

Influence of trace As or Ni on pyrite precursors and nucleation kinetics at low temperature: high resolution mineralogical and crystallochemical analysis

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Pyrite formation in sedimentary environments is a key step in the biogeochemical cycles of Fe and S. Analyses of pyrite composition can be used as proxies to infer redox conditions prevailing in ancient sedimentary environments at the time of precipitation. However, the mechanisms of pyrite formation at low temperature are recurrently discussed because of its various potential mineralogical precursors involved at the molecular level. Parameters favouring or slowing pyrite nucleation have to be determined given this mineral's pivotal role in anoxic surface environments. While traces of certain elements such as As and Ni are widely observed in sedimentary pyrite, their influence on pyrite precipitation kinetics when present in solution prior to pyrite formation has been scarcely studied. Additionally, understanding the geochemistry of As and Ni during pyrite formation is crucial to predict the trapping and release of these potentially toxic trace elements in contaminated environments.

We present recent results that show the influence of trace level As and Ni in pyrite nucleation kinetics at low temperature [1]. Pyrite syntheses were carried out at ambient temperature under conditions representative of early sediment diagenesis in the presence or absence of As or Ni. The mineralogy and the crystal-chemistry of the solids were monitored during pyrite formation at the molecular scale using X-ray diffraction, X-ray absorption spectroscopy at the S and Fe K-edges, wide angle X-Ray scattering coupled with pair distribution function and SEM-EDXS microanalysis. We show that (1) the Fe-S mineralogical precursors of pyrite are identical in each scenario and correspond to a continuum of multi-sized FeS-type particles and (2) a delay or speed-up of pyrite formation is induced by the presence of traces of As or Ni in the initial solution, respectively. We suggest that their modes of incorporation to pyrite's precursors differ, explaining these antagonistic effects. These results imply that the presence of impurities of some trace elements could have a significant impact on pyrite formation in natural media and point