

## Production of functional carbons through chemical or electrochemical methodologies: mechanistic study and scale-up

One of the functionalized nanocarbons, graphite oxide (GO), has been known for more than 150 years, its individual layers, often termed graphene oxide, have recently gained extraordinary attention for potential use in a broad range of applications.<sup>1–8</sup> The attraction is due to the material's high chemical potential, which stems from the myriad functional groups decorated on its surface. GO is typically prepared by treating graphite with a strong oxidizer followed by exfoliation, and many variations of this methodology exist.<sup>9–12</sup> An early (1958) yet recently popular (>10,000 citations over the past five years) method was reported<sup>13</sup> by Hummers et. al. who demonstrated that GO may be obtained by exposing graphite to a mixture of  $\text{KMnO}_4$  and  $\text{NaNO}_3$  in  $\text{H}_2\text{SO}_4$ . Subsequent variations of this method have enabled access to single-layer GO and other forms of oxidized carbons.<sup>14–17</sup> Despite its prevalence, the underlying chemistry of the Hummers' method is not well understood and varying or irreproducible outcomes are often obtained despite many experimental<sup>18,19</sup> and computational<sup>20–24</sup> studies.

While in situ analyses of Hummers-type oxidations could help to enrich our understanding of the underlying mechanism(s), such reactions are typically performed in concentrated  $\text{H}_2\text{SO}_4$  and in the presence of a strong oxidant which excludes the use of conventional characterization techniques, such as X-ray photoelectron spectroscopy (XPS), FTIR spectroscopy, and elemental analysis. To overcome this limitation, we utilized in situ X-ray diffraction (XRD) and in situ X-ray absorption near edge structure (XANES) analyses with synchrotron radiation to monitor the oxidation of graphite under the aforementioned conditions. These data in conjunction with a series of atomic absorption spectroscopy, UV-Vis spectroscopy and elemental analysis measurements enabled the Hummers-type oxidation of graphite to be optimized. Ultimately, we found that additives, such as  $\text{NaNO}_3$ , were not necessary to form GO. A simple recipe that consists of only graphite,  $\text{H}_2\text{SO}_4$ , and  $\text{KMnO}_4$  facilitated the efficient oxidation of graphite.<sup>25</sup>

Not only Hummers type GO production, various methods for the production of functional carbons were developed, such as electrochemical treatment, plasma irradiation, and mechanical exfoliation

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## Figures

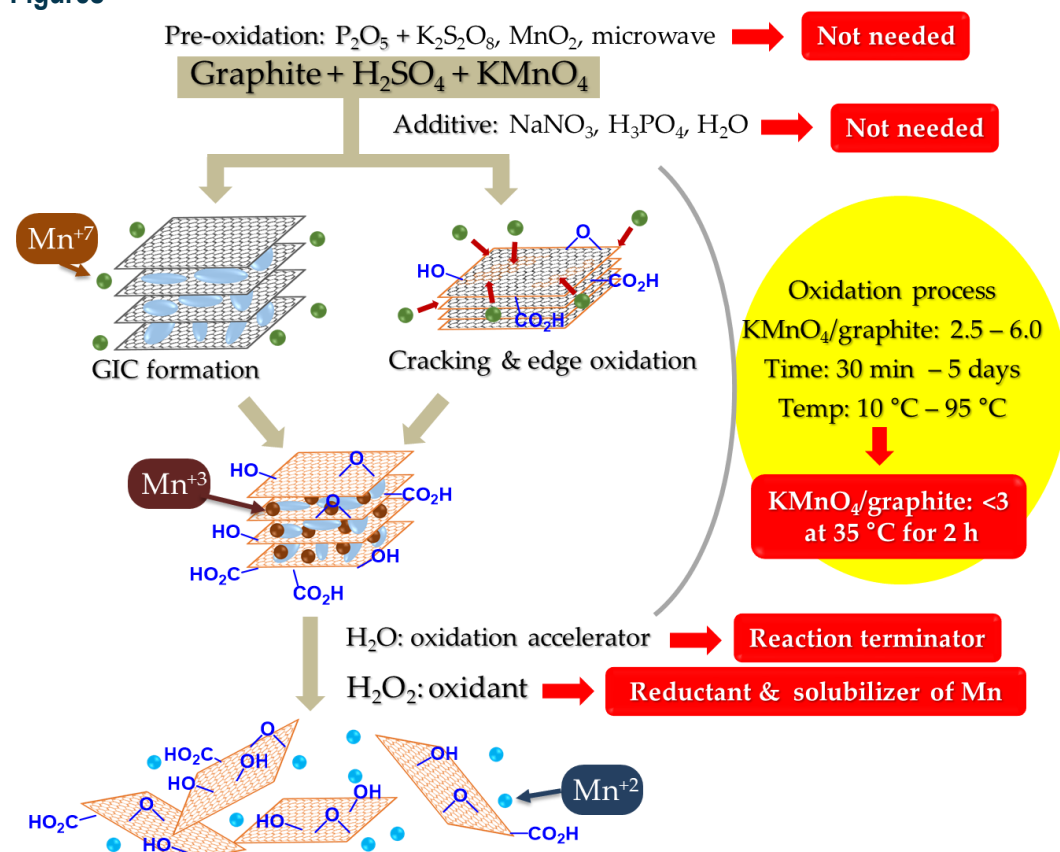


Figure 1: Optimization of graphite oxidation.