

New methods of the luminescence dating in Gliwice laboratory

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Abstract: After many years of experience with multiple aliquots method using both thermoluminescence and, more recently, optically stimulated luminescence it become possible to introduce a new, single aliquots method. The method enables determining the absorbed dose using one single portion of the sample. This eliminates many problems such as properties variation among individual aliquots of the same sample. It also makes possible to date even very small samples. The paper describes the method and procedures recently introduced in Luminescence Dating Laboratory in Gliwice in comparison to those being successfully used for a routine dating.

Key words: luminescence dating, laboratory technique, measurement protocols.

Introduction

Last few years brought a big improvement in luminescence methods. Among widely used multiple aliquot methods the new group of procedures was proposed, which could in general be described as a single aliquot method. All multiple aliquot methods for both thermally and optically stimulated luminescence and all different kind of used procedures, always require relatively big amount of material, which is a significant limitation of the method. Additionally, since luminescent properties of natural materials such, as quartz or feldspars may vary among individual aliquots of the same sample, or even single grains, the use of many aliquots is a source of an error, which can be partly reduced by some kind of normalization procedure. Thus, methods mentioned above are material and time consuming and cause some unavoidable uncertainties.

The single aliquot method first proposed by Duller (1991) eliminates those problems. Simplest idea of the method is to carry all measurements using the

same aliquot and to determine the absorbed dose for this particular aliquot. This eliminates the most popular high temperature TL method, since repeated heating may cause sensitivity changes in the material itself. Optically stimulated luminescence procedures do not require heating to temperatures high enough to cause changes in the sample sensitivity so it is the best technique to be used for a single aliquot method.

Single aliquot method

For single aliquot measurements both: infra-red and green light excitation are suitable. There also is few different procedures which can be used. In **additive dose protocol** (Duller 1991, 1995) known laboratory doses are subsequently added to the natural signal in few cycles. Every irradiation is followed by an OSL measurement. Measurement has to be short enough, not to destroy the natural signal. Yet, it always causes some signal loss. Also repeated preheating, necessary after each irradiation, cause a signal loss. Nonlinearity of the luminescence growth curve, caused by factors listed above, can be corrected using the following procedure:

- beside main aliquot, an additional one being measured, using identical procedure but without adding laboratory doses. This gives the information about signal loss in each cycle and can be used to correct the growth curve of the main aliquot luminescence;

- after few cycles with added laboratory doses, few cycles without adding any dose are performed. It may be used to check if the correction procedure is adequate.

In case of the **regeneration dose protocol** (Murray & Roberts 1998) the accumulated OSL signal must be zeroed before each irradiation cycle. The OSL measurements could be prolonged to clear the entire signal or some additional procedure has to be performed. It is the most important to choose the process that will not significantly change sample properties. This can be dependent on the sample kind. It was observed that prolonged IRSL measurement changes feldspar sensitivity up to several percent. The prolonged preheating may cause sensitivity changes in quartz samples.

Single aliquot regeneration on additive doses (SARA) (Duller 1995) is the method that unifies both procedures listed above. It requires, in fact, few aliquots. Besides natural, unchanged one, few other aliquots were preliminary irradiated with different laboratory doses as an addition to the natural signal. Single aliquot regeneration protocol is performed for all aliquots and the accumulated dose is determined separately for each one. Than the calculated doses are plotted as a function of added laboratory dose and the plot is extrapolated to obtain the corrected accumulated dose.

Procedures being used in Luminescence Dating Laboratory in Gliwice

For many years luminescent methods are in use in Luminescence Dating Laboratory in Gliwice for quartz samples. Until recently the used method required many aliquots of the same sample (multiple aliquots method). Procedure used most often for both, TL and OSL measurements, was the combined regeneration–additive method that proved to be most accurate and effective. Some of aliquots have given known, laboratory doses as an addition to the natural signal, while others were irradiated after previous bleaching with the strong light. Because we observe significant differences in luminescence properties of individual aliquots of the same sample, it is necessary to measure at least few aliquots of every kind. The table below shows a typical sequence used for dating in our laboratory:

Number of aliquots	Bleaching with laboratory light source	Laboratory dose	
8	–	–	Natural dose
5	3 h	–	Sample bleached for 3 h.
5	3 h	β_{1r}	Regeneration doses after 3 h.
5	3 h	β_{2r}	of bleaching
5	3 h	β_{3r}	
5	3 h	β_{4r}	
5	6 h	–	Sample bleached for 6 h.
5	6 h	β_{1r}	Regeneration doses after 6 h.
5	6 h	β_{2r}	of bleaching
5	6 h	β_{3r}	
5	6 h	β_{4r}	
5	–	β_{1a}	Additive doses
5	–	β_{2a}	

The measurement itself always consist of 10 s of preheating in 200°C and then, in case of TL method, a linear heating up to 500°C with a ramp rate 10°C/s, while the light output is measured and collected. In case of OSL, measurement consist of 99 seconds of green light excitation with light output detection. Many times additional procedures are required to correct errors caused by luminescence properties differences:

- preliminary IR light excitation to detect an unwanted feldspar content in quartz samples,

- normalization procedure to correct a scatter between aliquots. The procedure consists of irradiating all aliquots with identical test dose and then repeating the procedure.

