SUMMARY

This report presents results of a short study of the thermoluminescence TL properties of talc. Talc is a magnesium silicate mineral utilised widely in industrial and pharmaceutical applications. It occurs in well defined geological situations with several known formation processes, and is extracted by quarrying and mining. The use in foods and dietary supplements is mainly as a filler. The work was initiated to address questions as to the possible impact of talc on application of EN1788 methods to detect irradiated foods.

Authentic samples were obtained from extraction and processing facilities in France and Italy. Supplementary samples from Sardinia, China, Mexico and Australia were obtained indirectly. Irradiated pharmaceutical grade talc was obtained from a UK commercial source for comparison. All samples were characterised by TL analysis after initial preparation, and also following irradiation to a 200 Gy dose. The dose response and behaviour under storage at ambient and elevated temperatures and in a light box, were investigated for two samples in detail. Exploratory kinetic analysis was performed to provide first order estimates of trap parameters.

All samples of talc exhibited measurable TL response in the 0-500°C region, with at least four recognisable TL peaks present in response to radiation. Sensitivities vary by some 3 or more orders of magnitude from sample to sample, accompanied by less pronounced changes in glow shape following radiation. The dose response increases progressively in the 100-6400 Gy range investigated. At higher dose levels the response is non-linear with the onset of saturation occurring in the kGy region for the majority of signals. Radiation induced signals from all four peaks were stable at ambient temperatures over the duration of the study. Samples stored at 50°C showed some thermal erosion of the low temperature signals. This observation together with analysis of the distribution of natural TL and the kinetic analyses confirms that lower temperature signals have comparable stability with other silicates, and that higher temperature signals are the dominant components of geologically induced signals. The stability requirements to explain natural signals in terms of simple production mechanisms have been defined. Exploratory kinetic analysis confirms that there are multi-trap contributions to the main TL peaks, but provides broad support for a simple explanation of the natural TL.

The TL response following high dose irradiation can be distinguished from geological signals by 1-2 orders of magnitude in low temperature peaks. There may be scope for further investigation of phototransfer effects, but these would clearly be overtaken by bleaching in prolonged exposures to light. Thermal processing of pharmaceutical grades of talc reduces residual low temperature signals by some 1-2 orders of magnitude leading to product with negligible low temperature natural signals. TL could in principle be used to verify the effectiveness of such thermal treatment, and potentially to authenticate commercial products. It can also quite clearly be used to identify irradiated talc, providing the amount of prior exposure to light is limited. In samples containing talc in extreme dilution with other unirradiated products, there might be ambiguity in interpretation of glow curves with minor low temperature peaks dominated by high temperature signals. Care would be needed in interpreting EN1788 results of this sort.

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GLOSSARY

Thermoluminescence (TL) - Light emitted from a heated sample in addition to black body radiation as a result of thermally stimulated release of trapped charge carriers.

TL intensity – The amount of light detected over a given temperature interval at a given heating rate, usually expressed in photon counts.

TL glow curve - Variation of TL intensity with temperature

Photostimulated Luminescence (PSL) – Light emitted from a sample exposed to optical or photo-stimulation resulting from release of trapped charge carriers from defect centres

Geological signal – TL or PSL signal induced by long term exposure to ionising radiation in the natural environment over geological timescales. Note that these signals are observed at high TL glow curve temperatures, since they originate from TL traps with long thermal stabilities.

Phototransfer – the transfer of charge from deep to shallow traps arising from photostimulation during optical illumination and subsequent re-trapping at shallow traps.

Multi-trap systems – systems or samples whose luminescence signals arise from more than one trapping state. These systems can be expected to show complex kinetics and dose response behaviour.

1. Introduction

This reports presents a short investigation of the luminescence properties of talc, a material sometimes used as a filler in the manufacture of dietary supplements. Following the 2001/2 FSA survey of undeclared irradiated foods it was suggested (Schmitt & Hungerbühler, 2002) that thermoluminescence (TL) signals from talc may confound the application of EN1788 methods to dietary supplements containing talc. The purpose of this study was to investigate the TL signals associated with talcs from various origins, including pharmaceutical grades to assess the potential impact on EN1788.

Authentic samples of mined and quarried talcs from a number of sources were obtained and characterised. Processed materials included optically sorted and thermally processed pharmaceutical grades of talc, and also irradiated pharmaceutical talcs. Both natural TL, originating from the accumulated effects of long term exposure to ionising radiation in the natural environment, and artificial TL induced by artificial irradiation were studied. Storage tests were also conducted to evaluate the stability of radiation induced TL signals at different glow-curve temperatures and to assess the effect of illumination during storage of both irradiated and unirradiated samples.

This report describes the background to the study, the samples obtained through fieldwork and supplementary activities, and the TL results of characterisation tests and storage tests. The potential impact of talc on EN1788 analysis is discussed together with an assessment of the use of TL methods to identify irradiated talc.

2. Background

Luminescence methods have been applied to detection of irradiated foods now for nearly 20 years (eg Sanderson et al, 1989, 1992, 1994,1995,1996, 1998, Sanderson & Carmichael 2000). Thermoluminescence (TL) from minerals associated with herbs, spices, seasonings, fruits and vegetables and shellfish has been studied extensively and validated methods are described in the EN1788 international standard. Photostimulated luminescence (PSL) has been validated for herbs and spices and shellfish, and can be utilised either as a screening technique (screening PSL) to select samples for further analysis, or for certain pure materials as a reference method in its own right (calibrated PSL). EN13751 describes both PSL approaches.

Detection methods for food irradiation are relevant both in respect of verification of commercial activities, and in support of regulations governing the application of ionising radiation to foods and the subsequent labelling of processed products. At time of writing the UK position permits irradiation of certain food classes, subject to licence controls administered by the Food Standards Agency, and to explicit labelling. Only herbs and spices for culinary use have been granted a licence to date, and it is understood that the quantity of material being processed in the UK is very small. European regulations, while currently permitting the diverse national regulations governing food irradiation, effectively only permit the irradiation or herbs and spices across the whole community, and also require full labelling of treated products. There is also a requirement on member states to report the results of national surveillance programmes, based where possible on validated and standardised analytical methods, to the European Commission for compilation and publication on an annual basis.

A UK survey of some 200 samples of herbs and spices, 200 samples of shellfish and some 150 samples of dietary supplements was undertaken in 2001/2, and published in 2002 (FSA,2002, EC, 2002). Both PSL and TL methods were used in this study, the analyses being undertaken at SUERC. Whereas very few cases of undeclared samples for herbs, spices or shellfish were encountered some 42% of the dietary supplements examined were either irradiated or identified as containing irradiated materials in this study. In the majority of these cases the results indicated that major proportions of the luminescence-sensitive parts of the products tested were irradiated. In a few cases the data implied minor components in a mixed product. This finding reflects an apparently widespread use of radiation in microbiological control of some of the pharmacologically active ingredients (most notably herbal ingredients) used in these products. There is evidence that the issues surrounding the use of irradiation are now more widely appreciated within the dietary supplement industry, and that steps are being taken to improve compliance with UK and EU law, partly in response to the FSA survey. Both FSA and the European Commission have called for follow up studies to monitor this process.

In the case of simple herbal products or ingredients the application of TL and PSL methods to health foods and dietary supplements is relatively straightforward. The techniques have been extensively studied and validated for many varieties of herbs and spices, and the limits of application are quite well known. However within the dietary supplement and health food products sectors there are manufacturing processes and minor ingredients, some of whose luminescence characteristics are less well known, and may require further attention to deal with marginal cases.

This study concerns talc, which is used in some tablet formulations as a filler. The name talc is applied generically to naturally derived products of various purity comprising mainly the mineral talc, a hydrated magnesium silicate $Mg_3Si_4O_{10}(OH)_2$, together with accessory minerals. The main formation processes are hydrothermal alterations of ultra-basic rocks, or metamorphic reactions between dolomites and silicates (Bowen & Tuttle, 1949, Bray 1994, Douillard et al, 2002, Read, 1934, Sharer et al 1999, Wiik, 1953). Talc has a variety of industrial uses which exploit its major physical and chemical properties (Chernovsky, 1973, Sarquis & Gonzales, 1998, Bailey & Holloway, 2000, Stixrude, 2002). As a durable silicate with specific gravity in the same range as the quartz/feldspar clay minerals recovered during standard EN1788 extractions it is likely that talc, if present in the original sample, would be recovered into the polymineral silicate suite on which routine TL analyses are based. Its signal contribution relative to other minerals present will depend on the concentrations and TL sensitivities of talc relative to the other silicates present. Whether or not this affects EN1788 analyses depends both on these considerations, and on the extent to which the TL properties resemble those of more common silicates. Although there is limited literature on cathodoluminescence of talc (Gotze et al 2002), we are not aware of systematic studies of thermoluminescence that would address these matters.

Prior to this study the suggestion that talc was responsible for adverse results obtained in the FSA survey was put forward by Schmitt & Hungerbühler, 2002. Work was undertaken by the Tela laboratory in Berlin on behalf of the Lichtwer Pharma company, whose Kwai garlic tablets had been identified as fulfilling the EN1788 criteria for the presence of minor quantities of irradiated material. TL

analysis of further tablets from the same batch reproduced the FSA result. Samples of the garlic used for production of tablets in 1999, and at later dates, however produced negative EN1788 results for irradiation. Retained samples of calcium carbonate, talc, magnesium stearate, titanium dioxide, silicon dioxide and gum Arabic, which formed part of the tablet formulation were also examined. Of these small low temperature signals and low glow ratios were obtained from the talc sample, leading to the inference that this may have been responsible for the adverse result in the FSA survey. An analysis of the dose represented by the natural signal was performed based on a linear assumption, without at that stage examining the dose response characteristics. A further 10 products, plus three additional talc samples (from Italian and French Luzenac sources) were also examined. These also showed weak TL signals in the 150-250 degree temperature range, and a distinctive series of glow peaks on re-irradiation.

SUERC had also conducted additional studies outwith the FSA survey. Pharmaceutical grade talc had been submitted for examination to address the same underlying question. In this case the glow curves as received showed peaks at 260-270 degrees and from 320 degrees and above. After irradiation a further two peaks at approximately 160 and 220 degrees were also observed. It was recognised that the shapes of the post-irradiation 4 –5 peak structure are atypical of common silicate extractions, and that in the absence of information on the stabilities of signals from all peaks in the glow curve that it would be imprudent to apply EN1788 criteria. The analytical result in this case was indeterminate with respect to irradiation, but it was noted that material of this type (whether irradiated or not) would not interfere with EN1788. Following discussions with FSA additional attention was given to the 5 garlic products tested in the 2001/2 survey. The separated samples used in the survey analyses had been retained, in keeping with practice in the SUERC TL laboratory, and were subjected to Scanning Electron Microscopy and Energy Dispersive X-ray (SEM-EDX) analysis. The glow curves were also reviewed again to assess whether signals from talc may be relevant. The results of this investigation confirmed that talc was present in the Kwai tablet, along with other silicates. The other four garlic products identified as irradiated did not have talc in significant quantities; one was dominated by Calcium Phosphate; the others showed quartz-alumino-silicate mixtures which are very consistent with normal soil derived silicates. Rutile (TiO_2) was identified in one case also. Finally some talc samples from non-commercial geological steatite sources in Shetland were subjected to TL analysis (Bray, 1994, Buttler 1989). These did not give signals that would have confounded EN1788. These results are summarised further in Appendix A.

