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Ban Chiang pottery: thermoluminescence dating problems

We published, in 1972, a note by Bennet Bronson and Mark Han on a series of thermoluminescence dates from Thailand (1972, 322-6) and, two years later, comments by Dr H. H. E. Loofs on these dates (1974, 58-62). We now publish further comments from Gary W. Carriveau of the Detroit Institute of Arts, Michigan, 48202, and Garman Harbottle, also of the Brookhaven National Laboratory, who call for a critical re-examination of the published thermoluminescence dates from Ban Chiang. Since their publication, thermoluminescence (TL) dates from the site of Ban Chiang, Thailand (Bronson & Han, 1972; Ichikawa & Nakagawa, 1973) have raised questions concerning the reliability of the applied thermoluminescence dating techniques for this South East Asian material. Loofs (Loofs, 1974) has commented on the consequence of accepting these dates (which extend back into the fifth millennium BC), and the implications that this material would then be the oldest pottery in Asia. Furthermore, his paper gives cause for concern when one considers that this pottery has repeatedly been found in association with copper, bronze, iron and glass.

The results of recent research (Carriveau & Harbottle, 1981) illustrate one reason why Ban Chiang material has been so problematic for TL dating and suggest that a critical review of their published TL dates is in order.* The new experimental technique is used to measure certain departures from secular equilibrium in the uranium and thorium decay chains; most notably the emanation of radon and thoron gas. In most age determinations, the annual dose rate is calculated *assuming* that individual members of decay chains are in secular equilibrium (Carriveau &

Troka, 1979; Bell, 1976, 1977). When this assumption is not met, serious errors occur in age determinations. Our new technique, which uses experimentally-measured natural gamma ray intensities, can be used to check the assumption, and then to assess departures from secular equilibrium. In some cases, appropriate corrections can be made, ensuring the most accurate age determination. Experimental details of the method are contained in the papers of Carriveau & Harbottle (1979, 1981).

Table 1 contains data used to study several of the possible departures from secular equilibrium in the uranium decay scheme. These data appear as gamma ray intensities from naturally occurring radio-isotopes, normalized to the intensity of the 609.3 keV transition in the isotope Bi-214. The columns labelled CANMET series and IAEA series are obtained from radiometric standards considered to be in equilibrium (uranium ore mixed with clay). These values can be used as 'bench mark' values for material in secular equilibrium. Normalized intensities from material in question can then be compared against the 'bench marks'. Those that exhibit secular equilibrium in the decay chain will produce normalized activities within the range set by the bench mark material. If a match is not found, then a departure from equilibrium can be seen.

* Fleming appears to support the acceptance of the early TL dates. He also makes the curious statement that Ban Chiang wares 'are usually quite undatable because of their lack of significant TL output' (Fleming, 1979, 10). This evidence is quite contrary to this author's (GWC) experience, and to that of others engaged in TL dating of Ban Chiang material (Han, 1974, pers. comm.; Huxtable, 1978, pers. comm.; van Zelst, 1978, pers. comm.; Bortolot, 1980, pers. comm.).

Isotope	Energy (keV)	CANMET Series	IAEA Series	Ban Chiang	
				988	2114
Th-234	92.8	0.30-0.44	0.30-0.42	0.86	0.94
Ra-226	186.0	0.52-0.59	0.56-0.71	0.73	0.83
Pb-214	295.22	0.94-0.97	0.97-1.00	0.89	0.98
	351.99	1.52-1.57	1.57-1.58	1.53	1.64
Bi-214	609.3	1.000	1.000	1.000	1.000
	1120.4	0.18-0.21	0.17-0.18	0.20	0.22
Th-234					
		0.64	0.58	1.18	1.13
Ra-226					

Table 1. Normalized intensities for selected transitions in the uranium decay chain

Table 1 also contains data comparing the normalized intensities from two groups of Ban Chiang material with the bench marks, clearly illustrating non-secular equilibrium for this material. (Sherd material from the 1975 excavation, square D4. BCES 988, layer 14; BCES 2114, layer 21.) Although the values for the Pb-214 and Bi-214 match with the bench mark values, the Th-234 and Ra-226 normalized intensities clearly indicate non-secular equilibrium. This behaviour, the pre-radon normalized intensities higher than expected, could possibly be explained by emanation of radon gas. However, FIG. 1 shows an absence of emanation from this material. This is indicated by the lack of an increase of post-radon (Bi-214 and Pb-214) or post-thoron (Bi-212 and Tl-208) daughter products as a function of time after the experimental test cell is sealed (see Carriveau & Harbottle, 1979, for details). These emanation measurements have an experimental uncertainty of the order of a few per cent. Similar measurements were made on sherd material from a different level and also showed no emanation. Therefore we must conclude that the departure from secular equilibrium occurs before the gaseous phase of radon or thoron. This phenomenon has been seen in other geological and ceramic material. As to sherds from Hong Kong (Carriveau & Harbottle, 1981; Wu & Kendall, 1981), one may question whether the material can be correctly dated with thermoluminescence techniques.

