Thermoluminescence Dating of Volcanic Material

Abstract

Thermoluminescence (TL) dating proposed in the early fifties for archaeological and geological materials has developed in such an extent that it is now established as a chronological framework in palaeolithic archaeology and quarternary geology. Since the early days of thermoluminescence dating research, various attempts were made to determine the cooling date of volcanic formations (Sabels, 1962 ; Aitken *et al.*, 1968 ; Hwang, 1970 ; Aitken and Fleming, 1971). It is observed that the dating of volcanic lava has its own unique set of problems. These are mainly anomalous fading, excessive spurious thermoluminescence and chemical fractionation. Various other techniques or the use of associated material is investigated to overcome these difficulties. In this paper the problems involved in the dating of volcanic materials and the methods developed to solve them will be evaluated and discussed.

PRINCIPLES OF THERMOLUMINESCENCE DATING

Thermoluminescence dating makes use of the fact that many crystalline materials such as quartz and feldspar have the ability to store energy from the natural radiation over a period of time. The stored energy exhibits itself as light when the material is heated.

Let us examine the accumulation of radioactive energy in solids more closely. When radioactive isotopes like uranium, thorium and potassium decay, they emit ionizing radiation. When the ionizing radiation interacts with the material, it knocks out electrons from the atoms. Some of the ionized electrons can return to their original states by emitting light in the process. A small fraction of the ionized electrons are trapped in lattice imperfections of the material. These electrons can stay in these traps for various durations. In TL dating, we are looking for those electrons which can stay in those traps long enough to be considered stable during the period of interest. When these solids are heated in the laboratory, the trapped electrons are released and are thus able to recombine with other types of lattice imperfections. The energy released during this transition is emitted as visible light which can be detected by sensitive photomultipliers (Fig. 1).

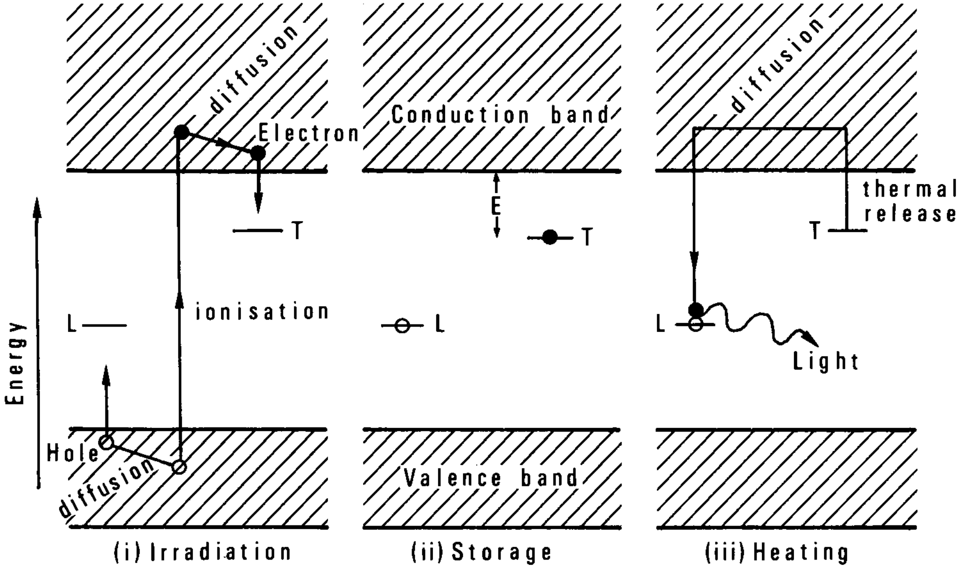

Fig. 1. Electron energy levels and allowed transitions during (i) irradiation, (ii) storage and (iii) heating, for non conducting solids. The valence band which is filled with electrons and separated by an energy gap from the conduction band (L refers to the luminescence centers and T refers to the electron traps).

