

An Experimental Study Related to Dose Scattering on the Routinely Used Minerals with Comparison of Grain Sizes and Disks Materials

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Abstract: The determination of the dose rate of a ⁹⁰Sr/⁹⁰Y radiation source prevalently used for luminescence dating studies was investigated over quartz, feldspar and polymineral aliquots in different grain sizes. These minerals are deposited over commonly used stainless-steel and aluminium discs. Luminescence dating includes protocols incorporating artificial irradiation, which is a considerably important point to understand the rate of the absorbed dose of the used samples. In this study, ⁹⁰Sr/⁹⁰Y beta sources, which are housed within Elsec and Riso commercial luminescence readers were used to compare the dose absorption ratio on fine grain (<20µ) feldspar, polymineral pottery (<20µ) and coarse grain (90-100µ) quartz. The absorbed dose rates were investigated using aluminium and stainless-steel discs with a thickness of 1 mm and a diameter of 1cm. Also backscatter and bremsstrahlung effects were experimentally investigated on stainless-steel and aluminium discs with the Ortec ZnS beta scintillation counter. As the result of this study, by comparing luminescence results dose absorption ratio by Riso's beta source irradiation was seen lower than Elsec's beta source irradiation.

Keywords: Bremsstrahlung, backscatter, luminescence, ⁹⁰Sr/⁹⁰Y, beta irradiator.

1. Introduction

In luminescence dating and dosimetry, there are several well-established protocols already reported in the literature. [1, 2, 3]. Nevertheless, on-going research still takes place in both the areas of instrumentation as well as measurements protocols. Thereafter, measurement

procedures have been developed, in which the equivalent dose is obtained on single aliquots for quartz, feldspars as well as polymineral samples [4, 5]. In single-aliquot regenerative-dose (SAR) procedures, the natural Optically Stimulated Luminescence (OSL) signal is compared to the OSL signals resulting from doses being artificially given to the same aliquot. In particular, a SAR protocol was developed [6], in which there is a correction for the sensitivity change during the reading sequence. The ease of use of this protocol, the ability to make multiple equivalent dose determinations with a limited amount of prepared quartz as well as the low relative errors have led to its widespread application mostly in OSL dating. For the former, a new reading head design using a laser source [7] or different MAAD normalisation procedures [8, 9] are trying improve the equivalent dose response.

The Riso TL/OSL reader and the ELSEC 9010 Optical Dating systems are widespread luminescence readers all around the world. In general, for irradiation, artificial beta emitters are used within commercial luminescence readers as attachments, due to the fact that both beta particles and gamma rays yield the same luminescence efficiency. The dose rates for these artificial irradiators are very swift compared to the naturally occurring environmental dose rate. Underneath the soil, samples are always dosed in a slower speed naturally; backscatter and bremsstrahlung events are not a main issue as compared to laboratory irradiation procedures. Studying with x-rays and gamma rays, the backscatter factor should be evaluated. Furthermore, the size of mineral grains with a beta source bremsstrahlung effect also should be considered.

In luminescence based retrospective dosimetry and dating studies, aliquot irradiation is very important for the accurate natural dose response determination. Almost all dating laboratories use $^{90}\text{Sr}/^{90}\text{Y}$ ($t_{1/2} \sim 30$ y), which is a well-known pure beta emitter source. Within 15 different arranged beta source experiments performed as source-sample placing at a distance of 5mm, the measured dose rates were yielded by standard deviations around 5 % and 8 % [10]. These deviations come from the nature of the radiation source, which is also related to backscatter and bremsstrahlung effects. These effects depend on sample types, sample to irradiator distance, and type of the metal disc, which holds the samples also related irradiator energy.

In addition to initial irradiation, several types of secondary radiations also create, such as secondary electrons, bremsstrahlung radiation, backscatter radiation and auger electrons. Nevertheless, both auger and secondary electrons have much less energy as compared to backscatter and bremsstrahlung radiation types. Backscattered and bremsstrahlung radiation energies come from deeper layers of the sample holding metal. This energy would increase the radiation absorption ratios of minerals especially mineral grains bonding over the surface.