As it shows, the method requires about 70 aliquots (5 mg each) of extracted quartz, which is relatively big amount, especially for small samples, or samples

with poor quartz content. Necessity of using correction procedures makes the method also time consuming. Multi-step measurement (samples are irradiated in the external beta irradiator) may negatively influence method accuracy since the sample is moved between steps and thus, can be accidentally exposed to destructive influence of light. In practice, the accuracy observed in our laboratory is about 10%.

After purchasing the additional OSL unit and the automated radiation source, which works as a part of luminescence detection unit used in TL Laboratory in Gliwice it become possible to use the new, single aliquot method. It would have many advantages: It requires smaller amount of material for dating, which allows dating even very small archaeological samples or the samples with very poor quartz content. Since accumulated dose is calculated for only one aliquot, differences of luminescence properties among aliquots has no influence on the result and provides a higher precision of dose determination (~4%). The used apparatus enable to automate the whole process. The operating system was modified for the new purposes.

To use in laboratory an additive procedure was first selected because it seems to be more useful for quartz. To avoid statistic errors the procedure is carried on few individual aliquots. Additionally some extra aliquots are used to evaluate the signal loss in each cycle. The table below shows the measurement procedure:

Number of cycle	Measuring aliquots no. - 5	Test aliquots no. - 5
1 (natural signal measurement)	preheat: 200°C/10s 10s OSL measurement	preheat: 200°C/10s 10s OSL measurement
2-7 (repeated additive dose cycles)	irradiation with laboratory dose β_1 preheat: 200°C/10s 10s OSL measurement	preheat: 200°C/10s 10s OSL measurement
8-10 (repeated test cycles)	preheat: 200°C/10s 10s OSL measurement	preheat: 200°C/10s 10s OSL measurement

We observe about 5% signal loss in each cycle. The laboratory dose β_1 should be approximately half of a value of the natural dose we expect.

The single aliquot regenerative protocol (SAR) was also tested in our laboratory. In this case the OSL measurements must be long enough to zero the signal before the subsequent cycle. Due to this and also to preheating, required before every OSL measurement, sensitivity changes may occur. To correct this an additional sensitivity tests are performed between measurement cycles (Murray & Wintle in press).

Every measurement cycle consist of a laboratory irradiation with different laboratory doses $\beta_1, \beta_2, \beta_3, \beta_4, \beta_5$, (except the first cycle, which is a measurement

of a natural signal N) followed by an OSL measurement, and then correction irradiation with test dose β_t , followed by another OSL measurement. The table below shows the possible sequence:

Number of cycle	Procedure
1 (natural signal measurement)	preheat: 220°C/10s 60s OSL measurement irradiation with test dose β_t preheat: 160°C/1s 60s OSL measurement
2–5 (regeneration dose cycles)	irradiation with one of laboratory doses β_x ($x = 1,2,3,4$ subsequently) preheat: 220°C/10s 60s OSL measurement irradiation with test dose β_t preheat: 160°C/1s 60s OSL measurement
6 (test cycle)	irradiation with laboratory dose β_1 preheat: 220°C/10s 60s OSL measurement irradiation with test dose β_t preheat: 160°C/1s 60s OSL measurement

The OSL measurement time was arbitrary chosen to ensure the signal zeroing before irradiation in the next step. It was proved that 60s of green light excitation is enough if excitation light strength is maximized.

Conclusions

The results we obtained so far prove the single aliquot method usefulness for dating. The dates we obtain for the samples which were being dated before, with the “old”, multiple aliquot method shows a high precision and are similar to previous ones, while the measurement itself was much shorter. Although we need more experience, the results are very promising, and we plane to include those new techniques in laboratory routine to increase our dating abilities.

References

- DULLER G. A. T., 1991: Equivalent dose determination using single aliquots. *Nuclear Tracks Radiation Measurements*, 18: 371–378.
- 1995: Luminescence dating using single aliquots: methods and applications. *Radiation Measurements*, 3: 217–226.

- MURRAY A. S. & ROBERTS R. G., 1998: Measurements of the equivalent dose in quartz using a regenerative-dose single-aliquot protocol. *Radiation Measurements*, 5: 503–515.
- MURRAY A. S. & WINTLE A. G. (in press): Luminescence dating of quartz using improved single-aliquot regenerative-dose protocol.