Journal of Radioanalytical and Nuclear Chemistry, Vol. 247, No. 3 (2001) 679–683

Luminescence dating: Limitations to accuracy attainable

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(Received August 25, 2000)

There are two components involved in evaluating age by luminescence. One is the "equivalent dose" determined from luminescence measurements on mineral crystals (usually quartz or feldspar) extracted from the material to be dated. The other is the "dose rate" to which the crystals have been exposed throughout antiquity. The age is then the ratio (equivalent dose)/(dose rate). Factors which influence the accuracy of the two components, and so the accuracy of the age, are discussed. Limiting factors are identified in order to recognize aspects of measurement on which future development must concentrate to achieve an improvement in accuracy of age determination.

Introduction

There has been no general account of all the factors influencing the accuracy attainable in luminescence dating since the texts by AITKEN[1] or by FLEMING.[2] The question of overall accuracy has not figured prominently in more recent reviews, for example by AITKEN[3] or by WINTLE,[4] although tests for accuracy in specific aspects of the dating procedure do figure therein. The intention here is to summarize the factors which limit the accuracy of an age determined using luminescence methods, identifying those for which improved accuracy has been achieved over the past decade and those for which there may be potential for improvement. Particular attention is paid to factors common to the various luminescence methods, whether thermoluminescence of long pedegree[5–7] (reviewed by AITKEN)[1] or optically stimulated luminescence,[8,9] all conveniently summarized by WINTLE.[10]

Accuracy or uncertainty is discussed numerically in terms of standard deviation in this discussion.

Age determination

In luminescence dating, age = (equivalent dose)/(dose rate), where the "equivalent dose" is determined from luminescence measurements on mineral crystals (usually quartz, feldspar or less commonly calcite) extracted from the material to be dated and a present day measurement of the "dose rate" from natural radioactivity to which the crystals are exposed is usually assumed to have applied throughout antiquity. Uncertainty in the age comes from both uncertainty in the equivalent dose and in the dose rate. In both cases the uncertainty will comprise a systematic uncertainty arising for example from the uncertainty in the calibration of the equipment and a random uncertainty arising for example from Poisson statistics associated with counting of luminescence photons and sample to sample variations due to inhomogeneity of the source material.

Uncertainty in equivalent dose

In essence, the luminescence from crystals of a mineral extracted from the material to be dated is compared with the luminescence resulting from exposure of the crystals to a known radiation dose in the laboratory in order to deduce the radiation dose accumulated by the crystals throughout antiquity, denoted the equivalent dose. Uncertainty can arise from the calibration of the laboratory radiation source as well as from the luminescence measurement procedures and the validity of basic assumptions of the process in a particular case. Table 1 summarizes many of the factors that can influence the uncertainty in the determination of equivalent dose with references in which each factor is discussed. For the sake of brevity and to avoid the need for caveats relating to particular minerals or methods, Table 1 and the following discussion concerns only quartz and feldspar (calcite is discussed by LIRITZIS)[11] and special methods, such as "pre-dose" thermoluminescence dating,[12] are not included.

The laboratory administered known radiation dose is usually provided by a ^{90}Sr beta-source with a dose rate which has been calibrated relative to a "standard" (usually gamma) source. At best, the accuracy of the beta source dose rate is limited by the accuracy of calibration of the standard gamma source. For "inclusion" dating, that is when working with mineral crystals larger than about 100 μm, only a beta source is required as it is usual to etch away the surface layer of crystal affected by alpha-particles. For "fine grain" dating, that is for crystals about 2–12 μm, the influence of natural alpha-particles cannot be avoided and the sensitivity of the crystals to alpha particles must also be determined. An ^{241}Am source is commonly used to provide alpha-particle irradiation. It is intercalibrated by luminescence measurements with a much weaker ^{241}Am source, the alpha particle emission rate of which can be determined by alpha-particle counting using a semiconductor detector.

0236–5731/2001/USD 17.00

Akadémiai Kiadó, Budapest
Kluwer Academic Publishers, Dordrecht

Table 1. Factors that can influence the uncertainty in determination of equivalent dose. Dates are given in parenthesis beside each reference in order to illustrate the chronological development of the aspects of equivalent dose determination listed

