

Optically stimulated luminescence response to ionizing radiation of red bricks (SiO₂, Al₂O₃ and Fe₂O₃) used as building materials

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Quartz is the most common mineral in our environment. It is found in granite, hydrothermal veins and volcanic rocks, as well as in sedimentary deposits derived from such solid materials. These sediments are also made into building materials, such as bricks and pottery. Thus the potential use of a dose reconstruction technique based on quartz grains is enormous, whether as a dating tool in archaeology and quaternary geology, or in nuclear accident dosimetry. This work describes the Optically Stimulated Luminescence (OSL) response of red brick to ionizing radiation. The bricks, from the state of Puebla, Mexico, represent another class of materials that can be used in retrospective dosimetry following nuclear or radiological incidents. The chemical composition of fifteen bricks (three samples from five different brick factories) was determined, using energy dispersive spectroscopy (EDS), to be primarily SiO₂, Al₂O₃ and Fe₂O₃ and is believed to be representative for this common building material. Individual aliquots from these bricks were powdered in agate mortars and thermally annealed. Replicate samples of the aliquots were then irradiated with beta particles from a sealed source of ⁹⁰Sr/⁹⁰Y. The OSL response was measured with a Daybreak Model 2200 High-Capacity OSL Reader System. We present here for this material the characteristic OSL response to beta particles; the reproducibility of the OSL response; the linearity of the response in the dose range 0.47 Gy to 47 Gy; and the fading characteristics.

Keywords: OSL; red brick; building material; retrospective dosimetry.

El cuarzo es el mineral más común en nuestro ambiente. Este se encuentra en el granito, estratos hidrotérmicos y rocas volcánicas, así como en depósitos sedimentarios derivados de estos materiales sólidos. Los materiales de construcción como tabiques y cerámicas, también contienen estos sedimentos. Así, el uso potencial de la técnica de reconstrucción de dosis usando granos de cuarzo es enorme, ya sea como una herramienta de fecho en arqueología y geología cuaternaria, o en la dosimetría de accidente nuclear. En este trabajo se describe la respuesta del tabique rojo a la radiación ionizante, de la Luminiscencia Estimulada Opticamente (OSL). Los tabiques del estado de Puebla, representan otra clase de materiales que se pueden usar en la dosimetría retrospectiva, después de accidentes nucleares o radiológicos. La composición química de 15 tabiques (3 muestras de 5 diferentes fabricas) fueron determinadas, usando Espectroscopía de Energía Dispersiva (EDS) siendo principalmente SiO₂, Al₂O₃ y Fe₂O₃, y se supone que son representativas de este material comúnmente usado en construcción. Alícuotas individuales de estos tabiques fueron pulverizados en un mortero de ágata y sometidos a un tratamiento térmico. Replicas de estas muestras fueron irradiadas con partículas beta de una fuente sellada de ⁹⁰Sr/⁹⁰Y. La respuesta OSL fue medida en un sistema de lectura OSL de alta capacidad, Daybreak® modelo 2200. Presentamos aquí la respuesta característica OSL de esta material a partículas beta, la reproducibilidad de la respuesta OSL, la linealidad de la respuesta en el intervalo de 0.47 Gy a 47 Gy, así como las características de apagamiento de la señal OSL.

Descriptores: OSL; tabique rojo; material de construcción; dosimetría retrospectiva.

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1. Introduction

The aim of this work is to investigate the Optically Stimulated Luminescence (OSL) properties of natural materials [1,2,3,4] - specifically, in this case, the characterization of red bricks made from natural soil and used as building material [5]. In Mexico, red brick is the most popular building material for houses, dwellings and buildings, and it can be used as retrospective dosimetry material after nuclear or radiological incidents [6,7].

In this paper is described the OSL response of red bricks from Cholula and Huejotzingo, in the state of Puebla, Mex-

ico, that for commercial reasons are the most common bricks used for the construction of dwellings in Mexico City.