Absorption dose ratio of quartz and feldspar minerals is changeable in terms of grain sizes. Furthermore, backscattered electrons will have much less energy as compared to first energetic beta rays and this would increase most of the absorbed dose in the ranges of fine grains. Aluminium filters with different thicknesses were used to harden the photon spectrums by [10]. The RISO $^{90}\text{Sr}/^{90}\text{Y}$ beta source also includes a 0.125mm beryllium filter window to harden beta rays. With this filter attachment, the beryllium window cuts off low energy beta rays. Thus, the dose dependence to grain size decreases.

In this work, the alteration of the integrated luminescence intensity was investigated for both different minerals as well as different grain sizes. $^{90}\text{Sr}/^{90}\text{Y}$ beta sources housed inside two different commercial luminescence readers were used. Finally, in this study, the impact of using a beryllium filter window on the dose absorption rate was also evaluated.

2. Experimental Protocols

All luminescence measurements were carried out using a Risø TL/OSL reader (model TL/OSL-DA-20), equipped with a $^{90}\text{Sr}/^{90}\text{Y}$ beta particle source, delivering a nominal dose rate of 0.143 ± 0.004 Gy/s. A 9635QA photomultiplier tube was used for light detection. The stimulation wavelength is 470 (± 20) nm for the case of blue stimulation, delivering at the sample position a maximum power of 40 mW/cm². For Infrared Stimulated Luminescence (IRSL), the stimulation wavelength is 875 (± 40) nm and the maximum power is ~ 135 mW/cm² [11-16]. The detection optics consisted of a 7.5 mm Hoya U-340 filter ($\lambda_p \sim 340$ nm, FWHM ~ 80 nm). Three different $^{90}\text{Sr}/^{90}\text{Y}$ beta sources were used; in the first case both Elsec beta (0.0277Gy/s) and Riso beta (0.143Gy/s) high strength beta dose sources were used for irradiation of samples and as a third beta source, a 3.5 centimetre diameter beta low activity (2.15nCi) calibration source was used. This low activity calibration source was formed by electroplating a thin $^{90}\text{Sr}/^{90}\text{Y}$ layer on to a nickel plate. This source gives the opportunity to neglect beta rays coming from side parts. By neglecting the beta rays coming from side parts, the dose absorption ratio of mineral grains are being well proportioned with vertical irradiation. The Risø TL/OSL reader $^{90}\text{Sr}/^{90}\text{Y}$ beta source has got a 0.125mm beryllium window located between the irradiator and the reader chamber. The Elsec $^{90}\text{Sr}/^{90}\text{Y}$ beta source doesn't have a radiation filter through the reading tray. The two OSL reader equipment's dose sources were used for irradiation of fine grain and coarse grain samples on aluminium and stainless-steel discs. Beta spectra measurements were performed using an

ORTEC ZnS beta scintillation counter, which evaluates within the range of 97-774 keV. The scintillation counter high voltage was set to 900 Volts.

1mm thick discs made of stainless-steel and aluminium were used for the luminescence reader device experiment, for beta spectrum experiment also 1mm thick and 3.5cm diameter aluminium and stainless-steel discs were used beneath the beta calibration source.

Experimenting with various mineral compositions of soils would affect the dose absorption ratios. Thus, global material producers' specialty materials with high-purity and high homogeneity were preferred which would give repeatable experiment results. Consequently coarse grain Merck quartz minerals in %99 purity and Eczacıbaşı K-feldspar fine grain minerals ($<20\mu$) were used. In addition, polymineral fine grain mineral ($<20\mu$) was also used. Polymineral fine grain samples are obtained from a pottery sample which was also used in this subjected article [8].

