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Facets of Luminescence for Dating

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Facets of Luminescence for Dating

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ABSTRACT The aim of this article is to introduce luminescence dating and relate it to luminescence and Raman spectroscopy of minerals. The physical bases of luminescence signals used in dating and their relationships to other radiation-induced luminescence and Raman signals are briefly reviewed. The manner in which these signals are applied to evaluate luminescence ages is described. Archaeological and geological case studies from the author's experience are used to illustrate potentialities and issues related to different contexts, techniques, and materials.

KEYWORDS dating, dosimetry, OSL/PSL, TL/TSL

INTRODUCTION

Luminescence is a ubiquitous phenomenon. Chemiluminescence has been observed since antiquity in the form of bioluminescence. Although phosphorescence from minerals following exposure to light was reported earlier, thermally stimulated luminescence (thermoluminescence: TSL or TL) was probably first described in a scientific text when Robert Boyle reported the emission of light from a diamond that was slightly warmed, i.e., warmed too little to have produced incandescence.^[1] This distinction indicates the secondary nature of heating as a physical process in the production of the TSL signal: i.e. the need both for what may be termed a “primary irradiation” and a non-thermal emission process.

Non-thermal photon emissions, including luminescence^[2] and Raman emissions,^[3] may be differentiated according to precursor and emission processes and—to an extent—according to timescale, the effects of measurement temperature, and the relationship between irradiation and emission energy (Table 1). Names for different types of luminescence follow the nature and energy of the radiation used to induce the signal at the time of measurement (Table 2). Raman scattering and fluorescence may be considered intrinsic to a “centre,” if it is allowed that this can be composed of one atom and/or its immediate electronic environment created by a small number of strongly bound atoms, for example, in the unit cell of a crystal. Phosphorescence is produced by de-excitation following recombination.^[4,5,6] Recombination requires prior excitation to a dissociated state to have occurred. The state may be the conduction band or a metastable trap associated with a different centre. The timescale of phosphorescence emission depends on the lifetime of charge retention in the dissociated state: it may occur on a similar timescale to fluorescence, or gradually as a function

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TABLE 1 A Comparative Summary of Some Characteristics of Raman and Luminescence Signals

| Signal | Photon Emission | | | | | |
|------------------|---|---|--|--|---|--|
| | Process | Process | Timescale | Energy distribution | Energy region | Temperature dependence |
| Raman Scattering | Transfer of energy from photons to molecular electronic energy states | Vibrational relaxation | $\sim 10^{-12}$ s | Offset: difference between precursor energy and molecular ground state | Less than precursor energy (Stokes), unless molecule/atom already excited | - Energy (peak position). - Energy distribution (line broadening) - Intensity (quenching) |
| Fluorescence | Absorption of energy and promotion of electrons in atoms/molecules to excited states | De-excitation Delayed de-excitation (quantum state change) | $\sim 10^{-12}$ – 10^{-8} s $\sim 10^{-12}$ – 10^{-3} s | Permitted electronic transitions | | - Energy distribution (line broadening) - Intensity (quenching) |
| Phosphorescence | Absorption of energy and excitation of electrons of atoms/molecules to a dissociated state (ionization) | De-excitation following trapping and/or recombination | $\sim 10^{-12}$ s– ∞ | for de-excitation of centre | As above, or unrelated to precursor energy if recombination occurs in a centre other than the donor | - Decay rate (de-trapping) - Energy distribution (line broadening) - Intensity (decay rate/quenching/assistance) |

TABLE 2 Radiation-Related Luminescence Signals as a Function of Radiation Characteristics

| (Luminescence) Prefix-Primary Irradiation | Irradiation | | Detection Energy ^a |
|---|--|---|-------------------------------|
| | Type | Energy | |
| Photo- | Photon beam/radiation | Theory: any including ionizing. Practice: generally MidIR-UV: 10^{-2} – 10^2 eV | Mainly Stokes |
| Cathodo- | Low-energy accelerated electron | 10^2 – 10^3 eV | Any |
| Radio- | Ionizing radiation: X; γ ; β / high-energy accelerated electron; α | 10^3 – 10^7 eV | Any |
| Iono- | Ion beam: accelerated ion ranging in mass from proton to heavy charged particle | 10^3 – 10^7 eV | Any |
| Secondary Irradiation Thermo-/Thermally Stimulated- | Thermodynamic eviction of charge from traps filled during primary irradiation | Limits defined by coolant and incandescence: 77-1000 K = 10^{-3} – 10^{-1} eV | Any |
| Photon/Optically Stimulated- | Electromagnetic eviction of charge from traps filled during primary irradiation | Theory: any including ionizing. Practice: generally NIR-UV: 10^{-1} – 10^2 eV | Use Anti-Stokes |

