**FRAMBOIDAL PYRITE FORMATION BY THE OXIDATION OF IRON (II) MONOSULPHIDE BY HYDROGEN SULPHIDE.** I. B. Butler<sup>1</sup> and D. Rickard, <sup>1</sup>Department of Earth Sciences, Cardiff University, Park Place Cardiff, CF1 3YE, Wales, UK (ButlerIB@cardiff.ac.uk).

**Introduction:** The ubiquity of framboidal pyrite occurrences in modern and ancient sedimentary environments and in low to medium temperature ore deposits suggests a robust formation mechanism able to operate under a range of physico-chemical conditions. Furthermore, the appearance of framboidal pyrite subjacent to the onset of anoxia in sediments and water columns is consistent with a formation mechanism which is geologically instantaneous. Finally, the highly spatially ordered microarchitecture present in many examples of framboidal pyrite (e.g. A in figure) implies a formation mechanism which permits self-organisation of individual microcrystals during framboid formation.

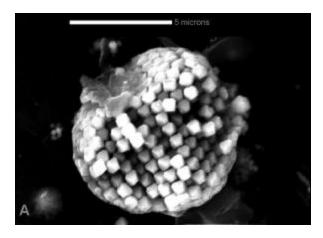
**Experimental Results:** Pyrite framboids have been synthesised via the oxidation of FeS (mackinawite) by H<sub>2</sub>S [1,2] in aqueous solutions at pH 6, Eh> -250mV, and temperatures above 60°C in the absence of O<sub>2</sub> (B in figure). Under these conditions, at Eh<-250mV, single crystals of pyrite are formed. The reaction proceeds via dissolution of solid FeS and subsequent reaction of an aqueous FeS cluster complex with H<sub>2</sub>S [3]. Circular structures on the surface of individual microcrysts indicate crystal growth via the development of screw dislocations. Greigite (Fe<sub>3</sub>S<sub>4</sub>), which has been heavily implicated in framboid formation models [4,5] was not formed in any experimental runs.

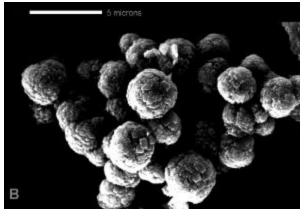
**Geological Implications:** The oxidation of FeS by  $H_2S$ :

 $FeS_{(mk)} + H_2S_{(aq)} = FeS_{2(py)} + H_{2(aq)}$ 

is by far the most rapid pyrite forming reaction so far identified [1,3]. The involvement of an aqueous FeS cluster complex in the reaction facilitates transport and reaction, allowing the development of comparatively coarse grained framboidal pyrite textures from the extremely fine grained dispersed FeS reactant in sedimentary environments. We suggest that the apparent Eh dependence of the reaction may be a supersaturation effect related to position within the pyrite stability field in Eh-pH space. The appearance of framboids close to the initiation of bacterial sulphate reduction in anoxic sediments and water columns may be unrelated to the presence of electron acceptors other than  $H_2S$  as has been previously supposed.

The size and form of the pyrite products of our reactions (both euhedral and framboidal) point to a nucleation vs crystal growth rate control over the nature of the pyrite texture. Framboidal pyrite appears to formed under conditions favourable for rapid, cascade style nucleation, but leading to slow, possibly substrate-limited growth. Single crystals indicate slow nucleation and normal crystal growth without substrate limitation. The results suggest that the framboidal texture results from rapid nucleation in environments where pyrite is strongly supersaturated. Where pyrite is metastable, or only slightly supersaturated, rapid pyrite nucleation is inhibited and single crystals form. The results demonstrate that pyrite framboids can form in the absence of molecular  $O_2$ , magnetic (e.g. greigite) intermediates or biological intervention.





**References:** [1] Rickard D. (1997) *GCA*, *63*, 115–134. [2] Butler I. B. and Rickard D., in preparation. [3] Rickard D. and Luther G. W. III (1997) *GCA*, *63*, 135–147. [4] Sweeney R. E. and Kaplan I. R. (1973) *Econ. Geol.*, *68*, 618–634. [5] Wilkin R. T. and Barnes H. L. (1997) *GCA*, *61*, 323–339.