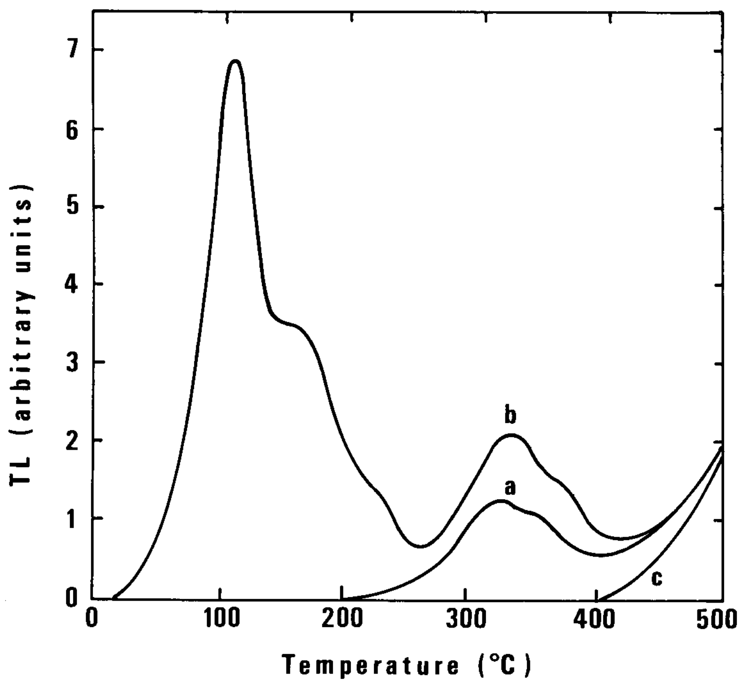

Fig. 2. A typical TL glow curve of quartz inclusions extracted from volcanic ash. a) Natural TL, b) Natural plus artificial irradiation, c) Black body radiation.

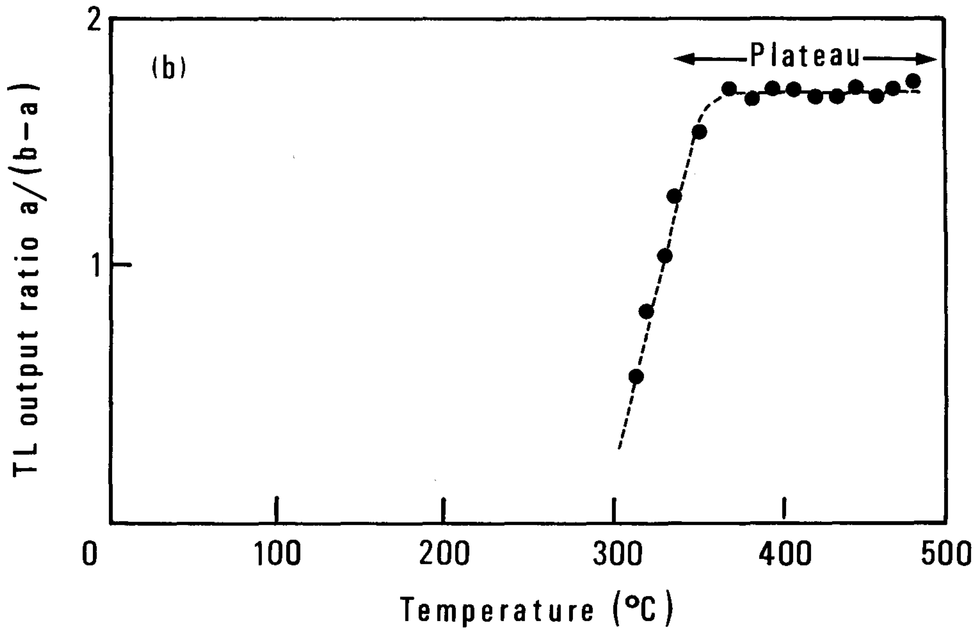

Fig. 3. Plateau Test. Plot is made of the ratio between (a) natural (b-a) and included glow. A horizontal line is obtained for regions which experience no fading.

A graphical plot of this light output versus temperature is called the « glow-curve » (Fig. 2). If we look at the light emission as a function of temperature, we will see no light at low temperatures, since the electrons trapped at relatively low levels would have already been drained by the environmental temperature cycling.

For archaeological dating, typically, the TL glow curve between 350° C and 500° C is of interest. From the kinetic studies carried out on various minerals, TL that occurs at temperatures above 350° C is likely to be stable over several hundred thousand years. However, the stable part of the glow curve have to be determined individually for each material by the « plateau test » (Aitken *et al.*, 1963) (Fig. 3). This is done by plotting the ratio between the natural and induced glow curve versus the temperature. Observation of a horizontal line is an indication of a temperature region which experienced no thermal fading. For age determination only this part of the glow curve is used (Fig. 4).

As it was mentioned above, TL emission intensity observed in the laboratory is proportional to the number of trapped electrons, therefore, the total archaeological dose the sample has accumulated since it was heated can be assessed. For this purpose one has also to take into account the specific TL emission sensitivities of various materials for a given radiation dose. This requires the measurement of TL sensitivity of each material by recording the light emission, after artificial irradiations with known doses.

