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Published version

SCIAU, P., RELAIX, S., GOUDEAU, P., BELL, Anthony M. T., JONES, R. L. and PANTOS, E. (2006). X-Ray synchrotron study of phase transforms in illite clays to extract information on sigillata manufacturing processes. In: Synchrotron Radiation in Art and Archaeology 2006, Berlin. (Submitted)

X-RAY SYNCHROTRON STUDY OF PHASE TRANSFORMS IN ILLITE CLAYS TO EXTRACT INFORMATION ON SIGILLATA MANUFACTORING PROCESSES.

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The technique of sigillata really began in central Italy during the first century B. C. with the development of red vitrified slips obtained through vitrification of a clay preparation. These ceramics, usually decorated with raised motifs and standardised shapes, quickly took over as semi luxury crockery. Given this success, this technique quickly extended to the entire Italian peninsula and then to the Mediterranean coast. From the very start of our era, great centres of production were set up in the south of Gaul.

The aspect of sigillata comes from the nature and the texture of its slip. Studies have shown that sigillata slips of quality were obtained from a non calcareous clay while the local calcareous clay was used for the bodies. During firing the slips are vitrified and get a specific microstructure containing hematite and nanometric corundum crystals [1]. An investigation of the clays surrounding La Graufesenque site started and it seems that only the Trias levels are chemically compatible with the composition of antique slips. Apart from the in depth study of the mineralogical nature of these clays realized at a geological Laboratory, we have studied the structural transformations as a function of temperature of two of these clays, chosen for the quality of vitrification in the firing temperature range of sigillata [1030-1080°C]. The main difference between the chemical composition of these two clays is the amount of Mg (2.4 % and 4.5 % in oxide weight).

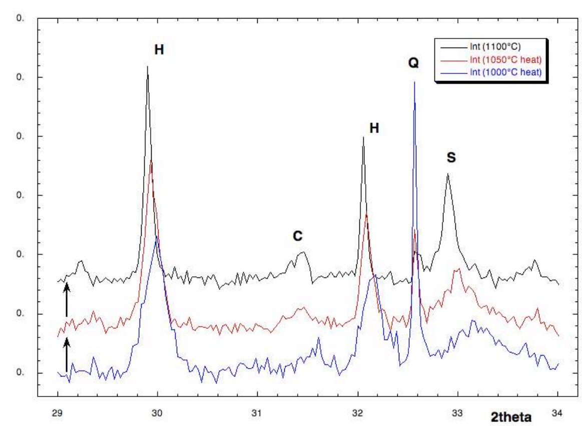

Fig. 1 Formation of spinel (S) and corundum (C) phases during the heating (clay with 2.4 % of MgO)

Time-resolved measurements were made at Daresbury (station 2.3) up to 1100°C in oxidizing conditions. An abrupt increase of the hematite cell was observed around 850°C. Above 1000°C, the hematite peaks get sharper which indicate an increase of coherence length (Fig. 1). A spinel phase with cell parameter close to $MgAl_2O_3$ was detected from this temperature. As for the hematite, its coherence length increases with the temperature but also during the beginning of the cooling. For the clay sample with the smaller amount of Mg, a corundum phase with very small coherence length was detected above 1000°C. Slips were prepared from the last clay by modern potters and firing at 1050°C in oxidizing atmosphere. A mineral quantitative analysis performed using the Rietveld method revealed that the amount of spinel phase is very high while the corundum contributes to a small part of crystal phases. It is the inverse in the antique slip where the amount of Mg in oxide weight is around 1%. It is clear that the amount of Mg plays a key role in the corundum/spinel competition and that the present slips contain too much Mg. Two questions arise: (i) As the Trias levels are quite heterogeneous is it possible to find clay with less Mg? and (ii) Did the gallo-roman potters eliminated a great part of Mg during the slip preparation process? We discuss the merits of these two alternative hypotheses.

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1. Sciau Ph., Relaix S., Chabanne D., Roucau C., Kihn Y., *J. Am. Ceram. Soc.* **89** [3], 1053 (2006)