In order to know the chemical composition of brick from different producers in this region close to Mexico City, Energy Dispersive Spectroscopy (EDS) analysis was performed on the materials. Bricks from different factories, but in the same region, were found to have similar chemical compositions. Results of the EDS analyses provides assurance that the OSL response of the different bricks, no matter the original producers from the same geological formation, will have similar responses to ionizing radiation.

2. Experimental procedure

The OSL response of red brick, like that of other OSL material, can be significantly reduced by bleaching from the blue and ultraviolet components of natural sunlight and indoor fluorescent lighting. Sample handling, from material preparation until the final analysis, was carried out under soft incandescent light (red or yellow) for this study.

2.1. Sample preparation

Fifteen bricks were analyzed, three bricks from five different producers. Samples taken from the bricks were pulverized individually in an agate mortar and pestle. The resulting powder was passed through a 100- μm sieve after the first grinding.

2.2. Annealing

Fines from the 100- μm sieve were annealed in ceramic dishes at 440°C for 1 hour, and then were allowed to cool inside the furnace over a 24-h period to reach room temperature, avoiding thermal stress. This pre-exposure annealing procedure helps to erase background and spurious signals from transport and storage.

2.3. Aliquot preparation

Individual aliquots (10 from each of the 15 brick samples previously ground, sieved and pre-heated) were carefully weighed and transferred to circular aluminium disks, 9 mm in diameter. The powdered samples were held on the disk by a thin, homogeneous film of vacuum grease previously deposited on the surface. (Previous measurements of the aluminium disks coated with the vacuum grease showed no resulting contribution to the OSL signal.) Several disks were prepared with a roughly equivalent mass of red brick powder and several others with different quantities of material. Figure 1 shows powdered brick aliquots deposited on the aluminium disks.



FIGURE 1. Red brick aliquots deposited on aluminium disks.

3. Instrumentation

3.1. Energy Dispersive Spectroscopy analysis method

Energy dispersive x-ray spectroscopy (EDS) is a chemical microanalysis technique performed in conjunction with a scanning electron microscope (SEM). The technique utilizes x-rays that are emitted from the sample during bombardment by an electron beam to characterize the elemental composition of the analyzed volume. Features or phases as small as about 1 μm can be analyzed. When the sample is bombarded by the electron beam of the SEM, electrons are ejected from atoms at the sample's surface. A resulting electron vacancy is filled by an electron from a higher orbital, and an x-ray is emitted, having energy corresponding to the energy difference of the two orbital. The EDS x-ray detector measures the number of emitted x-rays and their energies. The x-ray energies are characteristic of the element from which the x-rays were emitted. The resulting energy spectrum is obtained and evaluated for qualitative and quantitative determinations of the elements present in the sampled volume [8]. For this work, a scanning electron microscope JEM5600-LV equipped with a Noran Vantage EDS detector was used, and 15 red brick samples were analyzed.

3.2. Irradiation (beta exposure)

The red brick powder prepared was exposed to beta radiation from a $^{90}\text{Sr}/^{90}\text{Y}$ sealed radioactive source contained in a Multiple Sample Irradiator, Daybreak[®] Nuclear Systems, Inc. model 801E, with a measured dose rate of 0.147 Gy s^{-1} [9].

3.3. OSL Reader

Three excitation methods are commonly used in OSL analysis: Continuous Wave (CW-OSL), Linear Modulation (LM-OSL) and Pulsed (P-OSL) [10]. The CW-OSL method was chosen for this study. A commercial Daybreak[®] Nuclear and Medical Systems, Inc.[®], model 2200 High-Capacity OSL Reader was used, with the software to provide continuous-wave (CW) laser stimulation, and with a platter that holds 60 samples in an automated transport system (Bernal and Bogard, 2004). The measurements were made in a flowing nitrogen atmosphere using green/blue excitation light (480 nm) at a sample temperature of 125°C. The laser was operated at 90% of full power (30 mW/cm²), and OSL output was measured for 30 seconds (30 readings, each integrating the luminescence signal for 1 s) for each OSL curve. The representative OSL response of Puebla red bricks to beta particles is shown in Fig. 2. The red brick material presents a typical OSL curve similar to any other OSL material.