^aGenerally MidIR-UV: 10^{-2} – 10^2 eV.

of the energy-depth and other characteristics of the trap, or the stability of the centre itself.^[7,8,9] Temperature independent phosphorescence indicates tunnelling between centres.^[10]

Types of luminescence measurements and materials applied directly to the evaluation of luminescence dates are radioluminescence from scintillator crystals (e.g., NaI gamma spectrometry and ZnO beta counting^[11,12]) and thermally/optically stimulated luminescence from dosimeter crystals^[6,13,14] and from found materials.^[15,16] The signals detected in each case are of phosphorescence. The key difference between scintillators and luminescence dosimeters is that the former are chosen or developed to avoid trapping of charges prior to recombination, while for dosimeters the trapping of a proportion of charges in sites with lifetimes sufficient to retain them during the period of deployment is necessary. Such sites are potential wells with energies in the crystal's band gap: disturbances in its electronic environment created by defects, impurities, vacancies or interstitial atoms.^[6,17,18] Since charge retention in these sites is thermally metastable and may be affected by quantum (tunnelling) processes, TSL and OSL are therefore forms of phosphorescence where the signals emerge so slowly that they require secondary irradiation (stimulation) to be produced at detectable rates.

Thermal or optical stimulation may promote charges from their traps to the conduction band or directly to other traps or to recombination centers. The luminescent recombination centres of minerals used for dating, from which signals are measured, are better understood than the trapping and non-luminescent recombination centres.^[17] In many cases, associations with particular defect structures or elemental impurities are established,^[18] but the precise effects of the concentration and coordination of particular ions/structures in controlling luminescence-dosimetric behaviour in the principal minerals used for luminescence dating remains open to further investigation.^[19,20] The process of ionization, trapping, stimulation and recombination may be understood in energetic and, to an extent, spatial terms using configurational coordinate diagrams (e.g., Reference 17, Fig. 2). Consideration of certain spatial effects and the mobility of charges, ions or defects has been applied to dating problems (e.g., References 9, 20, 21), but in general understanding

is based on energy level diagrams consisting of systems of hole and electron traps (e.g., References 4–10, 15, 17). These facilitate the modelling of all the steps from ionization to recombination, albeit only in terms of energy, and can hence be used to model the response characteristic of a particular signal to dose or activated predose.^[22–24] Chemiluminescence may also result if stimulation produces reactions in the crystal.^[25–27] If this reaction is reversible through primary irradiation and stimulation then the signals may be used for dosimetry or dating: many luminescence-dosimetric processes may be described as chemical changes rather than charge movements.^[28–30] As such, the thermally- and optically- stimulated luminescence signals used for dating are intimately related to the electronic environment/defect structure of the crystals they are measured from, albeit through a complex series of interrelated processes.

AGE EVALUATION

Mineral crystals separated from samples of found materials are measured to evaluate the dose of ionizing radiation absorbed by them, since they were last subjected to sufficient exposure to heat or light to remove charges from the relevant trapping sites (Fig. 1). The average rate of dose absorption by the crystals since this event, from the samples and incident on them from their surroundings, is evaluated based on series of separate measurements. The luminescence age is calculated as the quotient of these:

$$\text{Age (a)} = \text{Absorbed Dose (Gy)} / \text{Dose Rate (Gy/a)}$$

The dose rate is composed of alpha, beta, and gamma radiation, produced principally by the decays of ⁴⁰K and the ²³⁵U, ²³⁸U, and ²³²Th series, and cosmic radiation. It is estimated from dosimetric, radiometric, and geochemical methods in situ and/or in the laboratory,^[11–14,31–35] from calculations of the long-term average cosmic dose rate,^[36] and from measurements of water retention properties.^[37] The results are corrected for expected temporal variability based on the hydrological, archaeological, geomorphological, or geological context of the sample and combined based on the geometries of the measured volumes relative to the crystals measured by luminescence.^[38,39]

