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Nucleation of aragonite upon carbonation of calcium oxide and calcium hydroxide at ambient temperatures and pressures: a new indicator of fire-related human activities

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Abstract

Wood ash found at archaeological sites is the most direct evidence of the presence of combustion features. The preservation of wood ash within archaeological deposits is often poor due to diagenetic processes and therefore its identification requires the use of spectroscopic methods. FTIR spectrometry is able to detect the main calcite component of wood ash, unless this is mixed with other calcitic phases of geologic origin. Using FTIR spectrometry we identified non-biogenic aragonite in archaeological heat-altered sediments and lime plasters. The presence of aragonite in such conditions is unusual, as it is unlikely to crystallize at ambient temperatures and pressures or in absence of Mg. The experimental conditions which favor the nucleation of this aragonite phase were investigated with FTIR spectrometry, XRPD and SEM imaging of modern lime plaster and quicklime samples prepared in the laboratory using different calcium carbonate starting materials. We show that aragonite forms at ambient temperatures and pressures upon carbonation of calcium oxide and calcium hydroxide together with calcite and its nucleation and growth are influenced by environmental parameters, such as carbon dioxide partial pressure, relative humidity and temperature. This pyrogenic aragonite is a reliable indicator for calcareous materials exposed to temperatures above 600 °C, and therefore it can be used to determine the presence of heat-altered sediments and ash in the absence of structured combustion features. This mineral phase could have applications in radiocarbon dating as well.

Keywords: aragonite; calcite; lime plaster; quicklime; FTIR; pyrotechnology; fire; microarchaeology