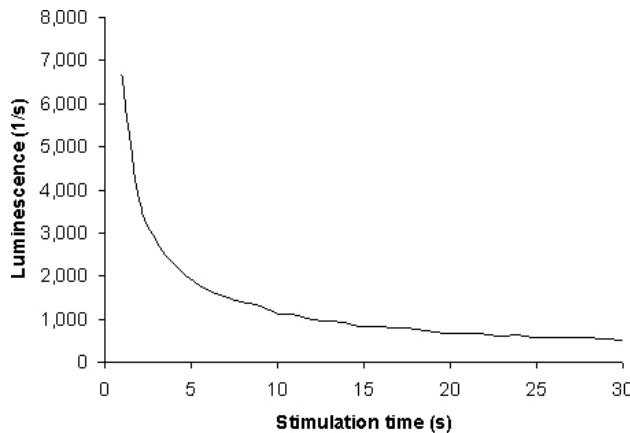


FIGURE 2. OSL response curves of red brick for a beta dose of 14.7 Gy.

4. Results

4.1. Energy Dispersive Spectroscopy analysis

The results of the EDS show that the red brick samples have: Si, O, Fe, C, Ca, K and Na. With the common composition of SiO_2 (57%), Al_2O_3 (13%), Fe_2O_3 (19%), K_2O (2%), Na_2O (0.67%) and CaO (6%). These values are given directly from the software of the EDS analysis. An EDS spectrum of one of the samples is shown in Fig. 3.

4.2. Mass dependence of the OSL response

In order to determine the dependence of CWOSL response with mass of the red brick material, aliquots with 6.1, 13.9, 16.5, 18.0 and 20.0 mg were prepared (5 samples for each mass) and irradiated with the same dose of 7.35 Gy, then read immediately after the irradiation of each set of five. The mass-dependent response is shown in Fig. 4. The OSL response is seen to be directly proportional to mass in the 6- to 20-mg range. A representative mass of 16.5 mg was consequently chosen for subsequent measurements, and OSL responses were normalized to the mass of the sample.

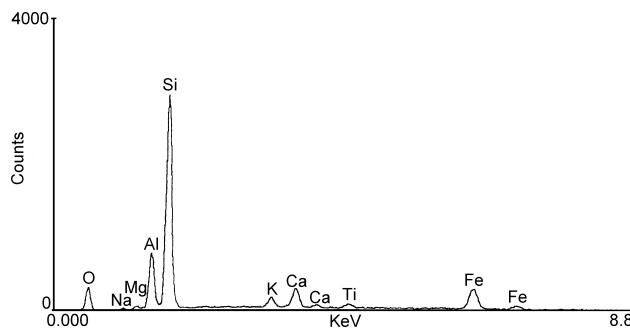


FIGURE 3. EDS spectrum of red brick material.

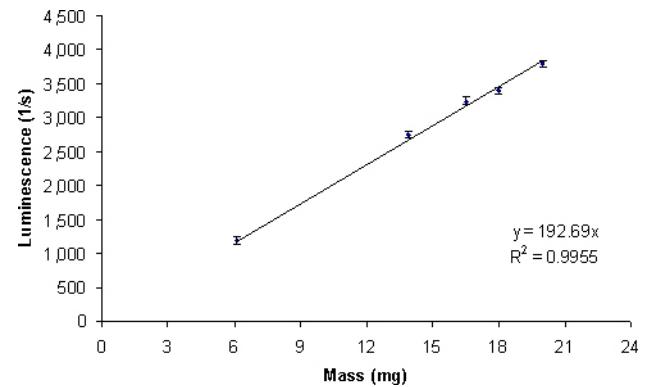


FIGURE 4. OSL response as a function of sample mass.