In addition to this information it was known at the outset of the study that pharmaceutical talc is irradiated in some instances; enquiries at the Isotron company and other commercial sources had confirmed that this was the case. Indeed the European Pharmacoepia specifically mentions irradiation in a footnote indicating that this is a suitable method for producing pharmaceutical grades of talc with extremely low bacterial activity. It was thus apparent that further work was needed to ascertain the potential impact of talc as an ingredient in foods on EN1788 analyses by TL. The main objectives of this work were : i) to establish whether authentic talc source materials have TL signals that would potentially interfere with EN1788, (ii) to establish the stability of radiation induced TL signals in talc under dark and light storage and (iii) to assess whether irradiated talc can be identified by TL. A series of

samples was assembled to address these aims, focussing on materials relevant to the European sources exploited by the Luzenac talc company, and supplied to Lichtwer Pharma.

3. Fieldwork and Sampling

3.1 Aims of Sampling

        To meet the needs of this study a comprehensive set of talc samples of known provenance and treatment was required. Fieldwork was carried out by SURRC staff accompanied by Lichtwer Pharma representatives to obtain authentic quarried samples from Trimouns in France and mined samples from Val Chisone in Italy. These products represent the source materials relevant to the largest European supply of pharmaceutical talcs, based on thermally processed material. To broaden the range of samples studied additional material was obtained through the Luzenac company to represent sources in Sardinia, China, Australia and North America. A further two samples, of material originating in China, were obtained from Richard Baker Harrison Ltd in the UK representing authentic irradiated pharmaceutical grade talc and its source material.

3.2 Samples from Luzenac

    The Luzenac Group was formed as the result of the acquisition by the Rio Tinto company of consolidated talc mines and quarries in the French Pyrenees, Canada, Austria,Italy, Sardinia, USA, Mexico, Australia. The result is a group with access to 11 active ore bodies, 19 processing plants and producing some 1.5 million metric tonnes of talc per year. By far the majority of the production is for industrial use. The European operation, based from Toulouse, produces and sells some 800 ktonnes per year from 7 sources and 11 processing plants. Again the industrial uses – mainly as fillers in papers, coatings and paints, ceramics and plastics are dominant. Proprietory grades of Pharmaceutical talc (the "Luzenac Pharma" product) are currently produced (based on information from the company) by selection of material from the Trimouns quarry source near the village of Luzenac in the Pyrenees followed by thermal sterilisation, milling and packing at the Val Chisone plant in Italy. The main selection criteria for pharmaceutical grade product are a high reflectance, coupled with low acid soluble contents. Microbiological controls are undertaken to verify the quality of the final product. The pharmaceutical grades of talc were in the recent past also manufactured from material mined in the North of Italy near Val Chisone. According to the company irradiation is not used as part of the process to produce pharmaceutical talcs. The possibility that products originating from the group are sometimes irradiated on behalf of customers cannot however be ruled out.

        To track the production of Luzenac Pharma product through from source to final product it was necessary to sample both in France and Italy. This was accomplished during a short fieldtrip in October 2002, accompanied by representatives of Lichtwer Pharma and the Luzenac group.

The quarry at Luzenac (eg de Parseval et al, 1991,1994, Larrasoana et al 2003, Martin et al 2000) produces approximately 450,000 tons of talc per year, out of a group total of 1.5 M tons. Material is extracted mechanically, and then roughly sorted into piles of different raw quality. These are sampled and graded prior to transfer to silos where they await transfer to the factory in Luzenac. After rough crushing and first size sorting, the material is transferred to the factory by cable car. The main impurity is chlorite; the main colour is due to particulate graphite. At the factory the material are sorted further producing the following grades : OXO which is

approximately 80% talc; OO approximately 90% talc; and OOO which is some 95% talc. Optical sorting is used to separate stones of different colour. During the first pass items >20 mm are sorted by reflectance. Thereafter they are reduced in size and passed through the final CCD sorting stage.

3.2.1 Factory samples

Table 3.1 summarises the samples obtained at the Luzenac processing works. The Luzenac Pharma grade of talc, produced in Val Chisone, uses OOO material selected for low acid solubility. This is shipped to Val Chisone by truck for processing which includes thermal treatment at 140 degrees for a minimum of 20 minutes. Thus sample A4 is the feed material for the pharmaceutical grade of product.

Sample	SUERC Reference	Details
A1	SP5726	< 20mm OXO grade talc after optical sorting; light fraction
A2	SP5727	<20mm OXO grade talc after optical sorting; dark fraction
A3	SP5728	<20mm OXO feed material prior to sorting
A4	SP5729	20-100mm OOO material awaiting shipment to Val Chisone

Table 3.1 Samples obtained from the works at Luzenac

3.2.2   Quarry samples

The quarry is located at 1800m above sea level in a slight pass between two mountains. On the eastern side Gneiss outcrops; the west has dolomite. The central area has thermally metamorphosed talc and chlorite. The quarry is worked in 6 areas, 5 out of which were in current operation. Each of these was sampled. Reference numbers, locations and descriptions are summarised below in table 3.2.

Sample	SURRC Reference	Latitude (N)	Longitude (E)	Area	Details
B1	SP5730	42°48.832	1°48.247	Fraisse	a: loose rubble; b face; c – road; d 3 m further south
B2	SP5731	42°48.742	1°48.188	Fraisse	a: shaded area of recent working; b from an exposed area
B3	SP5732	42°48.681	1°48.181	Alarcon	a: new face; b: dark material
B4	SP5733	42°48.645	1°48.184	Alarcon	Chlorite rich
B5	SP5734	42°48.602	1°48.151	Praderes	A : sub-pegmatic; b soft talc
B6	SP5735	42°48.574	1°48.128	Praderes	Base of 4 m high face
B7	SP5736	42°48.561	1°48.076	Blazy	No comment recorded
B8	SP5737	42°48.544	1°48.031	Blazy	From recently excavated pile
B9	SP5738	42°48.507	1°48.067	Carrierre	No comment recorded
B10	SP5739	42°48.432	1°48.033	Carrierre	From a 3-4 m smooth face
B11	SP5740	42°48.43	1°48.2		Dolomite sample
B12	SP5741	42°48.889	1°48.071		Gneiss Sample

Table 3.2 Details of Samples obtained from the Trimouns talc quarry in October 2002

Gamma dose rates recorded at Trimouns varied between 0.25 and 0.75 mGy/a. At present no further assessment of the composition of the natural radioelement assemblage has been attempted, but it is perhaps worth noting that given a typical crustal U,Th ratio and modest K content these gamma dose rates would be accompanied by alpha and beta components giving overall dose rates of some 1-10 mGy/a within the talc matrix, depending on how the alpha dose distributed relative to luminescence sensitivity.

Samples from Luzenac were packed and shipped by courier to SUERC. Two small subsamples of samples A1 and B1 were also shipped back by post to provide an independent control for potential x-ray inspection during shipping.

3.3 Samples from Val Chisone

Having collected quarry and process samples from the Luzenac works, additional material was also collected from Val Chisone, again assisted by personnel from Luzenac talc and Lichtwer Pharma.

3.3.1   Samples from the Rodoretto Mine , Val Chisone

The Luzenac "Pharma" product is currently produced using material delivered from the Pyrenees by truck, approximately 4 times per week. However locally mined product is also subject to the thermal decontamination process at Val-Chisone, and has been used for pharmaceutical grade product in the past. Lichtwer-Pharma had reported higher low-temperature TL signals from the Italian product than from the French material in their internal investigation. For this reason, and with the aim of obtaining authentic geological samples which had not been exposed to significant levels of daylight, samples were collected from the Rodoretto mine at the head of the Val Chisone valley. The Rodoretto mine took over from two earlier extraction facilities at the beginning of 2002; however it is believed to represent a folded extension of the same formations as the mines which had previously been exploited. Should the need arise, the mine used up until the end of 2001 has been turned into a museum, and it may be possible to obtain supplementary samples if needed.

The trip into the mine was facilitated by the mine manager S. Milani, and his colleagues, geologist F. Montecelli, and the shift foreman S. Pascal. Access was via a 2.5 km drift tunnel, leading to two working areas; the southern and northern extraction area .The deposit is formed between mica-schist with Gneiss, and a hanging wall of mica-schist. It is approximately 5 m. high, and contains marbles and granite fragments in places. The presumed dolomite source that preceded talc formation has been completely absorbed into the talc. Small samples were obtained from both southern and northern areas representing (i) light talc, (ii) grey talc, and (iii) impurity mass. They were extracted directly from the two working faces in operation in October 2002, and can therefore be relied on to indicate the natural geologically induced TL from samples which have not been previously exposed to daylight, heat, UV or other processes known to transform TL signals. These samples are described below in Table 3.3 The gamma dose rate recorded in the northern area for 300s, was 0.63 mGy/a – of similar magnitude to observations made at the Trimouns quarry.

Sample	SUERC Reference	Details
C1	SP5742	Southern Area (elevation 992 m). Light talc.
C2	SP5743	Southern Area (elevation 992 m). Darker talc
C3	SP5744	Southern Area (elevation 992 m). Impure material
C4	SP5745	Northern Area – elevation 996.5m. Light talc.
C5	SP5746	Northern Area – elevation 996.5m. Grey talc
C6	SP5747	Northern Area – elevation 996.5m. Impure material

Table 3.3 Samples collected from the Rodoretto Mine

3.3.2   Samples collected from the Val-Chisone processing facility

The processing facilities at Val Chisone were visited in the afternoon of 9th October, and a further series of samples obtained. Luzenac OOO grade material is delivered to this facility by truck, 3-4 times per week, and used to produce the Luzenac "Pharma" product. Other talcs are also processed, some of which were also sampled. The "pharma" product is passed through a furnace at a minimum of 140°C for 20 minutes, for both drying and thermal decontamination. Apparently microbial loads up to 10^6 g^{-1} can be observed prior to thermal treatment. The material is fed within closed containers to the mill, and also from there to the packing line. Hourly quality control samples are drawn from various parts of the process, and monitored for reflectance. If the reflectance falls below 90%, the process is stopped and the milling is cleaned prior to re-starting.

Details of the samples collected at the Val Chisone works are summarised in table 3.4. Samples code D6 was not used.

Sample	SUERC Reference	Details
D1	SP5748	Chinese pink talc
D2	SP5749	Chinese green talc
D3	SP5750	Sardinian talc from Luzenac Sammatta source
D4	SP5751	Val Chisone talc after crushing
D5	SP5752	Luzenac OOO talc after crushing
D7	SP5754	Luzenac OOO after heat treatment
D8	SP5755	Luzenac OOO after milling
D9	SP5756	Packed sample of Luzenac Pharma from the packing line.

Table 3.4 Samples obtained from the Luzenac-Val Chisone Processing site October 2002

Samples from Val-Chisone and Rodoretto were transported back to the UK in checked-in luggage. Two mine samples (C2 and C3) were subsampled and also carried back in hand-luggage.

3.4 Additional Samples

3.4.1 Additional sources provided by Luzenac Talc

Two additional samples of talc from the Australian and North American sources operated by the Luzenac group were kindly forwarded to SUERC by the Toulouse Office of the company for possible use within the study.

Sample	SUERC Reference	Details
Australian Talc	SP5757	Sample of Australian talc sent by LuzenacEurope
USA Talc	SP5758	Sample of North American talc sent by LuzenacEurope

Table 3.5 Australian and North American Samples

3.4.2 Irradiated and Unirradiated Commercial Pharmaceutical Grade Talcs

It is common knowledge within the dietary supplement manufacturing industry that irradiated pharmaceutical grades of talc are available. Moreover enquiries with the Isotron irradiator confirmed that this material is routinely treated. A pair of samples was obtained from a UK supplier (Richard Baker Harrison) representing commercial material irradiated in hermetically sealed containers to produce material with an extremely high microbiological integrity. The material supplied was a Chinese talc with trade name Magsil Diamond 200. Both unirradiated retained samples and material which had been commercially irradiated at Isotron were supplied.