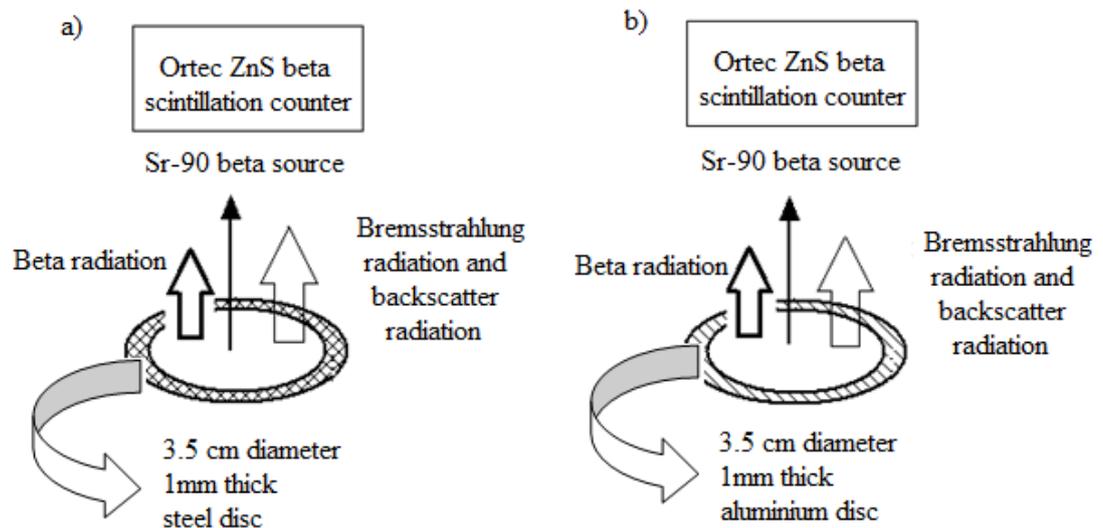


FIGURE 1. With this experimental configuration, the Ortec ZnS beta scintillation counter was also used to determine backscatter and bremsstrahlung radiation affects. Table 2 summarizes the results of this experiment. Plexiglas is not defined in this figure.

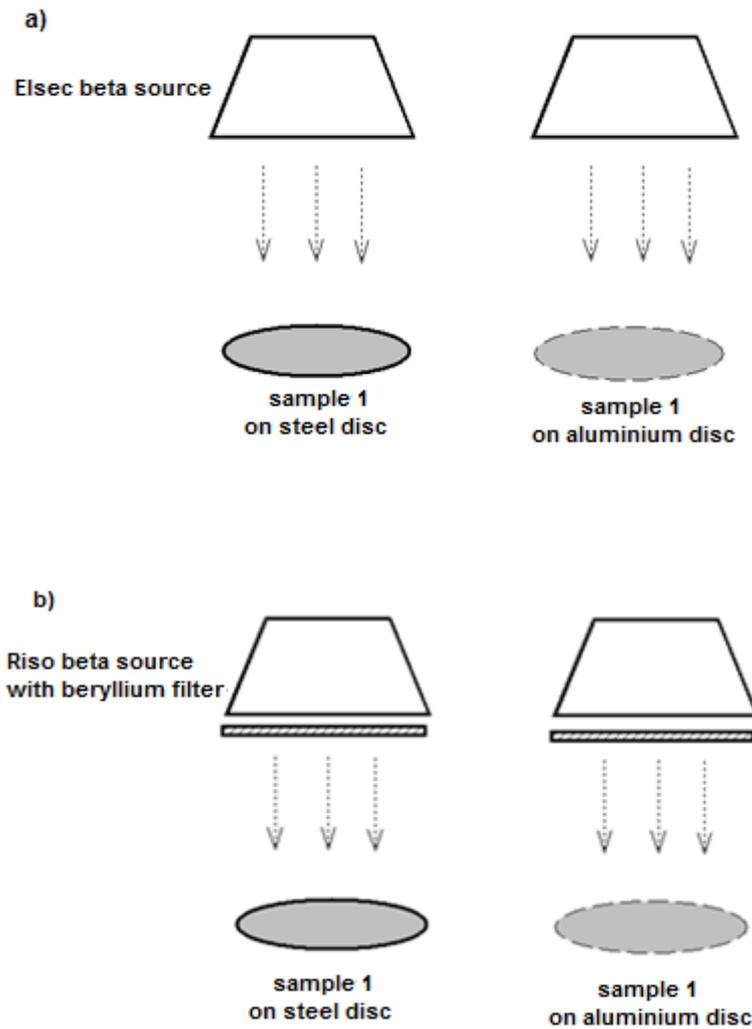


FIGURE 2. Three different mineral compositions were experimented with two different laboratory beta sources; one is the Elssec beta source and the other is the Riso beta source with beryllium filter. Three different samples were experimented, sample1: coarse grain quartz, sample2: fine grain feldspar, sample3: fine grain pottery.

2.1. Beta Spectrum Experiment Process

- Stainless steel (Ss), aluminium (Al) plates in 3.5 cm diameter and 1mm thickness were used as base material to perform the calibration beta source experiment.
- A white plexiglass material was fixed beneath the metal disc before the experiment.
- Each experiment stage was repeated five times, each time using a different metal plate (Ss,Al) and each time with a period of 600 seconds.