The “found materials” most commonly used for luminescence dating are pottery and sediments from archaeological contexts and young geological deposits.^[6,15] For pottery dating, the event of interest is heating during manufacture or use.^[37,40–44] For sediments it is their last exposure to sunlight before being (re)buried.^[16,45–50] However, a wide variety of other sample types and contexts are dated: anything containing inorganic crystals or microcrystalline inclusions that has suffered sufficient exposure to heat, light, mechanical action or crystallization during the event of interest may be attempted.^[6,15,51–57]

For dating, a defined mineral and grain-size fraction is generally prepared from a given sample by disaggregation, sieving, density, and/or magnetic separation and acid treatment.^[15] This permits the use of measurement protocols tailored to that material (e.g., thermal pre-treatments) and helps to constrain calculation of the environmental dose rate. The most often used fraction is sand-sized grains of quartz.^[41,46] Sand-sized grains of K or Na feldspar, and silt-sized grains of quartz or mixed silicates are also commonly measured.^[37,42,47,54] Less usual is dating with flint/quartzite, zircon, calcite and other minerals, which are less common in nature and/or exhibit significant signal instability and/or have complicated internal dosimetry due to the presence of alpha emitting radionuclides.^[10,55–58] Quartz and feldspar are not immune from such problems.

For dating measurements, mineral grains are generally presented as single layers on metal disks on a heater plate, with a large diameter PMT directly above and incident LED and/or laser sources.^[60–63] Thermal stimulation is by linear heating, often from room temperature to 500°C at 5°C s⁻¹. Optical stimulation is usually continuous wave (cw OSL), at either 470, 514, or 870 nm, with the sample held at elevated temperature to minimize re-trapping effects, but power may be ramped (lmOSL) or pulsed (pOSL).^[64] To exclude stimulating light and Stokes fluorescence from OSL measurements, and to minimize incandescence in TSL, the PMT is commonly filtered to detect bands in the region 300–450 nm. Other detection regions are used to analyze for particular materials or behaviours.^[8,17,20,44] NIR stimulation outside the spectral window of the PMT offers unconstrained detection, and generally produces little or no signal from quartz.^[65]

TSL and OSL signals are transient and are removed during their measurement (Fig. 1). Since different trap types have different thermal depths and optical cross sections, the signals from different traps appear at different temperatures or rates: de-trapping rates increase approximately exponentially with temperature, but approximately linearly with optical stimulation intensity. The TSL curve obtained by ramping the sample temperature consists of a series of peaks or a continuum (Fig. 3), depending on the trap distribution in the crystal.^[6–9,15,17,19,23,29,67] The cwOSL signal appears as a sum of approximately exponential decays (Fig. 3).^[16,22,67] Heating rates and optical power densities are chosen to facilitate analysis of different signals and components down to dark counts of c. 10 cps, while avoiding PMT saturation effects. Optical powers used for stimulation are of the order 0.1 W (10¹⁷ photons s⁻¹), requiring efficient extinction for cwOSL measurements.^[60] Integral photon counts above background in a given measurement vary by orders of magnitude depending on the signal used, sample size/sensitivity and dose.^[66]

The “as found” thermally or optically stimulated luminescence signal is calibrated relative to signals produced by controlled irradiations, typically from a ⁹⁰Sr/⁹⁰Y plaque (beta), to estimate the “as found” absorbed dose, which is often called “equivalent dose,” “palaeodose,” “stored dose” or “burial dose.” The calibration curve may be constructed in an “additive” and/or “regenerative” manner and may relate to the filling of traps or recombination centers.^[6,15,37–57] In additive measurements subsamples are given different doses in addition to their “as found” state, and signal versus dose is extrapolated back to obtain the absorbed dose.^[37,40,41,43,44,47,49,53] In regenerative measurements the “as found” signals are measured and interpolated through the response to subsequently administered calibration doses.^[42,43,45,48,49] Filling of traps is calibrated using thermal- and optical- stimulation of charge from those traps.^[37,45–55] Filling of recombination centres is calibrated using changes in the sensitivity of the sample as a function of previously administered and thermally activated dose.^[24,40,43,44,57]

The maximum age it is possible to date is constrained by saturation, with charge, of the traps that are the source of the particular luminescence signal being measured,^[40,57,66] the stability of this

signal,^[8–10,21] and the rate at which dose has been absorbed from the sample's environment.^[57,67] The minimum age is constrained by the need to obtain sufficient signal, that is, by the sensitivity of the signal to dose.^[43,48] Luminescence dating is commonly applied in the range 0–100 ka, with precision of at best $c. \pm 4\%$ at 1σ . Particular combinations of luminescence behaviour and/or environmental dose rate permit samples up to $c. 1$ Ma to be dated, and research continues to extend this range: certain types of luminescence signal have been observed to increase with doses into the multi-kGy range and over geological timescales.^[57,66]

