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Towards a robust fingerprint of natural graphite

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Graphite is an essential raw material in a wide range of applications due to its extraordinary chemical and physical properties. It is also a crucial raw material in energy storage; therefore, its responsible sourcing is of prime importance. In the European Union, ambitious goals were set (i.e., Critical Raw Materials Act) with respect to critical raw materials. Among others, not more than 65% of each critical raw material should stem from a single third country (i.e., outside EU) by 2030. However, currently there is no routine methodology available to distinguish natural graphite deposits from each other or trace graphite along the value chain.

In this study, we present results acquired by various analytical methods on a series of graphite samples from various stages of the value chain, spanning the range between raw ores and purified end products. X-ray diffraction (XRD), Raman-spectroscopy, carbon stable isotopy, laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) and laser-induced breakdown spectroscopy (LIBS) provide complementary structural and chemical information about the studied samples. XRD sheds light on the phases accompanying graphite and the structural change of graphite at various stages during processing, whereas Raman-spectroscopy reveals structural differences between graphite flakes from different origins, as a result of differing geological evolution and processing. On the other hand, the bulk $\delta^{13}C$ isotopic composition of graphitic carbon, largely inherited from its precursors, provides information about the origin of carbon. Complementary to that data, LA-ICP-MS and LIBS yield *in-situ* geochemical information about the chemical impurities in graphite/graphite products. LA-ICP-MS excels in low detection limits for a large suite of elements, whereas LIBS is ideally suited for acquiring quick multi-elemental maps, which provide invaluable spatial information about chemical heterogeneities. Using the above dataset, we discuss to what extent each method or the combination of multiple methods can be used for traceability. Given that individual methods yield data of different nature, opportunities for data fusion and multivariate classification are also presented.

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