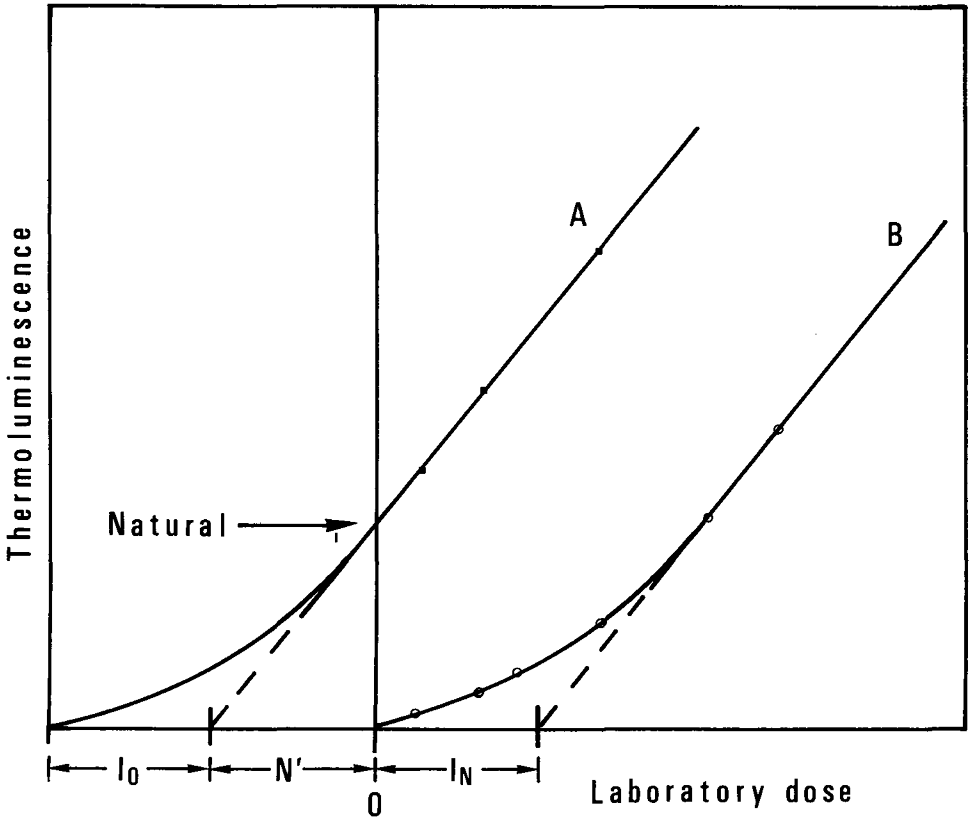

Fig. 4. Determination of archaeological dose. Curve A is the growth of natural plus induced TL, curve B is the second growth which is obtained after drainage of the natural TL. Archaeological dose is obtained by adding I_N to I_O.

For age determination, in addition to the accumulated archaeological dose, the annual dose rate has to be determined. The isotopes which are responsible for both internal and external dose rates are potassium 40, which is a gamma and beta emitter ; thorium 232, uranium 238 and their daughter products which are alpha, beta and gamma ray emitters.

Therefore thermoluminescence dating consists of two main steps. First, the determination of the archaeological dose which the sample has aquired during its history since the TL signal has been zeroed. Second, the determination of the corresponding dose rate or the annual dose. By dividing the archaeological dose with the dose rate the TL age of an object can be determined.

Complicating Factors in TL Dating of Volcanic Material

a. Anomalous Fading

It has been observed that a laboratory induced TL signal in feldspar decays in a few hours at temperatures below 400° C (Wintle, 1973). Since this decay does not follow the known decay behavior of the radiation induced centers in solids, it is called anomalous fading. It leads to an apparent decrease in age.

The anomalous fading of the feldspathic minerals is thought to be one of the main obstacles in dating. However, various indirect methods were found to avoid this problem. Huxtable *et al.* (1978) used clay minerals and sediments baked by lavas, Wintle and Brunnacker (1982) used loess in order to avoid volcanic material.

In the meantime Bailiff (1976) has reported that in some materials like plagioclase feldspar, the anomalous fading may be circumvented or at least substantially reduced by using phototransfer TL.

Another method proposed by Valladas *et al.* (1979), Guérin and Valladas (1980) was the use of high temperature region of the TL glow curve which is found to be free of the anomalous fading.

They were able to observe the TL from feldspar in the stable region of 500-700° C. In order to observe thermoluminescence in this high-temperature region it is necessary to restrict the light due to the black body radiation reaching the photomultiplier. This is done by means of a thick coloured filter. Furthermore, in order to obtain a good plateau it was necessary to eliminate this lower temperature thermoluminescence by pre-heating the sample for a minute before the glow-curve is taken.

Recently a new development was reported on the removal of anomalous fading in zircon by Templer (1985) which seems rather promissing for other volcanic material. The method is based on separation of the stable TL from the unstable component by using suitable heat or bleaching treatments. The use of such technique may give a new chance for unstable volcanic material which has not possible to date earlier.