Basic process	Variant	Potential uncertainty	Reference
Laboratory irradiation	Beta (gamma) dose rate	Usually relative calibration	[13](1979)
	Alpha dose rate		[1](1985), Appendix K, [14](1982)
Thermoluminescence	Fine grain	Alpha sensitivity different from beta/gamma	[15](1971), [16](1972), [17](1984)
	Inclusion	Etching of alpha dosed surface of crystals	[18](1970), [19](1982)
	Additive dose method	Functional form of dose response	[20](1978), [21](1990)
	Regeneration method	Sensitivity change due to procedure	[22](1993)
Optically stimulated luminescence	Additive dose method	Functional form of dose response	[8](1985), [23](1994)
	Single aliquot, additive dose	Correction procedure - Feldspar	[24](1991), [25](1994), [26](1996)
		- Quartz	[27](1997), [28](1997)
	Single aliquot, regeneration	Sensitivity change compensation	[29](1998), [30](1999)
Common factors	All multiple aliquot methods	Aliquot to aliquot variations (normalization)	[1](1985) pp.126-128, [31](1992), [32](1996)
	Mineral selection	Contamination	[2](1979), [33](1994), [34](1996)
	(quartz, feldspar)		[35](1989)
	Relevance of luminescence signal	Initial zeroing	[36](1980), [37](1994)
		Stability throughout antiquity	[1](1985), Appendix E, [38](1994), [39](1997)
		Some feldspars ('anomalous' fading)	[40](1973), [41](1997), [42](1994)
	Young samples	Background signal	[43](1988), [44](1994), [45](1997)

Table 2. Some examples of accuracy achieved in equivalent dose evaluations, in Gy, with percentage accuracy in parenthesis and followed by the reference to the source of the measurements. Accuracy values of 2% or better are underlined for ease of identification. Data between > < refer to the same sample material

	Optical stimulation Single aliquot regeneration	Optical stimulation Single aliquot additive dose	Optical stimulation Multiple liquot	Thermoluminescence additive dose
Sediments				
Quartz	> 0.014 ± 0.007 (**50%**)[29]	0.013 ± 0.001 (**8%**)[28]		< 73 ± 3 (**4%**)[50]
	> 0.049 ± 0.005 (**10%**)[29]	0.026 ± 0.006 (**23%**)[28]	−0.04 ± 0.07 (**175%**)[28]	< 253 ± 11 (**4%**)[50]
	> 2.76 ± 0.09 (**3%**)[29]	2.69 ± 0.04 (**2%**)[28]	2.4 ± 0.3 (**13%**)[28]	< 282 ± 20 (7%)[50]
	> 12.4 ± 0.2 (**2%**)[29]	11.2 ± 0.3 (**3%**)[28]		<
	> 50.7 ± 1.2 (**2%**)[29]	52.3 ± 1.0 (**2%**)[28]	58 ± 6 (**10%**)[28]	<
K-feldspar	(Infrared stimulation)			
			0.52 ± 0.05 (**10%**)[51]	
			> 2.32 ± 0.08 (**3%**)[51]	1.9 ± 0.3 (**16%**)[51] quartz TL <
		> 19.2 ± 1.7 (**9%**)[24]	19.5 ± 1.6 (**8%**)[24]	20.2 ± 1.0 (**5%**)[24] <
		> 22.5 ± 1.6 (7%)[24]	22.0 ± 0.8 (**4%**)[24]	21.6 ± 1.6 (7%)[24] <
Pottery				
Heated quartz	2.02 ± 0.04 (**2%**)[30]	5.4 ± 0.7 (**13%**)[27]		8.90 ± 0.36 (4%)[52]
	5.83 ± 0.13 (**2%**)[30]	10.3 ± 0.7 (7%)[27]		
	10.76 ± 0.08 (**1%**)[30]			3.82 ± 0.08 (**2%**)[53]
	^{1}1.2 ± 0.2 (**2%**)[30]	4.18 ± 0.10 (**2%**)[53]		7.14 ± 0.67 (**9%**)[53]
	19.40 ± 0.14 (**1%**)[30]	6.14 ± 0.71 (**12%**)[53]		16.25 ± 2.07 (**13%**)[53]

Turning to luminescence measurement, half way through the three decades surveyed in Table 1, the introduction of optically stimulated luminescence can be seen which has developed in exciting ways. In particular, "single aliquot" methods can realistically, in terms of effort and quantity of sample material required, provide many independent measurements of equivalent dose for the same sample. The distribution of equivalent dose values can be examined and, if appropriate, the standard deviation derived as a measure of the random uncertainty in the mean value. This is in marked contrast to traditional multiple aliquot methods which provide only one estimate of equivalent dose.

A great deal has been established about how to use the luminescence processes to avoid erroneous equivalent dose values, as exemplified by the references in Table 1. Nevertheless, further refinement should come from a more complete understanding of the details of the physical processes involved based on a combination of experiment and numerical modeling of the processes, e.g., References 46–49.

A few examples to illustrate the accuracy (standard deviation) that has been achieved in equivalent dose determination by different luminescence procedures are given in Table 2. Generally for equivalent dose values greater than 1 Gy the accuracy is better than 10% and indeed 3% or better for many single aliquot optically stimulated measurements on quartz. To emphasise the most promising methods, accuracy values of 2% or better are underlined in Table 2.