Dose rate calculations, based on an assumed secular equilibrium condition, would be seriously in error for these cases. This would be most serious when one used a technique where the radio-active parents are measured, using instrumental neutron activation analysis (Carriveau & Sayre, 1978) or

X-ray fluorescence spectrometry. It is difficult to estimate the magnitude of the error without having some knowledge of the amount and type of departure from equilibrium; errors up to hundreds of per cent are possible. Alpha counting measurements (Aitken, 1974, 98), would lead to less error as the method tends to be self-compensating; however, the correction is not exact.

Fortunately, a new method of measuring departures from secular equilibrium has been reported by Murray & Aitken (1981) and may be used to measure accurately most departures from equilibrium and, in some cases, appropriate corrections can be made for dose rate calculations. This procedure, however, requires a special high-efficiency solid state detector and sophisticated computer analysis and most probably cannot be used for regular dating determinations.

In conclusion, because of the evidence that the assumption of secular equilibrium in the uranium decay chain of some Ban Chiang sherds is not valid, we suggest that published thermoluminescence dates from Ban Chiang be examined critically. All previous calculations were based on assumed equilibrium, possibly introducing serious errors. For future age determinations of Ban Chiang material, equilibrium conditions must be accurately measured and appropriate corrections must be made whenever possible.

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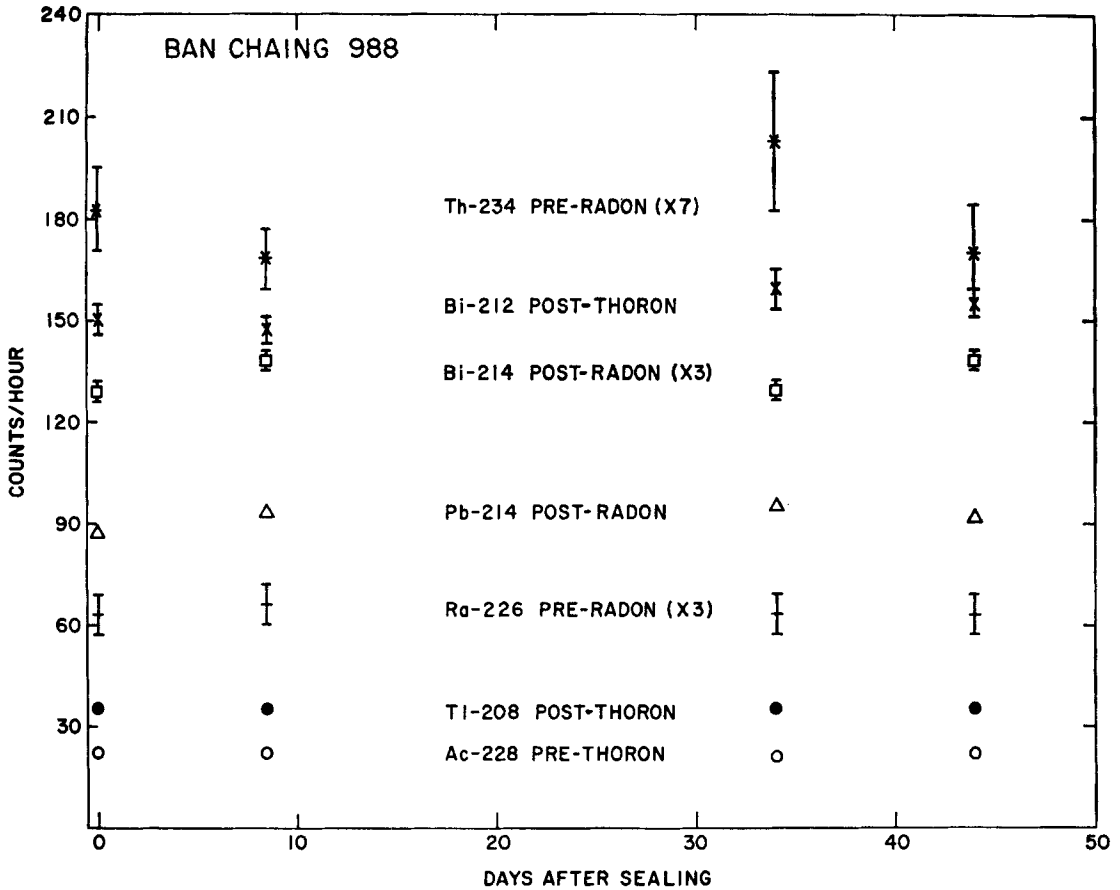


Fig. 1

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The Sutton Hoo Research Project

We all welcome the news of the new five-year research programme based on the Anglo-Saxon barrow cemetery at Sutton Hoo in Suffolk,

planned under the joint aegis of the Society of Antiquaries of London and the British Museum. These two organizations, together with other