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Title:	Single-step insertion of M-Nx moieties in commercial carbon for sustainable bifunctional electrocatalysis: Mapping insertion capacity, mass loss, and carbon reconstruction
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Keywords:	Bifunctional catalysis Oxygen electrocatalysis Metal insertion
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Abstract:	Atomically dispersed earth-abundant metals in N-doped carbon (M-N-Cs) have emerged as a new class of electroactive materials that can match not only the performance of the precious metals but can catalyze both the cathodic and the anodic reactions due to their bifunctional behaviour. This inspires the development of simpler strategies for scale-up production since the existing ones rely on precursors whose commercial viability is not yet ascertained. Herein, we demonstrate the insertion prospects of M-Nx (M = Fe, Co, Ni) moieties, the electrocatalytic centers in the M-N-Cs, into commercial carbon to establish that a single-step heating of the inexpensive precursors is sufficient to generate bifunctional electrocatalysts for oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) with efficiencies that bypass the majority of the known catalysts. Further importantly, we quantify both the ORR and OER trends and the metal insertion limits for each metal while maintaining an atomic dispersion, without the formation of surface migration-induced clustering, because such clustering is inevitable in the existing processes to necessitate an extra acid-leaching step to remove them. We further quantify and explain for each metal a negative mass balance originating from anomalous mass loss of both metal and carbon content, and a massive reconstruction of the carbon backbone catalyzed by the very metal, an event documented for the first time though it ought to be associated with other M-N-C syntheses too. The study establishes an incredibly simple and inexpensive strategy for the realization of M-N-Cs and outlines the parameters to be considered during mass-production.
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