4.3. Data reproducibility

Five already prepared aliquots of red brick, one from each brick producer and each weighing approximately 16.5 mg, were irradiated with a dose of 7.35 Gy. The aliquots were then immediately analyzed in the OSL reader. This was repeated five times in order to evaluate the reproducibility of the OSL response from each sample. Figure 5 shows the results of five measurements from one of the aliquots.

The average and standard deviation of the OSL maximum intensity from each readout were calculated for brick from each of the five producers. OSL responses for each sample (normalized for mass) are shown, along with the corresponding averages, in Table 1. The grand average for all samples and the estimate of its standard deviation (σ) are also shown. The resulting coefficient of variation (ratio of the sample standard deviation to the average) is 0.13%, showing remarkable agreement between the average responses of material from the five brick producers. These results are in agreement with the fact that the soil of the red brick material come from the same geological formation.

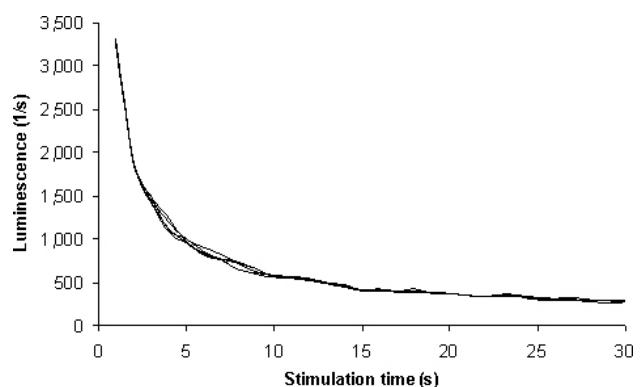


FIGURE 5. OSL results from five irradiations of the same red brick sample.

TABLE I. OSL reproducibility among bricks from different factories (photons per second per milligram).

Factory 1	Factory 2	Factory 3	Factory 4	Factory 5
191	192	192	191	190
193	191	191	192	191
192	194	190	191	194
193	192	192	192	192
192	190	193	193	191
192.2 \pm 0.84	191.8 \pm 1.48	191.6 \pm 1.14	191.8 \pm 0.84	191.6 \pm 1.52
Grand Average = 191.8	$\sigma_g = 0.245$	$\sigma/\text{average} = 0.13\%$		

4.4. Dose response

OSL responses of red brick to doses from 0.735 Gy to 14.7 Gy are shown in Fig. 6. The integration of the initial 1-s of the OSL decay curve is plotted as function of dose in Fig. 7. The linear dose response is a desirable characteristic of materials like brick that might be used for retrospective dosimetry after a radiological incident.

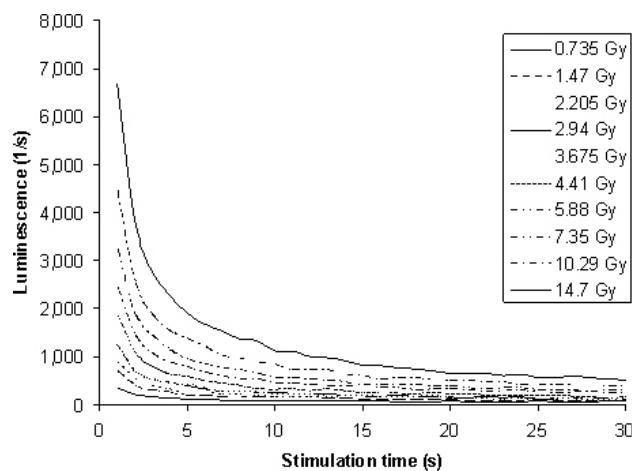


FIGURE 6. OSL response of red brick to different doses.