- The beta source was combined with a stainless-steel disc plate as seen in Fig. 1a, while spectrum measurements were performed.
- The beta source was combined with an aluminium disc plate as seen in Fig. 1b, while spectrum measurements were performed.

Plexiglass is a low density material, which gives an opportunity to neglect possible additional backscatter radiation coming from beneath the metal plates. Summarizing this part of experiment, we obtained averaged count values from the ORTEC ZnS beta scintillation counter which are summarized in Table 2. As a result of the beta spectrum measurements, bare calibration source counts were increased by 3 % after placing the calibration source on the aluminium plate. Placing the calibration source on the stainless steel plate as compared to the bare calibration source, the counts were increased by 8 %. The ratio of stainless steel aliquot counts over aluminium aliquot, an increase of 6 % counts was seen. It is sure enough to have a conclusion that the backscatter ratio from the high atomic numbered base plate would be bigger.

2.2. Quartz and Feldspar Grain Luminescence Reading Experiment

The Merck 99 % purity 90-110 μ quartz minerals, Eczacıbası (global feldspar producer for the domestic and international ceramic and glass industries) 99 % purity <20 μ K-feldspar minerals and <20 μ polymineral pottery minerals were used. One group consists of 8 quartz samples on stainless steel aliquots of 1 cm in diameter. The other group consists of 8 quartz samples on aluminium aliquots of 1 cm in diameter. Similarly, the two groups were also prepared for the case of feldspars, one of the sample groups placed on stainless steel and the other one placed on aluminium disks. All minerals were bonded on discs by using silicon spray. These four groups of aliquots were first dosed with the ELSEC $^{90}\text{Sr}/^{90}\text{Y}$ beta source after the bleaching stage; all aliquots were dosed with the RISO $^{90}\text{Sr}/^{90}\text{Y}$ beta source. Pre-irradiation, all aliquots were bleached within daylight until the background level was seen.

The experimental procedure was carried out in the following steps:

- Step 1: Bleaching of aliquots under sunlight to reset probable residue signals.
- Step 2: A beta test dose of 2 Gy was attributed to each aliquot with the ELSEC Sr90/Y90 beta dose source.
- Step 3: Quartz coarse grain aliquots were read with the RISO OSL (blue stimulation) reader, feldspar fine grain aliquots were read with the RISO IRSL reader.

- Step 4: All aliquots were bleached with daylight until the background level was seen.
- Step 5: A beta test dose of 2 Gy was attributed to each aliquot with the RISO $^{90}\text{Sr}/^{90}\text{Y}$ beta source.
- Step 6: Quartz coarse grain aliquots were read with the RISO OSL (blue stimulation) reader, feldspar fine grain aliquots were read with the RISO IRSL reader.

This experiment is summarized below in Table 1. The count ratios were calculated from the experimental results. The luminescence counts were compared with the steel/aluminium counts ratio. Irradiation with the RISO beta source steel/aluminium values yield as following 4, 3, 5 %, respectively, for the (coarse grain quartz, fine grain K-feldspar and pottery) samples.

Irradiation with the ELSEC beta source steel/aluminium count ratio values yield 16, 18, 21%, respectively, for (coarse grain quartz, fine grain K-feldspar and pottery) samples.

3. Results and Discussion

Optically stimulated luminescence dosimeter studies necessitate dosing samples with a stable dose source. In the ideal case for obtaining a stable irradiation process, the energy spectrum must be evenly distributed. Of course, in many cases this spectrum shows various values. Also backscatter and bremsstrahlung electron emissions must be minimized. Commonly used radiation sources are especially the $^{90}\text{Sr}/^{90}\text{Y}$ beta irradiator, which is easy to handle with appropriate shielding as attachments in luminescence readers in laboratories. Especially, two main minerals (quartz, feldspar) are being used in order to assess the equivalent dose in accidental retrospective dosimeter and dating. Dating related studies generally include fine grain and coarse grain mineral sizes. Grain sizes of minerals also affect the dose absorbing property of samples by backscatter factors. Furthermore, backscatter events and bremsstrahlung radiation increase the dose absorption ratios.

