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Optimization of iron and cobalt based organometallic redox couples for long-term stable operation of aqueous organometallic redox flow batteries

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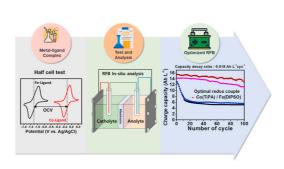
HIGHLIGHTS

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• Co(TiPA) and Fe(DIPSO) are proposed as redox couple for AMORFB full cells.

- Optimal transition metal-ligand ratio and KOH concentration are determined.
- Formation constant of organometallic redox couples is calculated.
- *In-situ* AMORFB cell is used to elucidate the performance of AMORFB full cells.
- \bullet AMORFB full cells show high CE of 99% and excellent capacity of 15.5 Ah L $^{-1}$.

G R A P H I C A L A B S T R A C T



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The optimal electrolyte condition for iron (Fe) and cobalt (Co) coordinated with Triisopropanolamine (TiPA) and 3-[Bis(2-hydroxyethyl)amino]-2-hydroxypropanesulfonic acid (DIPSO) is suggested for aqueous organometallic redox flow batteries (AMORFBs), while best combination for improving its long-term stability is determined. Co (TiPA) is optimal catholyte due to excellent redox reactivity and stability, while such benefits are proved by the comparison of formation constant which is proportional to the strength of coordination bonds within metal complex. That of Co(TiPA) is 10.8 times higher than that of Co(DIPSO) that is a competitor for catholyte. Regarding analyte, Fe(DIPSO) formed by 1:1.5 mol ratio of Fe ion to DIPSO ligand is more stable than Fe(TiPA) that is a competitor for anolyte. The stability is experimentally verified by linear sweep voltamogram. According to the measurements, In Fe(DIPSO), potential required for the solidification of Fe ions, which causes the performance degradation of AMORFB, is placed more negatively than that required in Fe(TiPA). This indicates the when Fe(DIPSO) is used, the stable cut-off voltage range for AMORFB tests is extended with its performance improvement. Furthermore, since the solidified Fe further acts as catalyst for undesirable hydrogen evolution reaction, retarding the solidification of Fe ions enhances the performance and stability of AMORFB. In in-situ AMORFB cell tests performed to validify the electrochemical evaluations of metal complexes, the stability of AMORFB using Co(TiPA) and Fe(DIPSO) is best. Regarding its performance, charge capacity is high as 15.5 Ah L^{-1} , while its capacity loss rate is low as 0.018 Ah L^{-1} cyc⁻¹ for 100 cycles.

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