CASE STUDIES

Luminescence Profiling and Dating of Sediments From Palaeolithic Archaeological Sites^[49,69,70]

Palaeolithic archaeological sites north and east of the Black Sea were examined as part of a project investigating whether present chronological data are biasing perceptions of Neanderthal climatic adaptation. The overall project objectives were to improve chronological constraints using tephtras, AMS ^{14}C , pollen stratigraphy, and luminescence dating of flint artefacts and sediments. Sediments are ubiquitous in these sites, but the materials and processes contributing to their accumulation are varied and complex. Since luminescence-dating results are intimately linked to sedimentary formation mechanisms, these were first investigated using "luminescence profiling".^[70] This is a rapid, semi-quantitative but stratigraphically detailed survey of the OSL and TSL sensitivity and absorbed dose of different grain size and mineral fractions prepared from many small samples taken through a sequence of deposits. Profiling of 15 sites was used to assess their data-ability and provide stratigraphic context for absolute-dating analyses. Absolute dating was conducted at 6 sites, selected on the basis of data-ability and archaeological importance. Absorbed doses were evaluated for both sand-sized quartz grains (200 μm , SAR OSL^[46]) and silt-sized grains of mixed silicates (10 μm , MAAD IRSL^[47]) for each of 35 samples. These fractions are affected differently by sedimentary processes and exhibit complementary differences in luminescence and

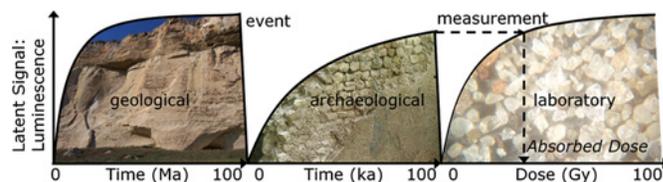


FIGURE 1 Dosimetric history of a mineral grain for luminescence dating. Adapted from Reference 68. (color figure available online.)

dosimetric behaviour. Residual natural luminescence signals, which produce age overestimates, were found to be less common in the silt fraction than the sand. Comparison of results from each fraction, in combination with stratigraphic information and the results of profiling, was used to obtain age estimates relevant to the archaeological and palaeoenvironmental history of each site (Fig. 2). These ranged from 98 to 26 ka (OIS5 to late OIS3): they indicated a mixture of single- and multi-period sites, and were relatable to independent dating evidence in terms of materials, methods and processes. In some cases the luminescence results indicated alternatives to extant chronological assumptions, including older chronologies for Neanderthal occupation than previously obtained using ^{14}C dating, thereby confirming the possibility of bias in the perception of Neanderthal climatic adaptation.

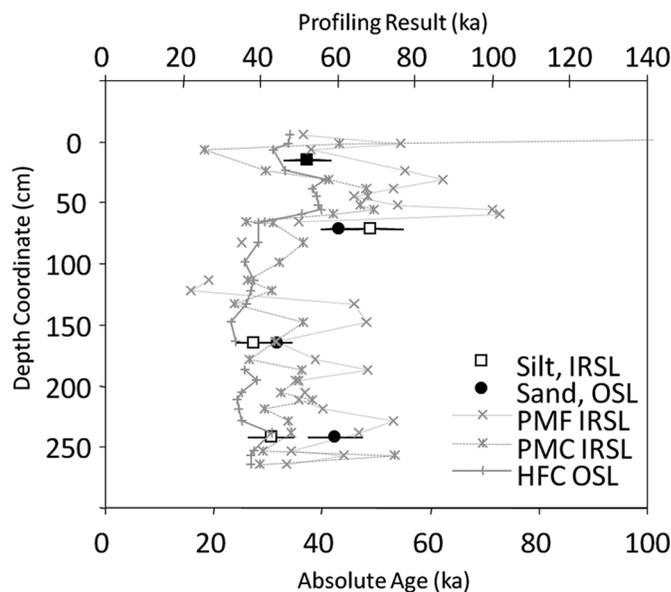


FIGURE 2 Comparison of absolute-dating results (Silt IRSL: polymineral 4–11 μm , Sand OSL: quartz 160–250 μm) and profiling data (PMF, PMC, HFC^[67]) through a sedimentary sequence exhibiting a phase of redeposition, above $c. 75$ cm, during which the luminescence signals were not reset. Adapted from Reference 49.

