二次元π共役高分子構造体が創発する革新的電極過程

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High-performance yet cost-effective advanced rechargeable battery is a game-changing device for effective use of electricity supplied from wind and sun and achieve “carbon neutrality”. The criteria of advanced battery are extraordinarily challenging: (1) Energy density of 500 Wh/kg in a battery pack form; (2) Price with < 100 $/KWh; (3) Duration with > 10 years.[1] In addition, because enormous number of devices are necessary, this battery should be constituted of sustainable elements. However, so far, there is no such battery system to satisfy whole these criteria. Here two-dimensional π-conjugated coordination frameworks (2DpCFs) are shown to be one of possible candidate electrode materials for such advanced batteries.[2-5] A key property in 2DpCFs is the designer electronic structure: we can tune their electronic properties by selection of metal cation and linkers;[6] therefore, we can also control the electrochemical properties of 2DpCFs. As the result, the bis(diimino)copper framework was found to show electron-transfer-number of 3.5 with cation/anion co-redox mechanism together with a dual-ion mechanism. In this talk, the role of the cation–anion interactions for this property will be shown by using an experiment/theory collaboration applied to a series of the model electrode systems based on different metal–nitrogen bonds. These models provide designer multielectron-transfer due to the tunable π-d conjugated electronic structures. In this work, we show that, in addition to tunable electronic structures, it is found that the specific cation-anion bonds in 2DpCFs show a unique reversible rearrangement upon electrochemical Li-intercalation. This dynamic structural change can be seen as an analogue to the well-known flexibility to host-guest interactions of this type of materials, for example, upon uptaking various small molecules,[7] and this dynamic process is also an essential for a significant reversible multielectron-transfer property.

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