STUDIES IN THERMOLUMINESCENCE DATING IN AUSTRALASIA

by

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Except as otherwise acknowledged or accredited in the text, the contents of this thesis are entirely my own work. No part of this work has been submitted to any other university or similar institution.

W.T. Bell
Canberra
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SUPPORTING PAPERS (available reprints in back pocket)

Bell, W.T., 1976. The assessment of the radiation dose-rate for thermoluminescence dating, *Archaeometry*, 18(1), p107.

Bell, W.T., 1976. Thermoluminescence dating of cooking stones from the Kuk Tea Research Station, New Guinea, *Arch. and Phys. Anthrop. in Oceania*, XI, No. 1, p51.

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Bell, W.T. and Zimmerman, D.W., 1978. The effect of HF etching on the morphology of quartz inclusions for thermoluminescence dating, *Archaeometry*, (in press).

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ABSTRACT

This thesis is presented in two parts: Part I dealing with the theory of the thermoluminescence phenomenon and with the origins and effects of the natural radiation environment; and Part II dealing with three applications of the TL dating method to archaeological sites in Australia.

The first chapter describes the theories and mechanisms thought responsible for the thermoluminescence phenomenon. The application of these theories to the principles of age determination using quartz is explained and a review of the thermoluminescent properties of quartz itself is given. Nothing further has been added to knowledge already extant in these areas as the purpose of this chapter is to lay the foundation for the work found later in the thesis.

A full description of the setting up and calibration of the TL dating equipment, including the alpha counter, is given in the second chapter.

The third chapter deals with the passage of alpha particles through matter. The theory of Howarth (1965) is modified in this chapter so as to quantify the absorbed dose from alpha particles passing through a 105 micron quartz grain and to describe the alpha dose reduction due to HF etching. The results are significantly different from the "first order approximation" given by Fleming (1969, 1970).

In the fourth chapter the theoretical approach of Charlton (1970) for solving the problem of the energy dissipation of electrons passing through matter is discussed. This method is modified in this chapter to be applicable to a quartz grain irradiated by the natural radioactive series. The contribution from the internal conversion electrons is included in the dose attenuation factors. The assessment of the dose dilution resulting from etching the grains in HF is then given.

The fifth chapter describes qualitatively the interaction of gamma

radiation with matter. The concept of energy-absorption buildup factors is described and is used in the evaluation of the absorbed dose from an overlying clay layer. The dependence of the TL response on both the photon energy and the particle size is explained and a method for formulating a general solution of this problem is put forward. Specific solutions for the samples to be dated in Part II of this thesis are obtained.

Chapter six presents detailed radiation data concerning the naturally-occurring radioactive series. Tables giving the energy released during every transition of every radioisotope are given. These data are used to evaluate dose-rate conversion factors from ppm of the parent and from the alpha activity of the sample to mrad/yr. The information given in this chapter has been published.

The seventh chapter commences Part II of the thesis and it is concerned with the TL dating of ancient Aboriginal fireplaces from Lake Mungo in western New South Wales. The inclusion technique of TL dating is used but particular attention is paid to the assessment of the alpha particle contribution to the dose-rate because of the irregularity of the etching of quartz grains in HF acid. Other complicating factors which are also considered in detail are the saturation of the electron traps, the internal radioactivity of the quartz grains, and the gamma dose from an overlying stratigraphic layer of much higher radioactive content than the fireplaces themselves. The ages of the Mungo fireplaces range from 31,400 to 36,400 years which confirms the antiquity of the site as suggested previously by radiocarbon age determinations on charcoal from the fireplaces.

The radiocarbon ages are given in the eighth chapter and a small systematic discrepancy of between 10 and 15% appears to exist between them and the TL ages, with the TL ages being the older in each case. A full description of possible sources of error in the radiocarbon method is presented and one of these, the variations in the Earth's geomagnetic field, is put forward as a possible reason for the TL/C-14 age differences.

The ninth chapter describes a further application of the inclusion technique, as modified in the seventh chapter, to Aboriginal fireplaces from Lake Jindabyne in New South Wales. The ages of these fireplaces lie between 2000 and 3000 years.

In the tenth chapter the TL dating of cooking stones from New Guinea is described. Large uncertainties in the ages for these stones were encountered and an impurity analysis of the samples suggests that this is due to the high level of impurity diffusion into the quartz grains. The pre-dose technique of Fleming (1973) was used for this investigation and the results given in this chapter have been published.

INTRODUCTION

The long usage of the term 'thermoluminescence' since its early discovery has caused the retention of the name to describe the emission of visible light upon heating of certain materials. The more appropriate term 'thermostimulation' might equally well be applied to the phenomenon to conform with the modern explanation of thermoluminescence as the liberation by rise in temperature of trapped electrons, whose transitions result in the emission of light.

The first recorded observation of thermoluminescence was undoubtedly that of Sir Robert Boyle (1663) who noted the phenomenon when he held a diamond near a hot but non-luminous piece of iron and saw it glow. Elsholtz(1676) discovered a similar property in the mineral fluorspar which was described by Oldenburg (1676) as,

> ".... said to be of this nature that it collects its light not so much from the Sun-beams, or the illuminated air, as from the Fire itself; Seeing that if some of it be laid upon a Silver or Copper-plate, under which are put some live coals or a lighted taper, it will presently Shine ..."

It was not until relatively recently that Daniels et al. (1953) suggested the possibility of geological and archaeological age determination by thermoluminescence. Initial studies by Tite and Waine (1962) gave archaeological ages to within ± 20% but this was based on a relative scale, it being necessary to calibrate the scale with similar material of known age. These studies were carried out at the Research Laboratory for Archaeology and the History of Art, Oxford which has since gone on to become one of the main centres for the development of the various absolute techniques of thermoluminescence dating.

Three of these absolute dating methods, (i) "the inclusion technique" first developed by Fleming in 1966, (ii) "the fine grain

technique" first developed by Zimmerman in 1967 and (iii) "the pre-dose technique" again developed by Fleming this time in 1973, have become the fundamental basis upon which the majority of the archaeological age determinations using thermoluminescence are carried out. Further details of these methods are given later in this thesis.

So we come to the work of this thesis itself which is essentially concerned with a review of the basic theoretical considerations involved in the dating techniques, resulting in a few cases in alteration of the previous data, and then the application of the theory to the age determination of three archaeological sites in Australasia. The relative success of these applications to the baked clay from Aboriginal fireplaces and to the cooking stones from New Guinean house sites is extremely encouraging and it is to be hoped that further work on similar materials will soon be undertaken.