SPURIOUS THERMOLUMINESCENCE

Thermoluminescence caused by other means than ionizing radiation is called spurious TL. It leads to an apparent increase in age. The spurious TL can arise from various sources including grinding, sawing, mechanical agitation, gas absorption, decomposition, combustion, etc. As a result the observed spurious signals appear to be associated with the surface area of the grains. Therefore one way to eliminate the spurious TL is to reduce the surface area of the samples used in the measurements. It has further been shown that it can be eliminated or reduced by using oxygen free atmosphere or proper evacuation of the heating chamber during heating of the sample (Aitken *et al.*, 1968a).

It has further been shown that some sort of spurious effect may be eliminated by chemical treatment or proper mineral separation procedure (Hwang, 1970).

DATING OF ASSOCIATED MATERIALS

The extraction of quartz portions from the lava appears a promising technique (Gillot *et al.*, 1978) despite the difficulties finding suitable quartz grains. However, dating stones baked by the lava which has flowed over them appears to be the best alternative to dating the lava itself. This approach which utilizes conventional dating techniques on quartz extracted from the stones has been successfully employed by a number of workers (e.g. Huxtable and Aitken, 1977 ; Huxtable *et al.*, 1978 ; Valladas and Gillot, 1978 ; Miallier *et al.*, 1983). One difficulty is that the thermoluminescence from the quartz is almost in saturation and therefore the natural signal is on the sublinear portion of the growth curve. Valladas and Gillot (1978) account for this by using a non-linear growth expression to calculate the equivalent dose.

APPLICATIONS

The lava in which anomalous fading has first been detected (Wintle, 1973) had come from the Chaîne des Puys in the Massif Central of France, the thermoluminescence ages obtained being low by an order of magnitude. The study was probably based on the feldspar component which exhibited anomalous fading. Few years later Valladas *et al.* (1979) used thick filters to observe the TL from plagioclase feldspar samples in the region of 500-700° C. They considered that the TL above 600° C was not subjected to anomalous fading.

When samples from this region were studied using thermoluminescence in the 600° C region of the glow curve (instead of 300° C), it was found firstly that a plateau could be obtained, secondly that any fading in a gamma-irradiated sample stored for a year was less than a few percent, and thirdly that the age obtained for the flow of Olby was (36 ± 4) 10^3 years, which was in agreement with other techniques.

Valladas and Gillot (1978) used quartz pebbles from the same flow of Olby. Quartz was almost in saturation and therefore they used non-linear growth expression to calculate the equivalent dose. By this method an age of (40.3 ± 3) 10^3 years was obtained, which was consistent with the above result. The quartz inclusion from granite found in the Lascham flow were also studied by Gillot *et al.* (1978). The mean age obtained was (34.6 ±3) 10^3 years. The various TL results of Olby and Laschamp are found to be consistent with the maximum and minimum ages for the flows provided by potassium argon and radiocarbon dates.

Baked clay from St. Saturnin and Royat has been dated by Huxtable *et al.* (1978) to an age of (8.9 ± 1.2) 10^3 years and (25.8 ± 2.2) 10^3 years which were obtained respectively. Bechtel *et al.* (1979) have reported the dating of plagioclase feldspar from a sample of basalt taken from the Mid Atlantic Ridge. They obtained a date of (203 ± 12) 10^3 years.

Another volcanic material of interest to both archaeologist and geologist is volcanic ash, which is the mixture of volcanic glass and feldspar.

Berger and Huntley (1983) used volcanic glass for dating volcanic activity. The glass has advantages over volcanic feldspars for not showing anomalous fading. With one exception good agreement with known ages was obtained, when five samples from the Seattle region in the age range 450-13000 years were dated.

Göksu (1978) used volcanic ash to determine the fossil human foot prints baked by volcanic ash. Ages of (36 ± 9) 10^3 years were obtained which were consistent with the history of the volcanic activity of the region.

Obsidian is also another volcanic product and has been investigated by TL and other methods for the purpose of distinguishing obsidians from different sources. So far no archaeological or geological TL dates have been reported (Huntley and Bailey, 1978 ; Göksu and Türetken, 1979 ; Carriveau and Nievens, 1979, and Rendell *et al.*, 1985).

CONCLUSION

TL dating of volcanic material has the advantage that the material dated is linked directly with the volcanic activity. By selecting suitable material which is free of the complicating factors mentioned earlier, an accuracy of ± 5-7 % can be achived. TL is generally less accurate than radiocarbon over the period covered by the dendrochronological calibration of radiocarbon (back to 5,000 BC). However, for periods pre-dating the dendrochronological calibration, and beyond the limit of radiocarbon (about 50,000) it is the only absolute method.

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