Sample	SUERC Reference	Details
Magsil Diamond 200 (Unirradiated)	SP5780	Control sample of Magsil Diamond 200 talc as received from supplier
Magsil Diamond 200 (Irradiated)	SP5781	Magsil Diamond 200 talc commercially irradiated in September 2002, 25-45 kGy.

Table 3.6 Commercial Irradiated talcs obtained for the study.

4. TL Analysis

4.1 Methods

Each sample was ground with a pestle and mortar, and approximately 1g of ground material subsampled. This was then washed with acetone, shaken and allowed to settle for 5s, allowing large agglomerated fragments to settle out. The solution containing finer talc particles was then decanted to another tube. Following resuspension, portions of the acetone/talc suspension were transferred to settling tubes containing clean 1cm diameter 0.25mm thick stainless steel discs, and the remaining acetone was evaporated off overnight in a 50°C oven. Duplicated aliquots were prepared for each sample from the Luzenac/Val Chisone fieldwork, and also of Magsil Diamond 200. These were used for a survey of TL signals and sensitivities described in section 4.2 below.

Two samples (one from Trimouns, the other from the Rodoretto mine, Val Chisone) were selected for more detailed examination. These were ground and split. One portion of each was irradiated at 5-10 kGy dose level at Isotron in November 2002. Both irradiated and unirradiated pairs were then dispensed as above in sets of 20 or more discs per sample to be used in storage tests.

TL analyses were conducted both using SUERC research TL equipment, with Electron Tubes 9883QB photomultipliers filtered with 3 mm Kopp 7/59 and 3mm of Schott KG1 filter glass,and with a Risø DA15 automatic TL reader. Samples were measured, irradiated with a $^{90}Sr/^{90}Y$ source (typically at 200 Gy dose) and re-measured. TL curves were recorded to 400°C, 450°C or 500°C at heating rates of 5°C s^{-1} in an atmosphere of oxygen-free nitrogen.

4.2 TL signals and sensitivities from all samples

The TL glow curves for all samples were recorded both on the SUERC PC1 TL reader and the Risoe DA 15 reader. Glow curve plots for the full series are shown in Appendix B; tabulated values of integrated data TL intensities and glow ratios (relative to the 200 Gy normalising dose) are presented for all samples in Appendix C.

In general the radiation induced response for the majority of samples can be described in terms of four main TL glow peaks, henceforth referred to as Peaks I, II, III and IV. The temperatures registered for these peaks differ slightly in raw data recorded on the two instruments as a result of differences in the apparent temperature calibration, and thermal lags of the two systems. Thus peak temperatures recorded by the Risoe instrument are some 15-20 degrees higher than the SUERC PC reader. Checks with LiF TLD-100 (as suggested by EN1788) produce peak V values of approximately 250°C for the SUERC instrument, and approximately 270 °C for the Risoe reader, in keeping with observations from interlaboratory trials. It is believed that the former are slightly closer to the reference value for this material than the latter. The apparent temperatures of peaks I to IV respectively are : Peak I 140-160°C (SUERC instrument); 160-180°C (Risoe); Peak II 190-210°C (SUERC); 220-240°C (Risoe); Peak III 260-280 (SUERC); 280-300°C (Risoe); Peak IV 320-340°C (SUERC); 360-380°C. The different optical filters on the instruments also affect absolute sensitivity.

Peak I - Glow 1 vs Glow 2 Plot

Figure 4.1 Glow 1 vs Glow 2 plot for Peak I in all samples

Figures 4.1 and 4.2 summarise the luminescence signals and sensitivities of the whole series of samples for the first two peaks, peaks I and II. Tables 4.1 to 4.3 give luminescence signal strengths for glows 1 and 2 plus glow ratios (G1/G2) relative to the 200 Gy normalising dose. The sensitivities of the individual peaks recorded in glow 2 (ie after irradiation) vary over some 3 orders of magnitude or more from sample to sample. There are also subtle changes in the relative proportions of the four peaks in talc samples from different origins, although the first three peaks appear to be highly correlated with each other. Within a single source area (eg the Trimouns

quarry) there are sensitivity variations of some 1-2 orders of magnitude from sample to sample. At present the origins of these variations have not be related to any specific mineralogical property; however it is perhaps worth noting that the variation in major mineralogy from sample to sample is not expected to cover such a wide range. The implication is that, in common with other mineral systems, luminescence sensitivity is responding to minor/trace phenomena within the sample – for example the concentrations of structural or impurity defects within crystal lattices. It is likely that these effects are characteristic of the formation processes and individual qualities of the materials. It is also noted that in some instances the TL glow curves could be considered to reflect a more continuous distribution of trap depths, which is more typical of other alumino-silicates.

Peak II - Glow 1 vs Glow 2 Plot

Figure 4.2 Glow 1 vs Glow 2 plot for Peak II in all samples

Sample		PI	PII	PIII	PIV
A1	SP5726	258	524	2851	1898
A2	SP5727	450	4229	31837	18924
A3	SP5728	402	4601	30817	14034
A4	SP5729	265	1154	1035	1521
B1a	SP5730a	940	20971	131494	105669
B1b	SP5730b	10184	75140	278180	366642
B1d	SP5730d	4085	84597	413296	230420
B2a	SP5731a	12878	217957	1097945	930616
B2b	SP5731b	14042	247816	1124951	801392
B3a	SP5732a	7180	83147	279832	230022
B3b	SP5732b	6057	216348	841912	667563
B4	SP5733	2991	19199	15379	33452
B5a	SP5734a	3116	40405	133797	175587
B5b	SP5734b	350	1044	2917	2305
B6	SP5735	297	265	401	361
B7	SP5736	7470	112862	607631	469442
B8	SP5737	7705	79370	431813	364988
B9	SP5738	1539	13080	69803	50099
B10	SP5739	1012	20357	108384	71002
B11	SP5740	583	4450	6907	9890
B12	SP5741	2205	84307	408487	374186
A1 posted	SP5726	294	1265	9776	6625
B1a posted	SP5730a	2719	34285	179789	182861
C1	SP5742	254	2541	64681	50266
C2	SP5743	372	5071	95277	13735
C3	SP5744	308	1520	47783	57542
C4	SP5745	334	4279	130147	49031
C5	SP5747	255	1822	32459	10873
C6	SP5748	250	936	23341	13673
C2 hand luggage	SP5743	321	1167	28190	8219
C3 hand luggage	SP5744	276	1182	29666	37705
D1	SP5748	1757	8666	9501	3378
D2	SP5749	489	2159	11099	2646
D3	SP5750	3166	19543	244013	57081
D4	SP5751	480	1782	24873	16312
D5	SP5752	5151	24787	87710	50986
D7	SP5754	254	2292	55387	17045
D8	SP5755	909	32080	956882	368531
D9	SP5756	670	12649	549647	267503
Magsil Control	SP5780	364	3049	14960	22166
Magsil 25-45 kGy	SP5781	20649	222280	293186	108231
B1a 5-10 kGy	SP5730	271970	1206249	1299811	556994
C2 5-10 kGy	SP5743	47237	184321	77949	5381

Table 4.1. Glow 1 intensities from initial screening (Risø DA15 automatic
TL reader)

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Sample		PI	PII	PIII	PIV
A1	SP5726	2566	2634	2744	1252
A2	SP5727	8437	15029	16384	6579
A3	SP5728	7021	11702	14611	5388
A4	SP5729	2524	4572	5827	1910
B1a	SP5730a	25054	49536	85006	25531
B1b	SP5730b	224758	427033	613042	222630
B1d	SP5730d	91398	192980	230365	74753
B2a	SP5731a	332170	615187	917114	292029
B2b	SP5731b	297839	597247	841169	253660
B3a	SP5732a	100354	191264	273195	108290
B3b	SP5732b	192085	367334	480615	223876
B4	SP5733	76801	141079	187466	74498
B5a	SP5734a	88121	178022	307281	113599
B5b	SP5734b	2356	3985	4847	1755
B6	SP5735	701	681	768	570
B7	SP5736	174755	356054	474658	146178
B8	SP5737	128921	261472	329657	106589
B9	SP5738	32619	54415	62184	25687
B10	SP5739	32401	54525	66743	27106
B11	SP5740	40068	52495	62147	24069
B12	SP5741	151407	184935	362199	212084
A1 posted	SP5726	4878	7024	10206	4348
B1a posted	SP5730a	87810	179645	274340	76040
C1	SP5742	64719	112586	153633	35006
C2	SP5743	39929	89861	70004	8718
C3	SP5744	88392	186296	244188	60945
C4	SP5745	91876	157583	227500	31037
C5	SP5747	14820	23905	27494	5148
C6	SP5748	18560	29654	43112	8608
C2 hand luggage	SP5743	26242	58686	51140	6943
C3 hand luggage	SP5744	51858	98063	116722	18237
D1	SP5748	9426	13579	14255	2103
D2	SP5749	2982	3322	7390	1578
D3	SP5750	34372	58660	152875	23470
D4	SP5751	20891	33805	47016	9749
D5	SP5752	23190	44264	59126	21401
D7	SP5754	12357	21786	49616	6638
D8	SP5755	258851	397569	886267	170146
D9	SP5756	266721	391915	608282	220074
Magsil Control	SP5780	13394	32644	38515	15165
Magsil 25-45 kGy	SP5781	16534	35823	50042	20583
B1a 5-10 kGy	SP5730	64981	134095	210051	58598
C2 5-10 kGy	SP5743	14212	29558	23925	3195

Table 4.2. Glow 2 intensities from initial screening (Risø DA15 automatic TL reader)

Sample		PI	PII	PIII	PIV
A1	SP5726	0.100546	0.198937	1.038994	1.515974
A2	SP5727	0.053336	0.281389	1.943176	2.876425
A3	SP5728	0.057257	0.393181	2.109164	2.604677
A4	SP5729	0.104992	0.252406	0.177621	0.796335
B1a	SP5730a	0.037519	0.423349	1.546879	4.138851
B1b	SP5730b	0.045311	0.175958	0.45377	1.646867
B1d	SP5730d	0.044695	0.438372	1.794092	3.082418
B2a	SP5731a	0.038769	0.354294	1.197174	3.186725
B2b	SP5731b	0.047146	0.414931	1.337366	3.159316
B3a	SP5732a	0.071547	0.434724	1.024294	2.12413
B3b	SP5732b	0.031533	0.588968	1.751739	2.981843
B4	SP5733	0.038945	0.136087	0.082036	0.449032
B5a	SP5734a	0.03536	0.226966	0.435422	1.545674
B5b	SP5734b	0.148557	0.261982	0.601816	1.31339
B6	SP5735	0.42368	0.389134	0.522135	0.633333
B7	SP5736	0.042746	0.31698	1.280145	3.211441
B8	SP5737	0.059765	0.303551	1.309886	3.424256
B9	SP5738	0.047181	0.240375	1.122523	1.950364
B10	SP5739	0.031234	0.373352	1.623901	2.61942
B11	SP5740	0.01455	0.08477	0.11114	0.410902
B12	SP5741	0.014563	0.455874	1.127797	1.764329
A1 posted	SP5726	0.060271	0.180097	0.957868	1.523689
B1a posted	SP5730a	0.030965	0.190849	0.655351	2.4048
C1	SP5742	0.003925	0.022569	0.42101	1.435925
C2	SP5743	0.009317	0.056432	1.361022	1.575476
C3	SP5744	0.003484	0.008159	0.195681	0.944163
C4	SP5745	0.003635	0.027154	0.572075	1.57976
C5	SP5747	0.017206	0.076218	1.180585	2.112082
C6	SP5748	0.01347	0.031564	0.541404	1.588406
C2 hand luggage	SP5743	0.012232	0.019885	0.551232	1.183782
C3 hand luggage	SP5744	0.005322	0.012053	0.254159	2.0675
D1	SP5748	0.186399	0.638191	0.666503	1.606277
D2	SP5749	0.163984	0.64991	1.501894	1.676806
D3	SP5750	0.09211	0.333157	1.59616	2.432084
D4	SP5751	0.022976	0.052714	0.529033	1.673197
D5	SP5752	0.222122	0.559981	1.483442	2.382412
D7	SP5754	0.020555	0.105205	1.116313	2.567792
D8	SP5755	0.003512	0.08069	1.079677	2.165969
D9	SP5756	0.002512	0.032275	0.903606	1.215514
Magsil Control	SP5780	0.027176	0.093402	0.38842	1.461655
Magsil 25-45 kGy	SP5781	1.248881	6.204952	5.858799	5.258271
B1a 5-10 kGy	SP5730	4.185377	8.995481	6.188073	9.505341
C2 5-10 kGy	SP5743	3.323741	6.235909	3.258056	1.684194

