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Support information for

Catalyst Durability in Electrocatalytic H_2O_2 Production: Key Factors and Challenges

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Challenges and limitations for a sustainable electrochemical H_2O_2 production

The effective commercialisation of electrochemical H_2O_2 production depends strongly on the development of cathode catalysts. Despite advancements in catalysts and cell design, the scale-up of this production is still in its early stages, constrained by challenges in designing highly active and selective catalyst materials, employing cost-effective fabrication methods, and achieving long-term operational stability. From an engineering standpoint, it is essential to reduce the costs of electrochemical H_2O_2 production to levels competitive with conventional methods such as the anthraquinone process. The costs involved in electrochemical H_2O_2 production can be brought down into electricity costs and capital costs. When disregarding operations and maintenance expenses, the total costs (C_{total}) can be expressed as $C_{total} = \frac{p_{electricity}UnF}{\lambda_{Faradaic}} + \frac{p_{capital}nF}{jt\lambda_{Faradaic}}$, where $p_{electricity}$ is the cost per unit energy of electricity (€ J^{-1}), U is the cell potential (V), n = 2 (the number of electrons transferred per H_2O_2 molecule), F is Faraday's constant, and $\lambda_{Faradaic}$ is the Faradaic efficiency. The significant role of Faradaic efficiency in affecting both electricity and capital costs underscores its importance in the economic viability of the process. Reducing cell potential can decrease electricity costs, while enhancing the operating lifetime (t) of the system lowers the capital costs. The ideal cathode catalyst should sustain high current densities without incurring potential losses or overpotentials, emphasizing the need for catalyst stability to manage costs effectively.[1]

The performance of catalysts significantly affects the cost-effectiveness of electrochemical technologies, making the enhancement of catalyst efficiency crucial for substantial cost reduction. Extended precious metal surfaces of Pt or Pd are highly active and selective towards the ORR-4e$^-$, promoting side-on oxygen adsorption and facilitating the breakage of the oxygen double bond. Conversely, the unique geometry of atomically dispersed catalysts tends to encourage end-on oxygen adsorption, which potentially impedes oxygen bond breaking and favors the ORR-2e$^-$. As a result, the most recent research in H_2O_2 electrosynthesis is intensely focused on developing noble metal alloys and atomically dispersed noble/transition metals on (heteroatom-doped) carbon materials, as discussed in section 2. SACs, in particular, are considered as next-generation ORR-2e$^-$ catalysts.[2,3]

Present challenges in scaling up synthetic processes for SACs, exemplified by a maximum yield of 1.6 kg using wet impregnation techniques, fall far short of the industrial threshold of approximately 100 kg. This underscores the critical demand for scalable and streamlined synthesis methods for SACs.[4,5] Furthermore, while augmenting metal loading in

SACs can boost catalytic conversion rates in a linear fashion, it frequently leads to aggregation, posing a barrier to large-scale manufacturing.[6]

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