Pre-dose TSL Behaviour of High-Fired Ceramics^[43]

Compositional and luminescence studies of high-fired ceramics such as faience and porcelain have parallels in the study of thermal alterations in materials relevant to earth and planetary science. Broken faces of sherds were cored (0.06–1.3 g), to avoid further damage to the pieces, then analyzed by XRD, INAA, and luminescence dosimetry. In addition to quartz, faiences contained gehlenite and diopside, indicative of firing above $\sim 900^\circ\text{C}$. Porcelains contained mullite, indicative of firing above $\sim 1050^\circ\text{C}$.

The “110°C TSL peak” yielded the largest signal per unit dose, from the small quantities of material available. The pre-dose response characteristic of the recombination centres that produce this signal depends on thermal activation: the transfer of holes, produced during primary irradiation, into them from non-luminescent centres.^[24,40] Thermal activation and deactivation characteristics of a sample of porcelain and another of faience were studied, by irradiating quartz prepared from the samples with a relatively large dose (8 Gy), then rapidly heating this to different temperatures, annealing them at higher temperatures for longer periods, and finally repeating the irradiation and activation sequence (Fig. 3). Between each heating/annealing the samples were given a relatively small dose (0.8 Gy) and the 110°C TSL measured to monitor the sensitivity of the 110°C peak.

The two samples exhibited differences in activation characteristics and in the effect of annealing on reactivation characteristics, which may be related to their firing temperatures based on mineralogical analysis (Fig. 3). Absorbed doses were evaluated for these samples using a “simplified pre-dose additive-regenerative” protocol,^[43] adapted from Reference 44 to include regeneration of the dose response characteristic following deactivation at 600°C . The effects of annealing on dose response also differed, apparently as an indirect function of firing temperature.^[43]

The observations were difficult to explain in terms of charge transfer mechanisms alone: minimal changes in the electronic environment of the centres involved would help to explain them and might be tested using other luminescence techniques and/or Raman spectroscopy. It is hypothesized that the change observed in the porcelain could have

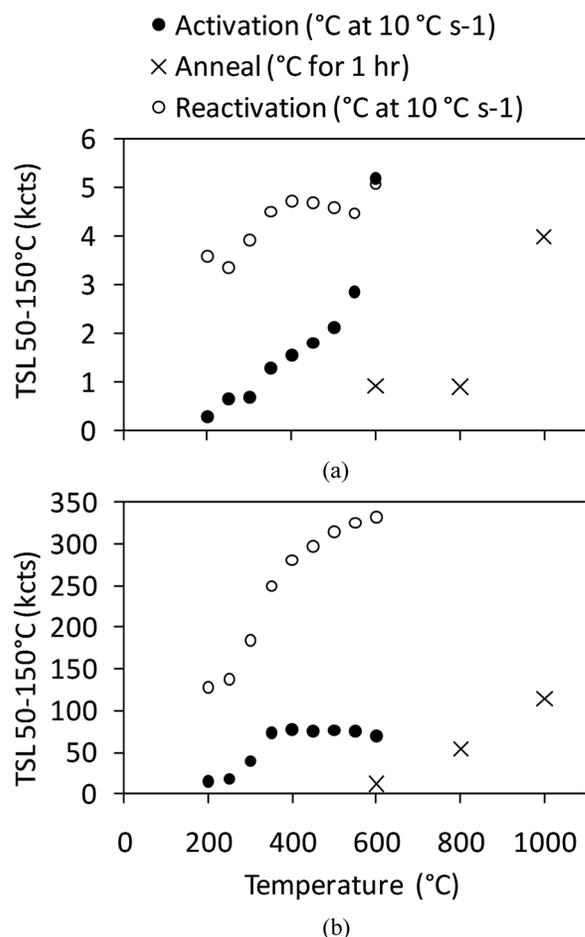


FIGURE 3 Thermal activation and deactivation characteristics of samples of a. porcelain and b. faience. a.: continuous activation with temperature; partial deactivation by annealing at 600°C ; reactivation to a plateau around the pre-anneal maximum. b.: activation to a plateau; deactivation by annealing; continuous reactivation to five times the pre-anneal maximum.

resulted from an increase in the proportion of luminescent-type hole centres^[24] able to emit in the wavelengths measured, with the total number of centres available remaining the same. That observed in the faience could be explained if additional luminescent-type hole centers had been brought into spatial association with the electron trap.^[20]

Effects of Internal Structure and Alpha Activity on OSL Signals in Quartz^[67]

OSL dating is being applied to understand the formation of the Mozambican coastal plain. Quartz from older dune sands has yielded natural signals higher than those attainable by continuous laboratory irradiation.^[50,71] The present study examined the

potential effects of impurities, inclusions and structural defects/voids, related to alpha radioactivity.