Table 4.3. Glow Ratios from initial screening (Risø DA15 automatic TL reader)

The TL glow shapes from individual talc sources are the consequence of the defect structure produced within the talc and impurity mineral lattices by substitional and intrinsic defects. Many of the individual talc sources sampled in this study appear to be visually distinguishable on the basis of the TL glow 2 shape, and there may be further potential for investigating the use of TL sensitivities as a tool for characterising the individual formations. The components of the radiation induced signals which are present in the first glow TL are themselves a response to the treatment condition and thermal and radiation history of each sample. Here the analysis of the data in terms of the four peak model is both convenient and instructive.

The form of the dose response for peaks I to IV is considered in the next section, and equivalent doses represented by the "natural" signals in the samples evaluated. From the glow 1 vs glow 2 plots it is immediately apparent that the relationship between initial and second reading is able to separate the three irradiated samples by 1-2 orders of magnitude despite the high sensitivity variations. For the unirradiated samples the initial contents of peaks I and II are very substantially lower than those induced by even the 200 Gy dose used for normalisation. For the mined samples there is essentially no significant signal in peaks I and II as recovered, making discrimination between irradiated and unirradiated samples of this material very easy. For the Luzenac source materials the first two peaks show low signal levels initially, albeit at rather variable levels in the quarried samples. After thermal processing the first two peaks are eroded by approximately 98-99% (Peak I) and by some 90% (peak II).

Condition	Sample	Peak I	Peak II	Peak III	Peak IV
Unirradiated	B1a	0.038	0.423	1.547	4.139
Irradiated	B1a 5-10 kGy	4.185	8.995	6.188	9.505
Irradiated/Unirradiated		111.554	21.248	4.000	2.297
Unirradiated	Magsil Control	0.027	0.093	0.388	1.462
Irradiated	Magsil 25-45 kGy	1.249	6.205	5.859	5.258
Irradiated/Unirradiated		45.955	66.433	15.084	3.597
Unirradiated	C2	0.009	0.056	1.361	1.575
Irradiated	C2 5-10 kGy	3.324	6.236	3.258	1.684
Irradiated/Unirradiated		356.757	110.504	2.394	1.069

Table 4.4 Signal to background ratios for irradiated and unirradiated samples

Table 4.4 shows the glow ratios for all three of the samples that were irradiated at commercial food processing or sterilisation doses, together with the ratio of the irradiated to unirradiated glow ratios for all four peaks. The results show very clearly the importance of signal stability and glow curve temperature in determining both natural signal levels and signal to background ratios following irradiation. Peak I and II, which are at relatively low glow curve temperatures show between 45 and 360 times more signal after irradiation than beforehand (peak I) or between 20 and 110 times more (peak II). By contrast the initial signals in Peaks III and IV are

systematically higher, leading to lower increments after irradiation. These observations can be very readily accommodated by conventional arguments about the thermal stability and saturation levels of these peaks. Thus lower temperature peaks lack sufficient thermal stability to accumulate large doses from the natural radiation environment, while the more thermally stable peaks at higher temperatures may also be close to dose saturation limits in some cases leaving little "headspace" for detection of extremely high doses. It may be significant that peak I in the Magsil sample, which had been irradiated in September 2002, some 1-2 months before the experiments were conducted, shows a lower signal to background ratio after irradiation than peak II – possibly implying that after 1-2 months there had already been significant signal losses in peak I.

4.3 Dose Response

Two samples were investigated for the form of the dose response curve from 100 Gy to 6.4 kGy. Irradiations were performed with a ^{90}Sr/^{90}Y beta source at a dose rate of 7 Gy min^{-1} on the Risø DA 15 reader. The samples of Luzenac (Trimouns) talc B1a, and the Rodoretto C2 sample were selected for this purpose, since they had also received additional attention through being irradiated and used for storage tests.

TL glow curves were recorded to 500°C at 5 °C s^{-1}. Figures 4.3 and 4.4 show the TL intensities of peaks I to IV for both samples respectively.
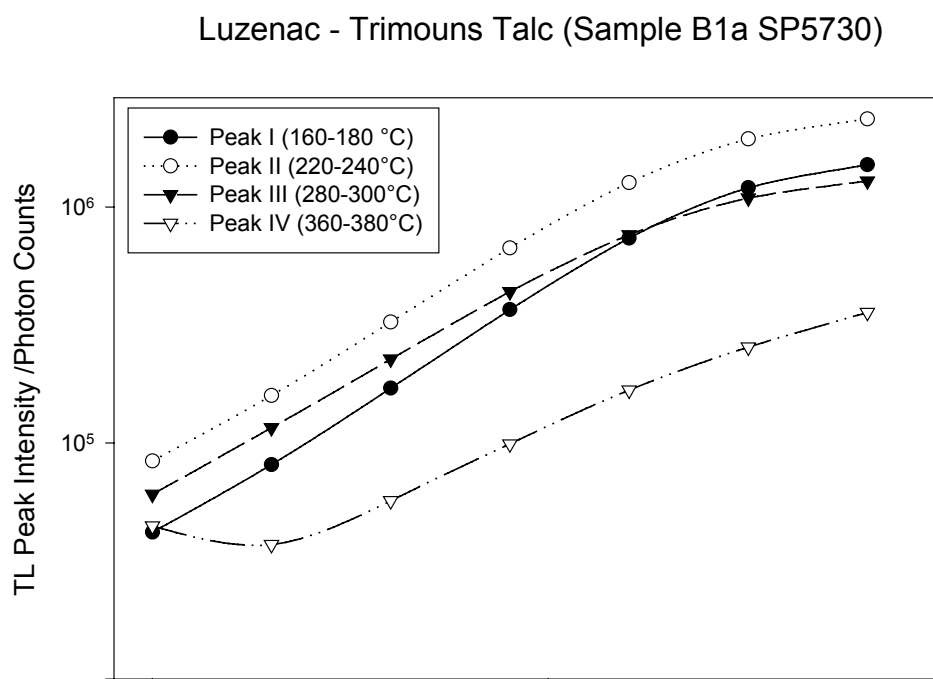

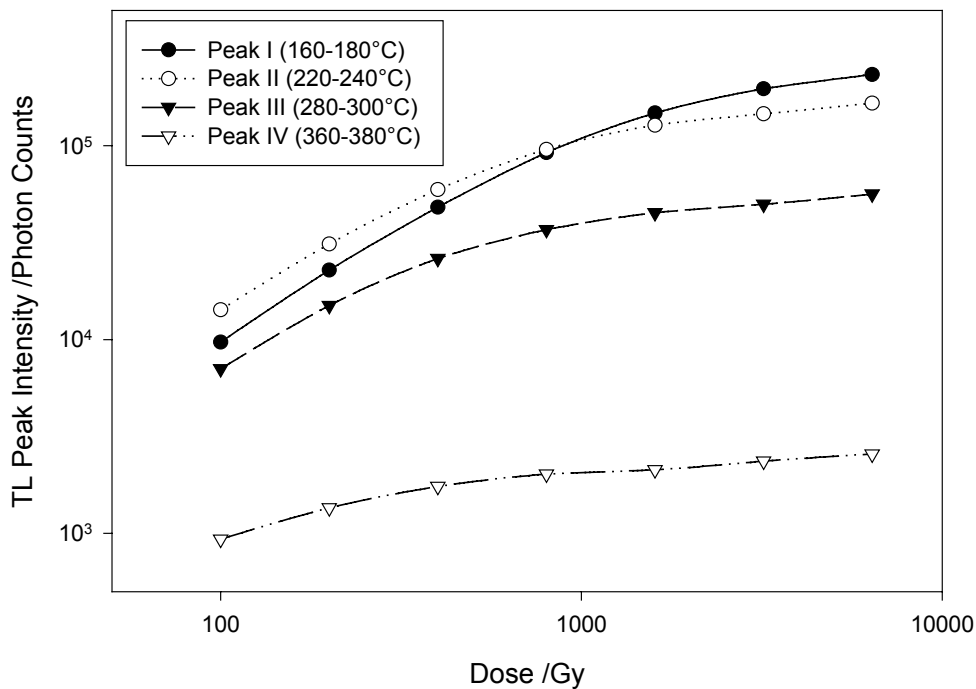Figure 4.3. Graph of TL peak intensities against dose, sample B1a.

Figure 4.4. Graph of TL peak intensities against dose, sample C2.

The dose response forms for all peaks, and for both samples show similarities and differences. In all cases there is evidence for partial saturation of the growth of TL with dose. The extent and dose level at which this occurs varies from peak to peak and samples to sample. For the Luzenac source sample peaks I and II show very similar forms of dose response with near parallel form on a log-log plot. The saturating component takes most effect in the kGy dose range, with some evidence of progressive growth at a lower gradient beyond this level. Peak III starts with a lower gradient and saturates at slightly lower doses, again with some continuing growth at higher doses. Peak IV by contrast shows a positive dose offset before the TL signal follows a similar dose response form. This may imply the existence of a competing process that suppresses expression of peak IV until a threshold dose is reached, or potential could be interpreted in terms of interactions between two trapping states which are linked together. This may merit further attention. The Val Chisone sample shows very conventional behaviour, with all four peaks showing simple saturating curves with different saturation doses and levels, progressing from 500 Gy up to the kGy level. Again there is some evidence for continued growth beyond the initial saturation stage. While forms of the dose response curves may not fit perfectly to single saturating exponential functions in all cases, implying the contribution of more than one trap, the data sets can be relatively well interpolated using such fits. It is therefore useful to examine the saturation coefficients for comparative purposes, and to assess the approximate doses represented by natural signals. The best fit parameters for curves of the form $y = a(1-e^{-bx})$ are shown in table 4.5

Sample	Peak	a	b	Mean saturation dose /Gy
B1 a	Peak I	$1.733 \pm 0.09 \times 10^6$	$3.407 \pm 0.36 \times 10^{-4}$	2940 ± 310
	Peak II	$2.587 \pm 0.084 \times 10^6$	$4.081 \pm 0.29 \times 10^{-4}$	2450 ± 174
	Peak III	$1.352 \pm 0.019 \times 10^6$	$5.035 \pm 0.17 \times 10^{-4}$	1990 ± 67
	Peak IV	$4.139 \pm 0.24 \times 10^5$	$2.585 \pm 0.34 \times 10^{-4}$	3870 ± 509
C2	Peak I	$2.37 \pm 0.04 \times 10^5$	$5.891 \pm 0.25 \times 10^{-4}$	1700 ± 72
	Peak II	$1.59 \pm 0.04 \times 10^5$	$1.097 \pm 0.087 \times 10^{-3}$	912 ± 72
	Peak III	$5.238 \pm 0.17 \times 10^4$	$1.553 \pm 0.16 \times 10^{-3}$	644 ± 66
	Peak IV	$2.286 \pm 0.1 \times 10^3$	$4.202 \pm 0.72 \times 10^{-3}$	238 ± 41

Table 4.5 Exponential fitting parameters for Peaks I to IV in samples B1a and C2

These parameters can be used to estimate the doses equivalent to the natural signals in samples B1a and C2. These are shown below in table 4.6 together with dose estimates based on linear extrapolation of the glow ratios determined relative to a 200 Gy normalising dose. It can be noted that the difference between linear and exponential models is relatively small when using the 200 Gy normalising dose. This would not however be so had the measured glow ratios been evaluated with a 1 kGy dose in keeping with normal EN1788 practice, since this would have put the measurements well into the non-linear response region.