The backscatter build up value of the 2.15nCi beta calibration source on a nickel plate was indicated as 40 % in its calibration certificate. These backscattered beta rays will increase the average luminescence counts on dosed grains. This backscatter increase was not calculated as near 40% in Table 2. As seen in Table 2, in this experimental configuration without lower energy beta rays being monitored, the energy threshold for the ORTEC beta scintillation counter is 97 keV. These disregarded beta rays and bremsstrahlung radiations would be absorbed easily by especially fine grains [17].

TABLE 1. Experimental results of dosed samples with two different $^{90}\text{Sr}/^{90}\text{Y}$ sources

Aliquots were dosed with a Riso90Sr/90Ybeta source *

Material	Mineral size (micron)	Mineral type	Count ratio of aliquots according to base material	Average percentage of count increase
Merck %99 pure quartz	90-110 μ	coarse grain	Steel/Aluminium	4
Eczacıbaşı K feldspar	<20 μ	fine grain	Steel/Aluminium	3
Polymineral pottery	<20 μ	fine grain	Steel/Aluminium	5

Aliquots were dosed with a Elsec $^{90}\text{Sr}/^{90}\text{Y}$ beta source **

Material	Mineral size (micron)	Mineral type	Count ratio of aliquots according to base material	Average percentage of count increase
Merck %99 pure quartz	90-110 μ	coarse grain	Steel/Aluminium	16
Eczacıbaşı K feldspar	<20 μ	fine grain	Steel/Aluminium	18
Polymineral pottery	<20 μ	fine grain	Steel/Aluminium	21

*Dose strength 0.143 Gy/s, **Dose strength 0.0277Gy/s

TABLE 2. OrtecZnS beta scintillation count result of $^{90}\text{Sr}/^{90}\text{Y}$ over steel and aluminium plates (energy detection range 97 – 774 keV)

	I	II	III	II/I	III/I	II/III
	$^{90}\text{Sr}/^{90}\text{Y}$	$^{90}\text{Sr}/^{90}\text{Y}$	$^{90}\text{Sr}/^{90}\text{Y}$	Count	Count	Counts ratio
	source	source	source	increase	increase	Count ratio
	over	over 1mm	over 1mm	percentage	percentage	of steel
	plexiglass	thick	thick	steel plate	aluminium	over
	base	stainless	aluminium	over	m	aluminium
	material	steel		plexiglass	plate over	
					plexiglass	
Average of counts	10565±200	11510±287	10899±155			
Count ratios	-	-	-	8	3	6
Total experiment count time (seconds)	3000	3000	3000			

This expected result was seen in Table 1. The ELSEC dosed backscatter values were increased far more as compared to the RISO dosed samples. The ELSEC $^{90}\text{Sr}/^{90}\text{Y}$ beta rays were not filtered using a window material; in other words, soft beta rays effectively contributed in dosing all grains.

These backscattered beta rays were increased to a count ratio of 21 % in Table 1. In a study, the backscattering build up rate on stainless steel to aluminium was calculated as around 14 % [17]. In summary, the backscatter build up value from stainless steel to aluminium in Table 1 was found between 16-21 %. The RISO $^{90}\text{Sr}/^{90}\text{Y}$ source dose responses show that a 0.125 mm beryllium filter hardens beta rays and the irradiation capability over fine grains to coarse grains does not change significantly (Table 1). The Beryllium filter window in front of the

RISO $^{90}\text{Sr}/^{90}\text{Y}$ source increase the reproducibility of aliquots. The filter attachments in front of the dose source increase the reproducibility of aliquots.

4. Conclusion

As results of this study, the following highlights can be proposed. In case of dating old sediments, high luminescence count rates frequently saturate the photomultiplier tube outputs. In a situation like this, a Beryllium filter or another filter sheet would be an advantage. Radiation inducing high luminescence jumps could be withdrawn to lower levels. This could be made easier in the evaluation of oldest sediments.

In dating studies of very young samples, for example forensic science related or recent historical based samples, a bare beta source could be an advantage. The small dose rates should give larger luminescence responses. As a result, evaluation of very young samples could be easier. In this situation using a Beryllium filter could be a disadvantage. With a short time irradiation, the maximum luminescence count should be seen without filter. In this type of study, using a stainless-steel based back material underneath the sample, also dosing samples with a bare beta source could be an advantage.

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