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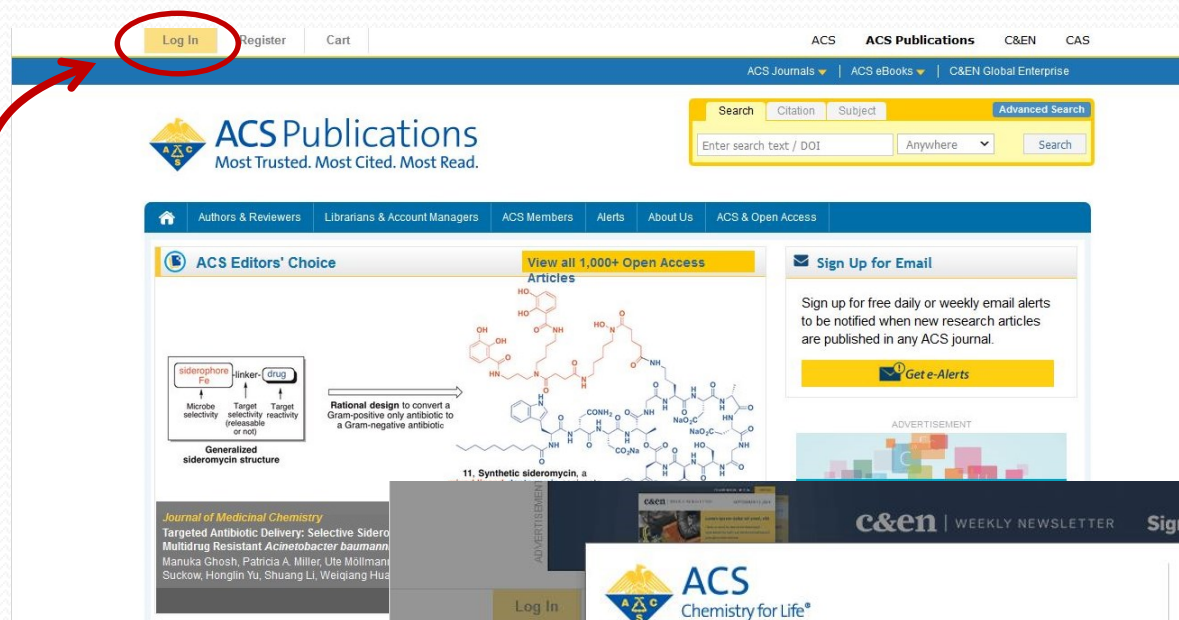
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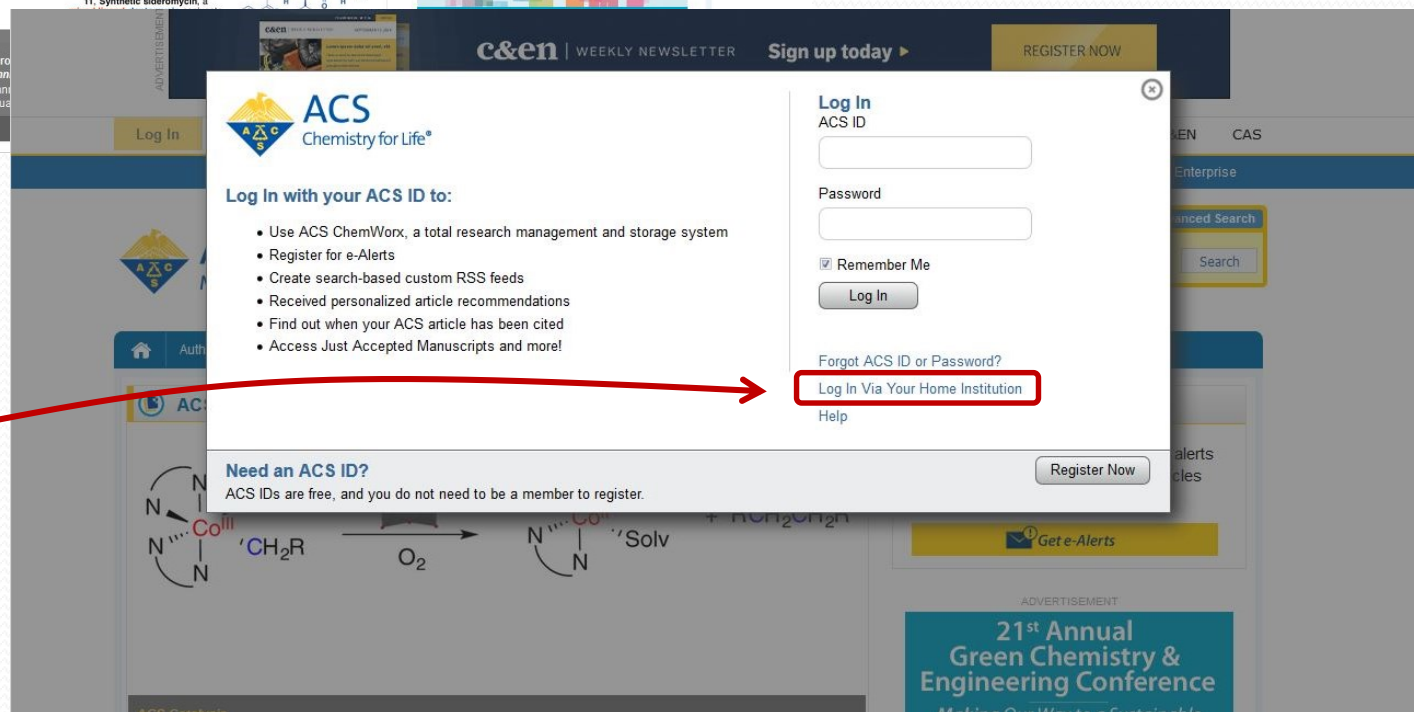
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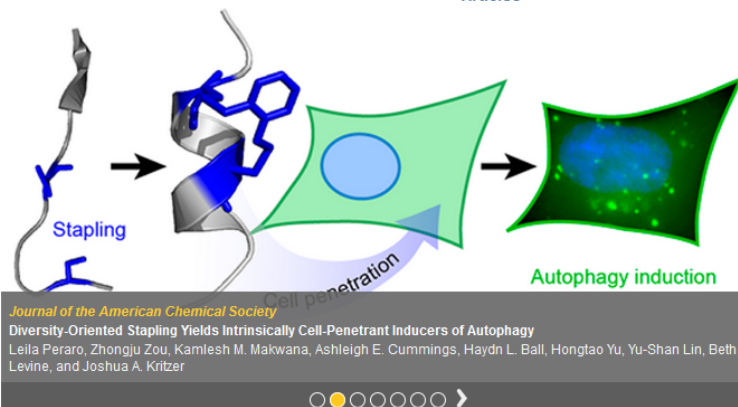