Sample	Peak	G1/G2 ratio relative to 200 Gy dose	Natural Dose by exponential fitting /Gy	Natural Dose by linear extrapolation of 200 Gy dose / Gy
B1 a	Peak I	0.03752	7.26	7.50
	Peak II	0.42335	82.69	84.67
	Peak III	1.54688	318.54	309.38
	Peak IV	4.13885	904.75	827.77
C2	Peak I	0.00932	1.76	1.86
	Peak II	0.05643	10.19	11.29
	Peak III	1.36102	290.78	272.20
	Peak IV	1.57548	537.72	315.10

Table 4.6 Natural dose levels in peaks I to IV from samples B1a and C2

Clearly the differences between saturation levels in the two samples investigated imply that caution should be taken in extrapolating linear dose estimates to other samples; particularly for higher dose levels. At low dose, providing the saturation levels of the survey samples are not significantly lower than those from sample C2, the dose estimated may, nonetheless be useful. Table 4.7 gives linear dose estimates for the survey samples. It can be seen that for unirradiated samples almost all peak I estimates are below 10-20 Gy, and many below 5 Gy.

Sample	Reference	Peak I	Peak II	Peak III	Peak IV
A1	SP5726	20.11	39.79	207.80	303.19
A2	SP5727	10.67	56.28	388.64	575.29
A3	SP5728	11.45	78.64	421.83	520.94
A4	SP5729	21.00	50.48	35.52	159.27
B1a	SP5730a	7.50	84.67	309.38	827.77
B1b	SP5730b	9.06	35.19	90.75	329.37
B1d	SP5730d	8.94	87.67	358.82	616.48
B2a	SP5731a	7.75	70.86	239.43	637.35
B2b	SP5731b	9.43	82.99	267.47	631.86
B3a	SP5732a	14.31	86.94	204.86	424.83
B3b	SP5732b	6.31	117.79	350.35	596.37
B4	SP5733	7.79	27.22	16.41	89.81
B5a	SP5734a	7.07	45.39	87.08	309.13
B5b	SP5734b	29.71	52.40	120.36	262.68
B6	SP5735	84.74	77.83	104.43	126.67
B7	SP5736	8.55	63.40	256.03	642.29
B8	SP5737	11.95	60.71	261.98	684.85
B9	SP5738	9.44	48.08	224.50	390.07
B10	SP5739	6.25	74.67	324.78	523.88
B11	SP5740	2.91	16.95	22.23	82.18
B12	SP5741	2.91	91.17	225.56	352.87
A1 posted	SP5726	12.05	36.02	191.57	304.74
B1a posted	SP5730a	6.19	38.17	131.07	480.96
C1	SP5742	0.79	4.51	84.20	287.19
C2	SP5743	1.86	11.29	272.20	315.10
C3	SP5744	0.70	1.63	39.14	188.83
C4	SP5745	0.73	5.43	114.42	315.95
C5	SP5747	3.44	15.24	236.12	422.42
C6	SP5748	2.69	6.31	108.28	317.68
C2 hand luggage	SP5743	2.45	3.98	110.25	236.76
C3 hand luggage	SP5744	1.06	2.41	50.83	413.50
D1	SP5748	37.28	127.64	133.30	321.26
D2	SP5749	32.80	129.98	300.38	335.36
D3	SP5750	18.42	66.63	319.23	486.42
D4	SP5751	4.60	10.54	105.81	334.64
D5	SP5752	44.42	112.00	296.69	476.48
D7	SP5754	4.11	21.04	223.26	513.56
D8	SP5755	0.70	16.14	215.94	433.19
D9	SP5756	0.50	6.46	180.72	243.10
Magsil Control	SP5780	5.44	18.68	77.68	292.33
Magsil 25-45 kGy	SP5781	249.78	1240.99	1171.76	1051.65
B1a 5-10 kGy	SP5730	837.08	1799.10	1237.61	1901.07
C2 5-10 kGy	SP5743	664.75	1247.18	651.61	336.84

Table 4.7. Natural doses for all survey samples based on linear scaling to the initial screening results.

Some source materials give higher dose estimates in peak I (most notably sample B6 – whose low sensitivity is believed to play a part in this). It is notable that the mined samples and Luzenac Pharma sources following heat treatment all show stored doses below 5 Gy in Peak I. For peak II natural dose estimates of some 50-100 Gy represent the upper limits; again with mined samples and the processed "Pharma" grade materials giving lower values.

It terms of stability requirements it is notable that peak I would require durations in excess of 10^2-10^3 years at the dose rates indicated in section 3 to acquire naturally induced luminescence, and peak II would require some 10^{3}-10^4 years or more. The subsequent sections consider how such timescales compare with the stabilities expected for these signals.

4.4 Storage Tests

A series of storage experiments was conducted using two samples :- Luzenac (Trimouns) sample B1a was selected to represent this source, Val-Chisone (Rodoretto) sample C2 was selected for the Italian source. At start of experiment ground mass was split and one portion sent to Isotron for Irradiation at a 5-10 kGy dose. Acetone deposited TL samples were prepared using the method described in section 4.1. Both irradiated samples and unirradiated controls were stored together under the following conditions : dark storage and ambient temperatures; dark storage at 50°C; and storage in an artificial daylight light box (ref- Sanderson et al 1993) with approximately 7 mWcm^{-2} total fluence. Thus six combinations of treatment and storage were investigated for two samples. Sub samples were removed from storage in duplicates for TL measurement, both on the Riso DA15 system and the SURRC PC1 reader at fixed intervals after start of experiment (the time of irradiation). Measurements were made after delays of 2, 3,8,16 and 34 days. The data sets are tabulated in Appendix D and summarised here.

Data have been examined both in 50°C bands, and as 20 °C integrals broadly centred on the four peaks described above. The Val-Chisone sample, as previously noted, shows signals in the four peak temperature bands, but in this case the peaks are less well resolved, and peak IV in particular is rather insensitive. Glow ratios relative to the 200Gy normalising dose have been evaluated and are discussed further here.

Figure 4.5 to 4.8 show the results for peaks I&II and peaks III and IV respectively for the Luzenac sample. Similar plots for the Val-Chisone sample are shown in figure 4.9 to 4.12.

It is immediately apparent from the dark-stored irradiated samples that all four peaks are thermally stable over the period of these storage tests at ambient temperatures, and that at 50°C there are some losses in peak I particularly, but still sufficient signal retention to permit discrimination between irradiated and unirradiated samples for more than the duration of this experiment. The unirradiated controls show progressively higher initial signals with elevated glow curve temperatures but again it is notable that for dark-stored samples the discrimination between irradiated and unirradiated samples is possible using all four peaks. However the increasing residual geological signals for peaks III and IV reduces the separation from some 2-3 orders of magnitude (cf peak I) to approximately a single order of magnitude.

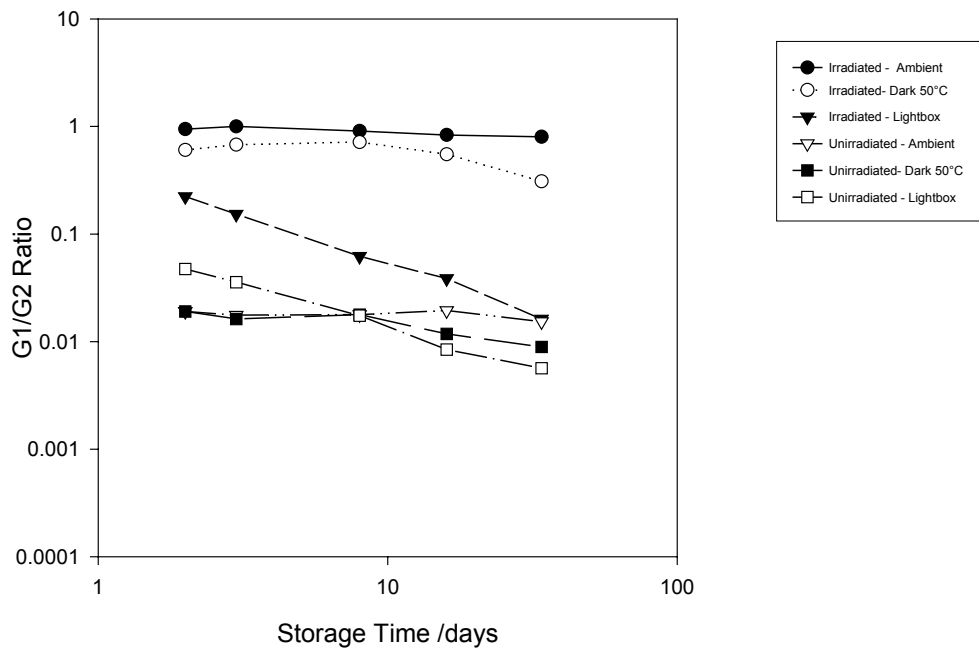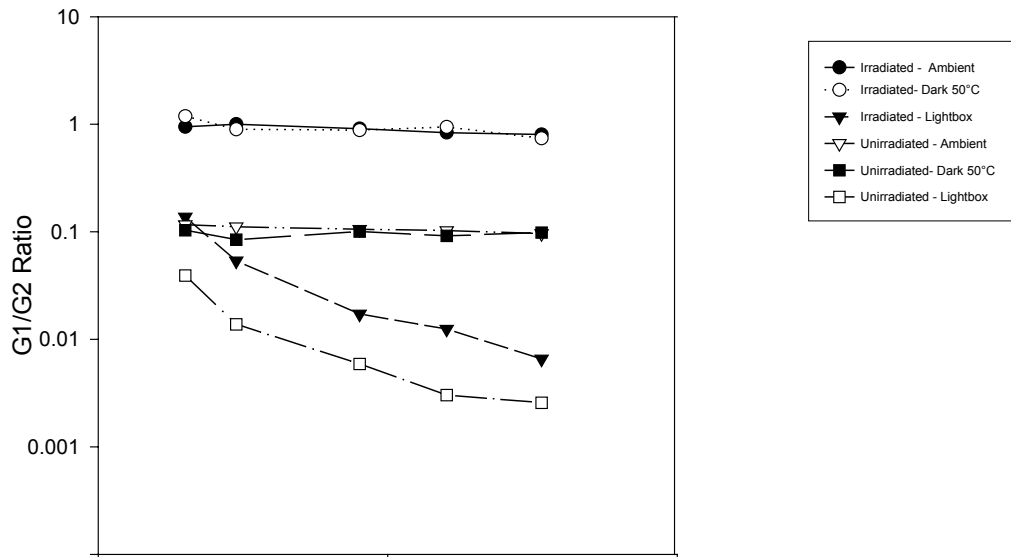21

