ferrocene-functionalized, thin-film metal—organic framework compound.

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Streszczenie

The installation of ferrocene molecules within the wide-channel metal–organic framework (MOF) compound, NU-1000, and subsequent configuration of the modified MOF as thin-film coatings on electrodes renders the MOF electroactive in the vicinity of the ferrocenium/ferrocene (Fc+/Fc) redox potential due to redox hopping between anchored Fc+/0 species. The observation of effective site-to-site redox hopping points to the potential usefulness of the installed species as a redox shuttle in photoelectrochemical or electrocatalytic systems. At low supporting electrolyte concentration, we observe bias-tunable ionic permselectivity; films are blocking toward solution cations when the MOF is in the ferrocenium form but permeable when in the ferrocene form. Additionally, with ferrocene-functionalized films, we observe that the MOF's pyrene-based linkers, which are otherwise reversibly electroactive, are now redox-silent. Linker electroactivity is fully recovered, however, when the electrolyte concentration is increased 10-fold, that is, to a concentration similar to or exceeding that of an anchored shuttle molecule. The findings have clear implications for the design and use of MOF-based sensors, electrocatalysts, and photoelectrochemical devices.

Słowa kluczowe

charge hopping, electrocatalysis, metal organic frameworks, permselectivity, redox shuttle

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