Quartz separates were analyzed by TSL and OSL (380 nm emission), INAA, SEM/XRF. The samples exhibited a continuum of broad TSL peaks that were sensitive to optical stimulation (Fig. 4): similar to zircon (inclusions), quartzite and flint (structural defects).^[55,56,58] Neutron activation analysis indicated high concentrations of Zr and its strong relationship with a group of mineralogically related elements. However, K, Th, and U were not related to Zr but exhibited moderate correspondence with Fe. SEM/XRF and optical examination indicated the presence of Fe in veins/cracks. Co-precipitation or co-infiltration of radionuclides with the Fe group would bring alpha emitters into spatial association with internal surfaces. Based on Fe concentration and inclusion size it was estimated that alpha radiation would affect little of the bulk crystal. Luminescence-dosimetric effects of alpha radiation in this context

were studied by comparing them with beta radiation, for compact crystalline quartz and milled quartz. The milled grains exhibited signal growth to much higher doses but probable instability, and higher alpha efficiency; particularly for a “medium” component in OSL (Fig. 4). These effects appear to be associated with higher defect concentrations produced by milling.^[72] Localized irradiation in nature could enhance signals from such defects, producing high natural signal levels that may be difficult to reproduce in the laboratory.

CONCLUSIONS

Luminescence signals used for dating are intimately related to the crystal and defect structure of the materials from which they are measured. This relationship has been studied since the origins of the field, but remains open to significant developments. Case studies have been used to illustrate different factors affecting luminescence-dosimetric signals, in the contexts of archaeological and geomorphological formation mechanisms, thermal history and mineralogical alterations, and mineral inclusions and defect structures. The studies highlight aspects amenable to analysis using Raman and luminescence techniques as a whole, where developments in understanding of the physical systems involved could be used to enhance the accuracy and broaden the applicability of luminescence-dating methods.

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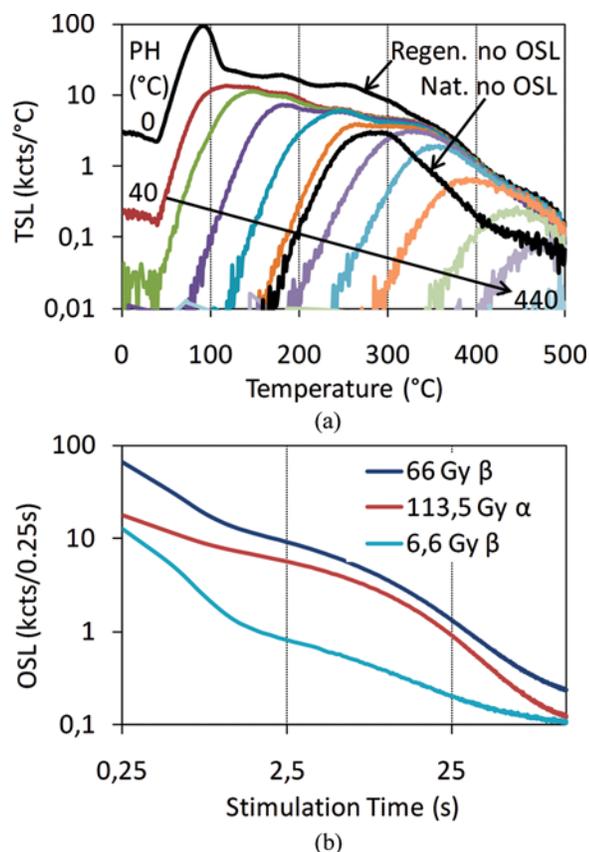


FIGURE 4 (a) TSL of quartz separates (160-250 μm); natural and following regenerative β irradiation, a range of preheats (PH), and OSL. (b) OSL of milled quartz (4-11 μm) following α and β irradiation. Adapted from Reference 64. (color figure available online.)

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