Luzenac Sample B1a - Peak I

Figure 4.5. Graph of glow ratio against storage time, sample B1a, peak I

Luzenac Sample B1a - Peak II

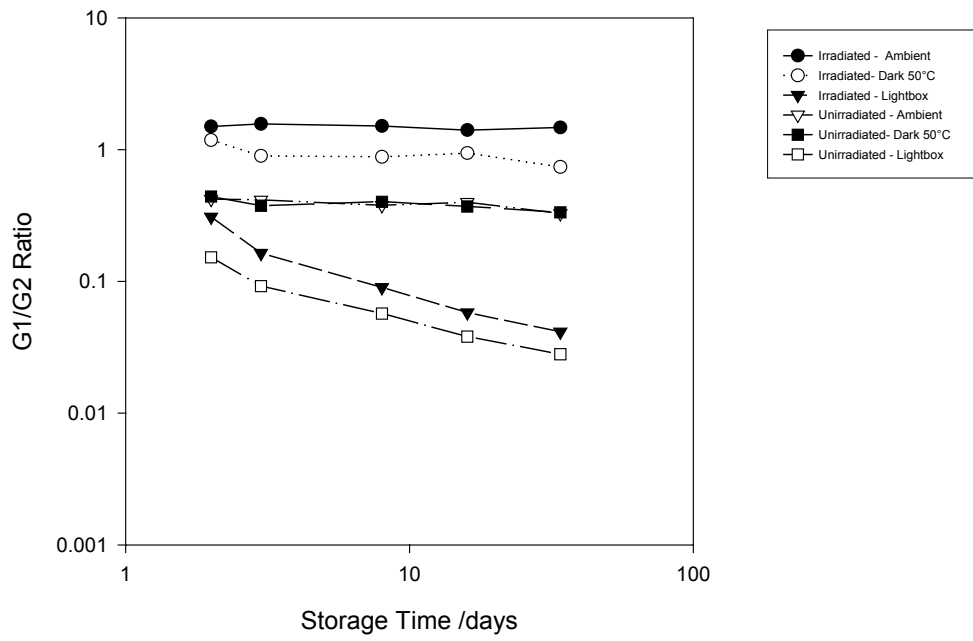## Luzenac Sample B1a - Peak III

Figure 4.7. Graph of glow ratio against storage time, sample B1a, peak III
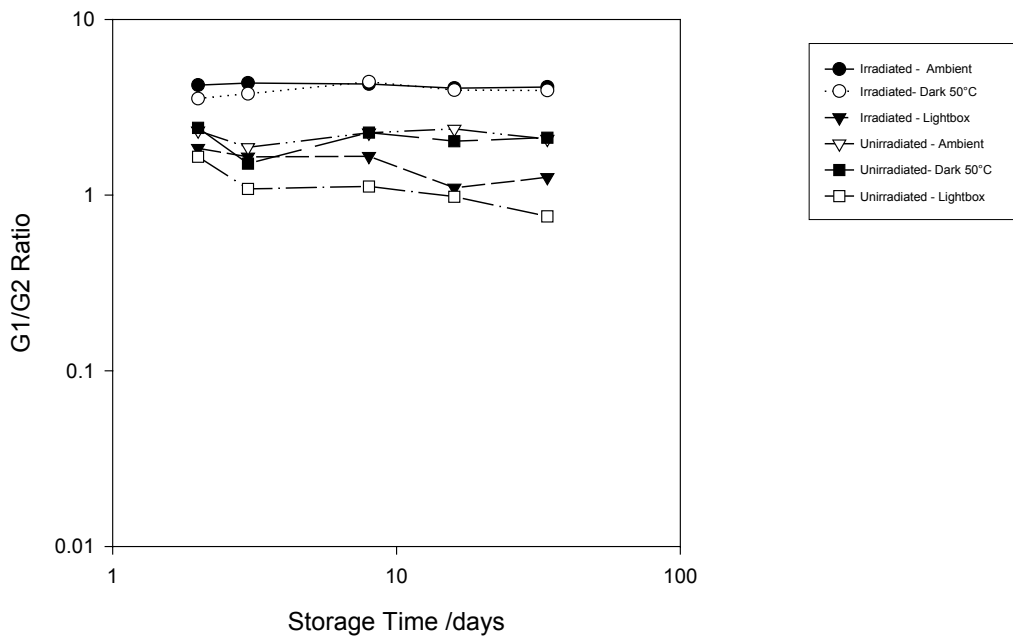
Luzenac Sample B1a - Peak IV

Figure 4.8. Graph of glow ratio against storage time, sample B1a, peak IV
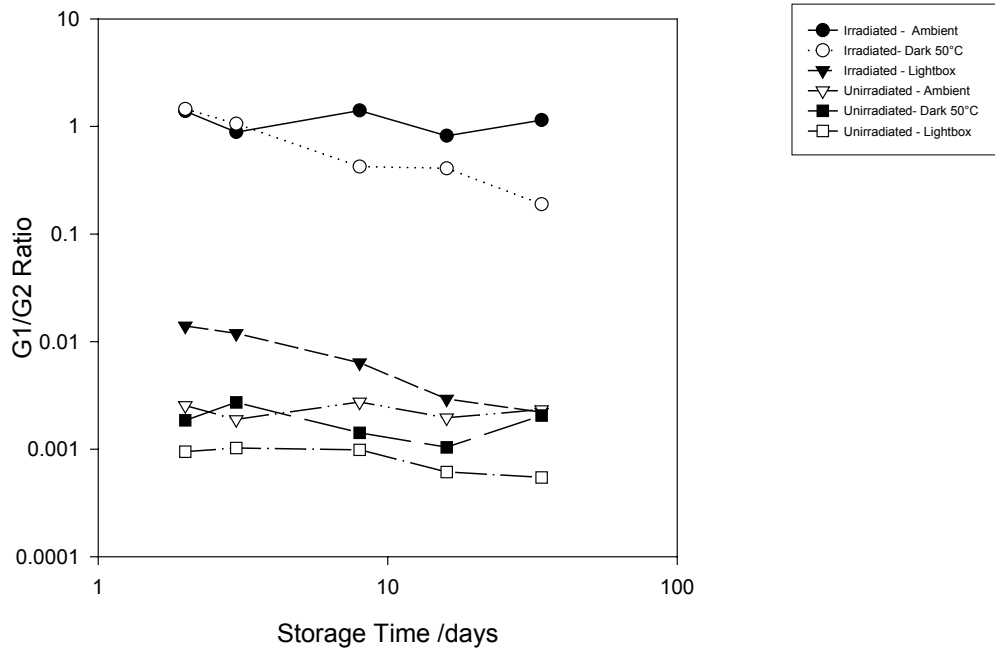
Val Chisone SampleC2 - Peak I

Figure 4.9. Graph of glow ratio against storage time, sample C2, peak I
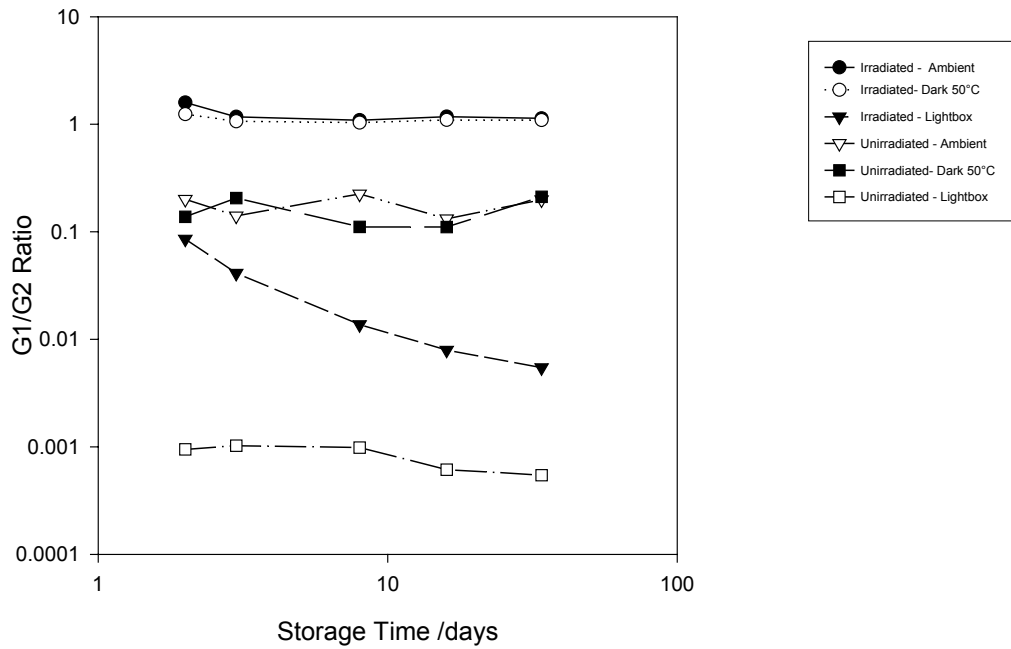
Val Chisone SampleC2 - Peak II