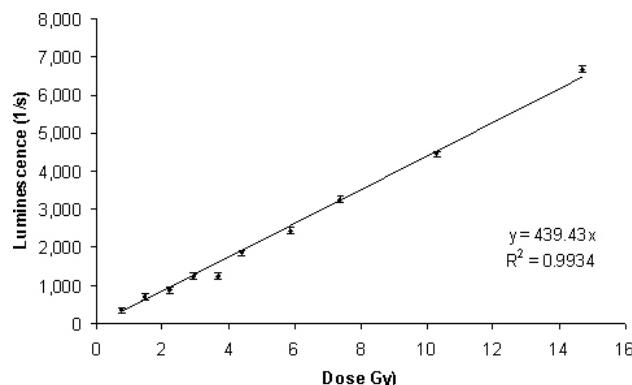


FIGURE 7. OSL response of red bricks with dose.

4.5. Fading

OSL signal intensity may fade with time. The usefulness of red brick as a material for retrospective dosimetry depends on an understanding of the signal fading characteristics, in order to allow correction of the dose estimate after a radiological incident. For this experiment, 12 red brick aliquots were irradiated to the same beta dose of 13.0 Gy. Three samples were measured immediately after the exposure, the next three samples after 24 hours, another three after 66 hours, and the last three after 150 hours. The average values of the OSL signal for each elapsed time are shown in Figure 8. The experimental values fit an exponential decay curve. OSL curves obtained immediately after irradiation (solid line) and after 66 hours (dotted line) are shown in Fig. 9. The shape of the two curves is the same, although the relative intensities differ.

5. Conclusions

These preliminary results show that the OSL response of red bricks from Puebla, Mexico, is linear with ionizing radiation dose and demonstrates a high degree of reproducibility. This material can be very useful for retrospective dosimetry in nuclear or radiological accidents.

Bricks from Cholula and Huehötzingo, Puebla, Mexico, used in this study consist primarily of silicon, iron and alu-

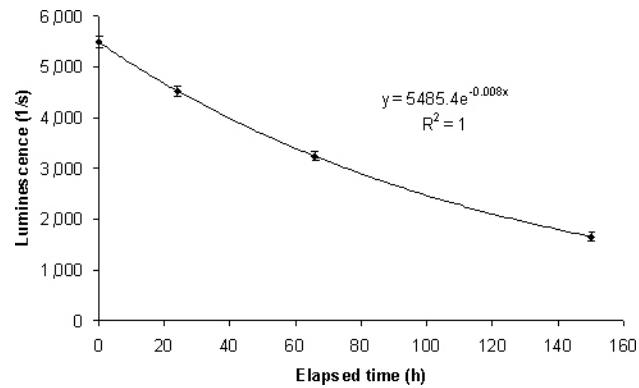


FIGURE 8. OSL fading in red brick.

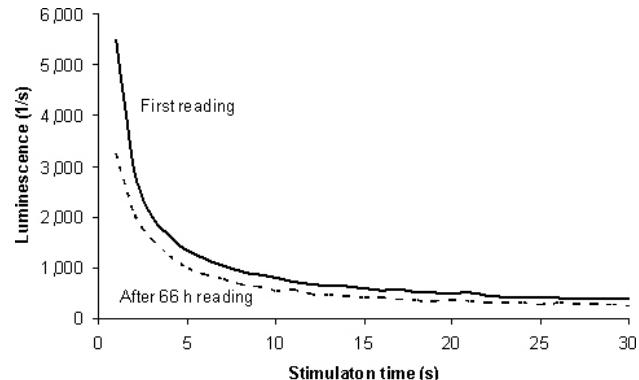


FIGURE 9. OSL response immediately after exposure (solid line), and after 66 h (dotted line).

minium oxides. Bricks from other geographic areas with the same composition would be expected to have the same or similar OSL radiation dose response, but a complementary analytical method, such as EDS, is needed to verify the chemical composition. Alternatively, if chemical composition is different or unknown, dose response studies after recording the as-found signals are required for precise dosimetry.

Other common materials are potentially useful as serendipitous dosimeters, useful for reconstructing doses from a radiological incident. Continued efforts to identify and characterize these materials should continue, given the increased likelihood of such incidents as a result of world terrorism and concerns about uncontrolled or unaccounted-for radiological and nuclear materials.

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