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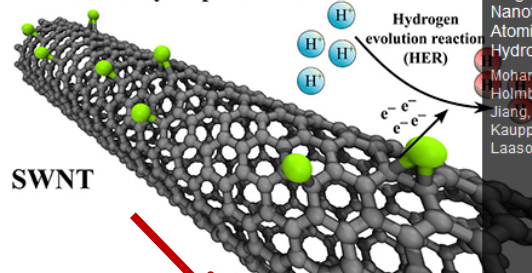
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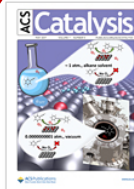
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Jessica E. Nichols  and Bryan D. McCloskey* 

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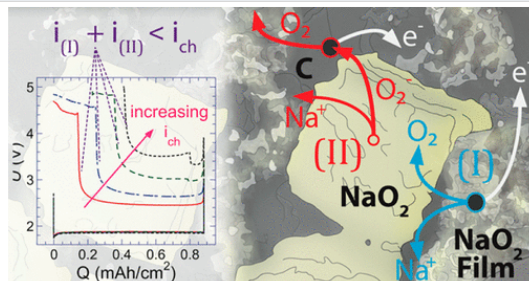
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Abstract



Metal–air (O₂) batteries have been intensely studied over the past decade as potential high-energy alternatives to current state-of-the-art Li-ion batteries. Although Li–O₂ batteries possess higher theoretical specific energies, Na–O₂ cells have been reported to achieve higher capacities on discharge and exhibit much lower overpotentials on charge than analogous Li–O₂ cells. Nevertheless, sudden and large overpotential increases (“sudden deaths”) occur in Na–O₂ cells on both discharge and charge, substantially limiting achievable capacity on discharge and increasing the average charge voltage, thereby reducing round-trip energy efficiency. In this work, we unravel the origins of these sudden death phenomena, which have been previously linked to the electrochemistry occurring at the cathode. On discharge, the maximum capacity was limited by pore clogging at low current densities and by surface passivation at high current densities, with concentration polarization playing only a small role in limiting the achievable capacity. On charge, the discharge and charge current densities were both found to influence the attainable capacity prior to sudden death. We propose a charge mechanism consistent with our data, where a concerted surface oxidation mechanism and a dissolution–oxidation mechanism both contribute to the observed overpotentials. Sudden death on charge is proposed to occur when these two pathways cannot support the applied current rate.

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