Figure 4.10. Graph of glow ratio against storage time, sample C2, peak II

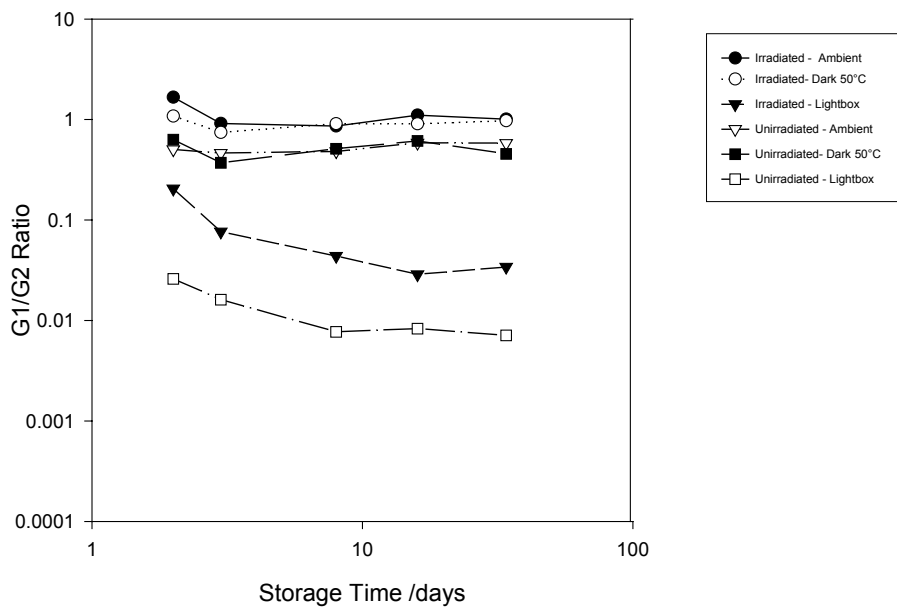## Val Chisone SampleC2 - Peak III

Figure 4.11. Graph of glow ratio against storage time, sample C2, peak III
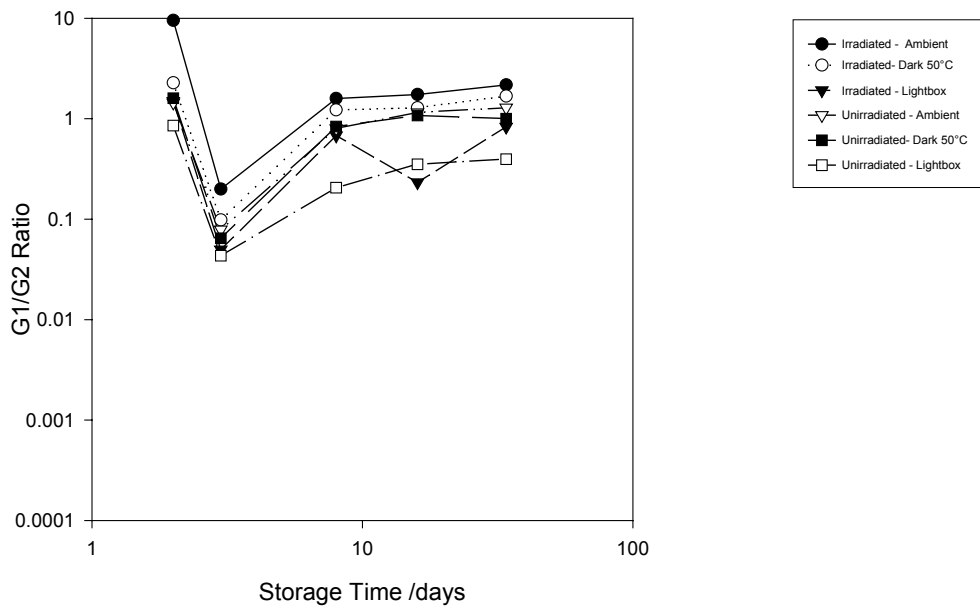
Val Chisone SampleC2 - Peak IV

Figure 4.12. Graph of glow ratio against storage time, sample C2, peak IV

Light exposure has a profound bleaching effect on the luminescence signals, most markedly on the lower temperature peaks (I and II) which are extremely sensitive to bleaching. By the time of the first measurement point the bleached samples have already lost the first two orders of magnitude of their initial radiation induced signals. Prolonged light exposure also reduces the levels of luminescence signals in the unirradiated samples, however a couple of important observations emerge from the two samples. For the Trimouns sample, which has been exposed to light in the natural environment, the implied amount of bleaching within the first day of the experiment differs between the natural background signal and the signal induced by artificial irradiation. The latter is reduced by nearly two order of magnitude, whereas the former starts out intriguingly with slightly higher levels of luminescence in peak I than it had prior to exposure the the light box. Thereafter both samples are bleached at a remarkably similar rate. This behaviour contrasts markedly with that of the Val-Chisone mined sample which had not been previously exposed to light. In this case both natural and artificial luminescence signals experience very similar proportions of post-illlumination signal reduction. It is not clear why the Trimouns sample shows an initial increase in TL following the first 24 hours or the experiment. The implication is that the may be partially influenced by prior optical exposure history of this material, in contrast with the mined sample. However the subsequent observation that the low temperature peaks are especially sensitive to optical bleaching, and therefore that prolonged light exposure would, if anything shift the natural glow curve shapes to higher temperatures, does not readily offer and explanation for the very small signals present in peaks I and I in some of the natural Trimouns samples.

Other than this observation the experiments seem to confirm that the thermal stability of talcs increases with increasing TL glow temperatures, in a manner which follows simple conventional behaviour.

4.5 Signal Stability

Signal stability is clearly relevant both to the use of TL for detection of irradiation, and also to help to interpret the origins of the residual natural signals in terms of geological irradiation at natural dose rates. Clearly the results of the work reported so far place constraints on the stabilities of the underlying TL trapping systems responsible for the main signals detected. From the survey evidence and storage tests it appears that peaks I and II could be considered as metastable over periods of interest to food irradiation, with peaks III and IV effectively stable at ambient temperatures. Light exposure is the main loss mechanism for irradiated samples and this clearly effects peaks I and II to a great extent. There is evidence in TL glow peak shape, and also in the storage test and dose response data to suggest that the peaks are not simply the product of single valued discrete traps, but that more than one component may be present within a given temperature range. It is nonetheless useful to conduct relatively simple analyses to ascertain the kinetic parameters associated with the majority signals. There are three potential indicators available here. Firstly the 50°C storage test and response to the Val Chisone 140°C 20 minute thermal treatment had measurable effects on luminescence in peaks I and II. It is possible to obtain first order estimates of kinetic parameters from these data. Secondly exploratory measurements of isothermal decays rates at elevated temperatures were conducted using samples B1 and C2 in an attempt to specify parameters for all four main peaks. Thirdly some exploratory progressive heat-stop measurements were made

from these two samples whereby each peak was glowed out sequentially, permitting application of the initial rise method for activation energy determination. The results of these approaches are summarised here. The methods used follow standard luminescence kinetic approaches (eg Chen & Kirsh, 1981).

4.5.1 Storage tests and thermal processing.

For a single trap of thermal activation energy E (eV) and frequency factor s (s^{-1}), the mean-life of trapped charge carriers is given by $\tau = s^{-1}\exp(E/kT_s)$ where k is Boltzmanns constant (8.617x10^{-5} eV K^{-1}),and T_s is the storage temperature in K. Providing relaxation processes, including luminescence production, are the most likely outcome of de-trapping, both charge carrier concentrations in a trap, and the remaining TL signal would follow simple exponential decay with mean-life τ. Thus knowledge of the values of E and s for traps associated with each TL peak can be used to estimate thermal stability at a range of storage temperatures.

From the empirical observations for samples C2 and B1a at 50°C it can be seen that there was an initial post-irradiation relaxation of peak I over the first 2-3 days, followed by a smoother signal loss. The proportions of signal remaining between 3 and 34 days (ie 31 day time interval) are tabulated below in table 4.8. Also shown are the measured signal survival in the Luzenac Pharma production following the first stages of thermal processing at Val Chisone . These were obtained by comparing samples D5 and D7. It was noted that samples D8 and D9 also showed continuing thermal erosion during the milling and packing stages. The product clearly continues to experience elevated temperatures during these processes, however their duration and magnitude are less well constrained than the thermal treatment at 140°C for 20 minutes.

Sample	Temperature /K	Duration /s	Peak I	Peak II	Peak III	Peak IV
C2	323	2678400	0.179	1.027	1.302	17.078
B1a	323	2678400	0.456	0.825	0.863	1.044
D5/D7	413	1200	0.093	0.188	0.753	1.078

Table 4.8 Surviving signal fractions over known thermal exposures.

For each storage temperature T_s, τ can be obtained as $\tau = -t/\ln(I/I_o)$, where I/I_0 is the surviving signal fraction, and t the duration. For two storage temperatures T_1 and T_2, with corresponding mean lives τ_1, τ_2 the thermal activation energy can then be obtained from $E = k.\ln(\tau_2/\tau_1)/(1/T_1 - 1/T_2)$. By substitution of E into the mean life expression given above, the value of s can then be obtained. Taking the B1a sample as potentially representative of Luzenac talc, and the D5/D7 pair as related to this these expressions can be used to evaluate E and s estimates for peaks I, and II. Peak III shows somewhat unstable estimates due to the close similarity between the mean life estimates for the two temperatures. Table 4.9 gives the estimated activation energies (E/eV), frequency factors (s/ s^{-1}) and mean life estimates at 0°C and 5°C for peaks I and II based on these observed values. It can be seen that thermal stability of some 10^2 years at ground temperatures is predicted for Peak I and of some 10^3 years for Peak II on this basis.

	T_{323} /s	τ_{413} /s	E /eV	s /s^{-1}	τ_{273} /years	τ_{278} /years
Peak I	3413567	504	1.13	1.11×10^{11}	179	76
Peak II	13928322	718	1.26	3.41×10^{12}	1772	676

Table 4.9 Kinetic parameters and predicted thermal stabilities for peaks I and II based on storage test data and thermal processing data.

4.5.2 Isothermal decay experiments

Experiments were conducted for samples C2 and B1a to extend the range of parameter estimates using isothermal decay methods. The measurements were taken with the SUERC PC1 TL reader which is believed to have a more accurate temperature calibration than other available equipment. Samples were held at increasing temperatures from 40°C to 400 °C in 20 °C while recording phosphorescence. The decay curves were fitted with exponential curves to determine mean lives at each storage temperature. In practice the decay curves recorded were not well described by single exponential decays, implying that the signal contributions originated from more than one trap. Fitting double exponential curves produced reasonably good fits,yielding two mean life parameters for each temperature.

The next step was to plot $\ln(\tau)$ vs $1/kT$ which for single trapping states should yield a slope of $-E$ (where E is the thermal activation energy) and an intercept equal to ln (s). Unfortunately the outcome was more complex. As shown in figure 4.13 for the B1a sample, the individual states were not sufficiently well resolved to permit unambiguous determination of kinetic parameters. A similar outcome was observed for sample C2. It may well be possible by using a combination of careful pre-heating, finer temperature increments and longer analysis times to apply the isothermal phosphorescence decay method to this material, however it appears that this may not be a particularly effective approach for separating the main traps responsible for the TL peaks.
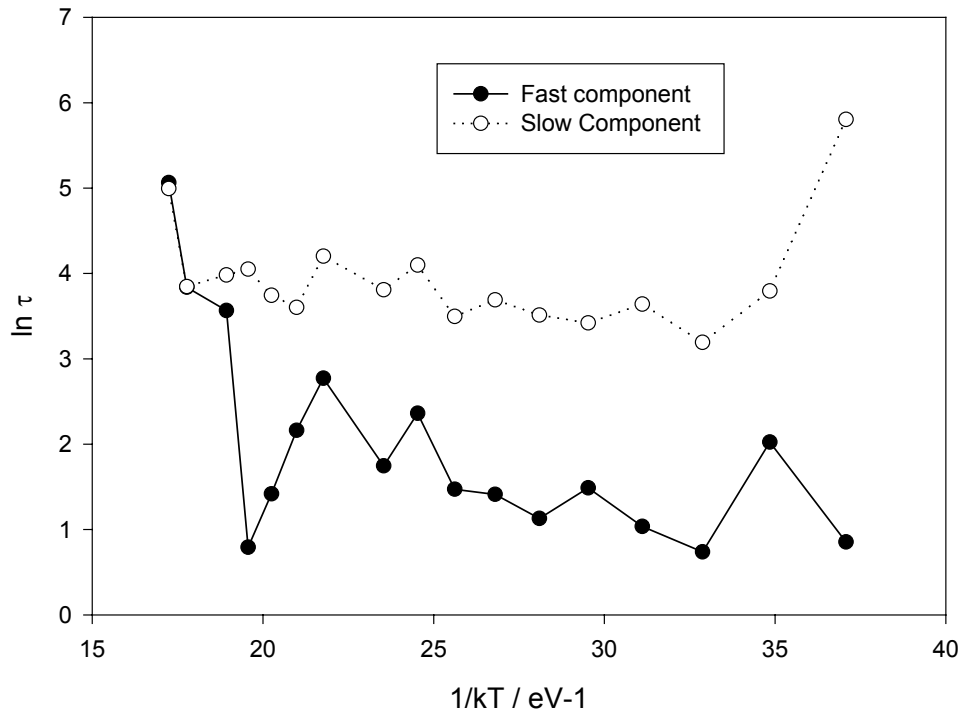
Isothermal decay results sample B1a

Figure 4.13 Isothermal decay results from sample B1a

4.5.3 Initial rise methods

The final approach investigated was sequential initial rise determination of the thermal activation energy, coupled to estimation of s from the observed peak maximum temperature. Once again both samples were investigated. A series of TL glows was recorded from each sample heating only up to the temperature of each peak, holding the sample at steady temperature for a short period (to anneal each component), and then cooling followed by re-heating and recording TL again until the next peak. This process was repeated sequentially throughout the glow curve for each sample. The underlying principle is that the initial part of a TL rise curve approximates Boltzmann dependence. By fitting an Arrhenius plot to the initial rise the thermal activation energy can be determined irrespective of the kinetic order of the process. This technique is extremely useful for dealing with materials with overlapping or interfering TL peaks. It was possible to obtain parameter estimates for both samples using this technique. The results for peaks I and II are reasonably consistent, both from sample to sample, and with the results obtained by storage tests and the thermal processing data. Peak I is the least stable peak in the system, followed by peak II, which by contrast has thermal lifetimes in the 10^4-10^5 year region. The higher temperature behaviour of the two samples differs in that whereas sampleB1a

shows sufficient stability to record geological doses, that of C2 appears somewhat limited, and potentially insufficient to account for the several hundred Gy doses inferred in section 4.3. The empirical observation that sample C2 can in fact deliver higher geological doses from its Peak IV temperature region despite the outcome of the kinetic analysis is itself an indicator that this temperature region has TL emission from more than one trapping system, offering a range of mean lives. More detailed kinetic analysis would be needed to produce a better model for the process.