Comparing "additive dose" and "regeneration" single aliquot methods, the former relies on measuring only a small fraction of the luminescence signal potentially available while the latter permits measurement of the whole signal. Thus for the same dose, the number of luminescence photons counted in the additive dose procedure would be much fewer than in the regeneration procedure. Since photon counting follows Poisson statistics, regeneration measurements could be expected to be more accurate than additive dose measurements. It follows that the lower limit of measurable dose should be smaller for the regeneration method. There is some indication of this in Table 2.

At very low levels of optically stimulated luminescence the background, due to an inevitable small amount of scattered stimulating light penetrating the optical filters intended to select only luminescence, may become significant. It has been shown with the dosimeter α-Al_2O_3:C, which emits luminescence with lifetimes of about 35 ms and 400–5000 ms, that pulsed simulation is beneficial in reducing background for detection of low doses.[54,55] Luminescence is detected after the end of the stimulating light pulse (of duration up to 1 s) and so in the absence of scattered stimulating light. For quartz, the half-life for the optically stimulated luminescence is only about 25 μs[56] and so for pulsed stimulation a pulse duration of the same order would be required.

Uncertainty in dose rate

The dose rate is the sum of components from beta, gamma, cosmic rays and alpha also for fine grain techniques. For dating pottery the beta and alpha dose rates required are for the material of the pottery with gamma and cosmic appropriate to the site. Measurement of the component dose rates may be by standard nuclear counting techniques (scintillation counters, semiconductor spectrometers) or by luminescence dosimeters (fluorite, $CaSO_4$:Dy) and the methods are well documented (e.g., References 1, 57–61).

Typically, the uncertainty in dose rate values is about 3% or so[27,50–53] which has generally been less important than the uncertainty in equivalent dose determination, Table 2. However with recent improvement in equivalent dose determination[30] it is now appropriate to seek to improve the accuracy of dose rate determination.

In nuclear counting methods, the detector is normally calibrated using standard ores so that the component dose rate is given by (detector counting rate)× (calibration factor) with the uncertainty dependant on both the Poisson uncertainty in the number of counts recorded and on the accuracy of the calibration procedure. The latter depends on the accuracy to which the activity of the standard source is known (of the order of 1 percent, e.g. References 62, 63) and the accuracy, which is very difficult to assess, of the factors converting activity to dose rate.[64,65]

In using luminescence dosimeters to determine dose rates, the dosimetry material must have a sufficiently high sensitivity to avoid the need for years of exposure. An important aspect is to use a material which has the same energy absorption properties as the quartz or feldspar grains used in equivalent dose determination, or to be able to correct reliably for any difference, e.g., References 67, 68. With an appropriate dosimeter, if the same beta source is use in the dosimetry measurements as in the equivalent dose measurements, then:

$$\text{age} = \frac{\text{(sample luminescence)(beta calibration factor)}}{\text{(dosimeter luminescence)(beta calibration factor)}}$$

and the beta calibration factor cancels out. However, the sensitivity of luminescence dosimeters is such that exposure for some months is required to obtain an adequate luminescence signal.

It should be possible to obtain a similar cancellation of calibration factors when using nuclear counters to determine the dose rate. In outline, a standard ore would give a known dose rate to a counter to provide the calibration factor. The same ore would give a known dose to a luminescence dosimeter, the luminescence would be measured and related to exposure time to the beta source used in equivalent dose determination, so relating the known dose to beta source exposure time. Thus the beta source calibration and the counter calibrations would derive from the dose rate from the same ore. A suitable dosimeter for such an intercalibration could be α-Al_2O_3:C, which is reported to have a linear response to dose over three orders of magnitude, and is highly sensitive when read by pulsed simulation.[54,55] The stopping powers and sizes of the dosimeter grains compared to the quartz or feldspar grains used in dating may have to be considered.[69–71]

However, accurately dose rate may be determined, there can be uncertainty about the relevance of the present day value through out the age being determined, the influence of water content for example often being of particular concern.[1,2]

Thermoluminescence methods have been developed to give age determination for pottery without requiring a knowledge of environmental dose rate, based on the difference between the equivalent dose determined from fine grains and from inclusions which should be due only to internal radioactivity of the pottery[72,73] or on the dependence of equivalent dose on the internal beta activity for different sized feldspar and quartz grains.[74] The potential of single aliquot optical stimulation methods to provide equivalent dose values of better precision than by thermoluminescence should encourage further work in this area.

Conclusions

Considering various aspects of luminescence dating which contribute to uncertainty, the following offer the prospect of improving accuracy in dating, (1) equivalent dose determination by single aliquot optical simulation methods, (2) investigation of pulsed stimulation for equivalent dose evaluation at luminescence levels such that scattered stimulating light would otherwise be a limiting factor, (3) concerning nuclear counting techniques for evaluation of dose rate, practical assessment of a calibration procedure in which counter calibration and beta source calibration cancel out, (4) re-evaluation of pottery dating methods which are independent of environmental radiation exploiting the improved precision of (1).

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