Sample	Peak	E /eV	s /s^{-1}	τ_{273} /year
B1a	Peak I	1.18±0.007	2.49×10^{13}	3.3
	Peak II	1.49±0.006	7.06×10^{14}	144000
	Peak III	1.67±0.007	1.38×10^{15}	1.56×10^{8}
	Peak IV	1.62±0.012	3.76×10^{13}	6.8×10^{8}
C2	Peak I	1.05±0.007	9.02×10^{10}	10
	Peak II	1.23±0.009	2.08×10^{11}	7730
	Peak III	1.21±0.03	8.42×10^{9}	72200
	Peak IV	0.89±0.03	1.57×10^{6}	478

Table 4.10 Kinetic Parameters obtained using sequential initial rise methods.

It is arguable that the use of a simplified 4 peak model to discuss these talcs is an oversimplification of the underlying processes, and this is partly the reason why kinetic parameters have shown some ambiguities by different methods. However the combination of stability analysis and kinetic analysis does in general provide support for the observed distributions of higher natural doses in the higher glow curve temperatures. Peak II appears to have sufficient stability to account for the natural doses observed between 10 and 100 Gy. Peak I may well consist of several overlapping states, in which case the more stable subset is most probably able to store signals in the 1-10 Gy region observed. At present it is hard to judge whether those few samples showing initial stored doses above 10 Gy are exhibiting additional phenomenon, or merely represent the limiting cases for natural radiation effects.

5. Discussion and Conclusions

At the outset of the study, while there was sufficient data through the work conducted at SUERC and by Tela laboratories on behalf of Lichtwer Pharma to be aware that talc exhibited thermoluminescence, there were significant knowledge gaps in the nature and distribution of TL signals in talc and its associated materials. It was known that there were potentially source specific differences both in natural luminescence signals and in sensitivities. It was also known that talc is routinely irradiated in some countries. The effects on source materials used by Lichtwer Pharma of the various processes utilised by their suppliers were arguably less well characterised, as were the origins of signal detected in finished products.

The work conducted in this study has significantly added to our knowledge of the thermoluminescence of talcs. More than 40 new samples have been analysed systematically in this study, representing some of the major sources utilised for industrial, pharmaceutical and food related applications. Significantly the study included authentic raw materials recovered from primary extraction sites as well as processed materials following the selection and treatments used to produced thermally decontaminated pharmaceutical grade talc. Samples of commercially irradiated talc were also obtained, for comparison, and additional experimental irradiations were performed using a commercial facility. The data sets obtained from the broad suite of samples confirm that while TL sensitivity of individual talc samples covers some 3 or more orders of magnitude, there are broad similarities between the radiation induced TL responses of the majority of samples. The response can be broadly described in terms of four major peaks,although luminescence emission between the peaks is also observed, and there are other indicators of multi-trap contributions to the response within each broad peak. The irradiated samples have TL signals which are 1-2 orders of magnitude higher than any of the source materials studied in the lower two temperature peaks.

The dose response of two samples has been investigated in detail, and shows saturating behaviour with saturation coefficients giving mean dose values ranging from several hundred Gy to a few kGy. There are differences between saturation levels in each peak, and between the two samples. The non-linear dose responses can be used to quantify the dose equivalent to natural signals in all peaks. It can also be used to investigate the magnitude of the errors introduced by making a linear approximation to dose estimation. Providing a normalising dose below the saturation limits is used, it is possible to obtain first-order estimates of the stored doses in samples using simple re-generation and linear scaling. The 200 Gy normalising dose used in screening measurements from all samples was thus utilised to define the dose levels equivalent to the natural signals. The higher levels of stored dose observed in the high temperature peaks are consistent with the expectation that these peaks originate from more stable trapping systems. In the lower temperature regions the majority of samples tested showed very low natural signals in peaks I and II, inmost cases corresponding to doses in the $<<5$-10Gy, and 10-100 Gy region in most cases. These values, taken together with natural dose rates suggest stabilities in the 10^2-10^3 year region for peak I at ambient temperatures, and 10^4-10^5 year range for peak II.

Storage tests with both irradiated and unirradiated controls at ambient temperatures and 50°C, plus light exposure tests, have provided verification of the short term ambient stabilities of peaks I and II under dark conditions. There is some short term thermal erosion of peak I at 50°C under dark conditions, and to a very limited extent of peak II; the higher temperature signals are thermally stable. Prolonged exposure to artificial daylight evidently has a strong bleaching effect, particularly on the lower temperature peaks. Quarry samples and optically sorted samples from Luzenac that had received greater levels of light exposure than those taken straight from faces did not have higher levels of natural luminescence. It was also noted that the thermal processing used in Val Chisone to dry and decontaminate Luzenac talc for pharmaceutical production reduced the levels of residual TL by a further 1-2 order of magnitude in the lower temperature peaks.

Estimates of kinetic parameters from the Luzenac and Val Chisone materials have been undertaken by three methods with a view to estimating thermal stabilities on theoretical grounds. Both initial rise analyses and isothermal TL decay based on data from the storage tests and processing information produce roughly consistent parameter estimates . There is evidence for multi-trap contributions from isothermal phosphorescence measurements which implies a range of lifetimes within each peak. Therefore the estimated thermal stabilities can only be taken as a general guide. For peak II it is clear that there is sufficient thermal stability from these modelled states to account for the observed natural distributions. For peak I this is also arguable for the majority of samples, however it might be argued that for some of the natural samples either higher natural dose rates at micro-level may be encountered (eg due to heterogeneity of alpha and beta emitting nuclides in the natural sources), or there are more stable states available in some samples than implied by the kinetic analyses performed so far. The possibility of a mechanism for transferring charge carriers from deep to shallow traps (for example by a tunnelling like mechanism, or potentially by photo-transfer in the very first stages of light exposure following natural extraction) cannot be ruled out at this stage, but there is no firm evidence to point to these mechanisms, not a clear need to invoke them.

As far as the practical questions are concerned it is clear that irradiated talc can be detected by TL, providing the signals have not been fully bleached by sustained exposure to bright lights. Regarding the pharmaceutical grades of talc produced by Luzenac, the processed material sampled at the thermal processing plant does not exhibit TL signals that would interfere in any way with the application of EN1788 methods. The presence of small scale, low-dose equivalent, signals in some of the source materials which are removed by thermal processing is interesting, and should certainly be taken into account in interpreting TL data from EN1788 analyses. However providing the thermal processing is conducted correctly, and the materials are not subsequently mixed with talcs from other sources, there is no reason to expect that products made with these materials would lead to adverse analytical outcomes. It might be useful for manufacturers to verify the TL character of supplied talcs prior to utilisation, both for quality control purposes on the supply, and to ensure as a precautionary step that there are no processes incurring after distribution that could introduce additional signals.

For pure talc samples the data so far do not suggest that EN1788 would be adversely affected by natural signals in talcs, especially if they had been subject to

effective thermal decontamination as had been observed in this study. Clearly irradiation has a very readily distinguishable effect on this product, in both low and high temperature regions; whereas the natural geological signals are primarily concentrated in high temperature glow regions. In this respect talc does not differ qualitatively from other silicates encountered in food samples.

The situation with blended products is more challenging. EN1788 recognises that in compound foods the TL glow ratios will be diluted in blended products, but that glow curve shapes may still indicate the presence of irradiated components. In cases where low temperature signals are the minor TL peak, accompanied by much larger geological signals in high temperature zones, and low glow ratios are obtained in the 150-250°C region, there could in principle be ambiguity between the analytical signature of low level incorporation of irradiated material to a mixture, and the residual geological signal.

Research has been undertaken by FSA to define the limits of both TL and PSL methods under conditions of extreme dilution (Sanderson et al 1999). Work is also ongoing to explore new approaches to imaging luminescence to enhance capabilities for sorting out complex mixtures. In the mean time it would be prudent to exercise caution in the interpretation of TL data from products where talc may be present. If the glow 1 peak shape is dominated by high temperature TL, then supplementary analysis of the components of the product would be useful prior to reaching an analytical conclusion. On the other hand glow curves dominated by low temperature peaks can be taken as indicative of irradiation. In cases of doubt it may be helpful to accompany EN1788 analyses with mineralogical investigation and possibly further separation.

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Annex A. Implications of Talc for EN 1788 TL analyses

The implications of talc in food samples for standard EN1788 analyses have been outlined in the report. Here some of the potential scenarios are highlighted to assist with interpretation of EN1788 analyses.

- **Samples containing irradiated talc as a dominant source of TL signal**

As noted in the report irradiated talc can be detected using EN1788 procedures providing radiation induced signals have not been entirely bleached out prior to analysis.

Samples with the characteristic 4 peak talc signal in glow 2, and with high glow ratios in peaks II,III and IV, are likely to contain irradiated talc.

More work is needed on the practical limitations due to optical bleaching for detecting irradiated talc.

- **Samples containing thermally decontaminated pharmaceutical grades of talc**

Providing the thermal decontamination has been conducted properly these samples should not produce glow 1 signals that would lead to adverse EN1788 results.

- **Samples containing industrial grades of talc**

In some sources of talc there may be signals in peak II from untreated materials that fall within the EN1788 temperature band of 150-250°C. Peaks III and IV should in all cases be larger than peak II (with typical peak III/II ratios >5 and IV/II ratios >10) from such interfering signals. Glow ratios in peak II should be <<1.

Where samples produce EN1788 data identifying irradiated components based on the presence of signals in the 150-250°C band, the following additional checks are suggested:

- o Look for the characteristic 4 peak signal in G2. If it is present then the sample sensitivity may be dominated by talc
- o Look at G1 shapes. If the natural TL shows strong asymmetrical signals in the positions of peaks II, III and IV, and also has III/II ratios >5 and IV/II ratios >10 then the G1 signal may be due to natural TL from talc
- o In such cases consider additional analytical work (to determine the talc content and its size distribution), or to conducting additional separation work.
- o The presence of interfering signals from talc should be noted on the analytical report.
- o If TL data cannot be explained in terms of talc normal EN1788 criteria stand.