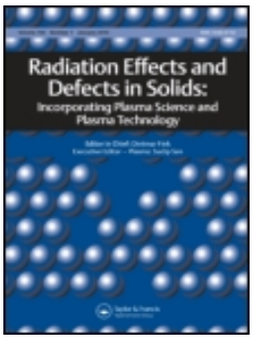

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Radiation Effects and Defects in Solids

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ISSN: 1042-0150 (Print) 1029-4953 (Online) Journal homepage: https://www.tandfonline.com/loi/grad20

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To cite this article: Nilo F. Cano , Neli T.G. Machado , Roseli F. Gennari , Rene R. Rocca , Casimiro S. Munita & Shigueo Watanabe (2012) TL dating of pottery fragments from four archaeological sites in Taquari Valley, Brazil, Radiation Effects and Defects in Solids, 167:12, 947-953, DOI: 10.1080/10420150.2012.702114

To link to this article: https://doi.org/10.1080/10420150.2012.702114

Radiation Effects & Defects in Solids
Vol. 167, No. 12, December 2012, 947–953

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TL dating of pottery fragments from four archaeological sites in Taquari Valley, Brazil

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(*Received 26 March 2012; final version received 8 June 2012*)

Sixty-three pottery fragments from four archaeological sites, numbered RST110, RST101, RST114 and RST114, in the Taquari Valley, vicinity of the city of Lajeado, Rio Grande do Sul state, southern Brazil, have been dated by the thermoluminescence method. Some of them from RST110 and RST101 are as old as 1400–1200 years, whereas those from RST114 and RST107 are younger than 800 years. This result indicates that RST101 and RST110 were peopled earlier than RST114 and RST107. The recent dates found are 302, 295 and 146 years and they are possible, since the first German immigrants who arrived in this region encountered Tupi–Guarani Indians still living there. One interesting result refers to the glow curves of quartz grains RST110, RST101 and RST114 that differ from the glow curves of RST107 quartz grains.

Keywords: thermoluminescence; dosimetry; ancient pottery; quartz

1. Introduction

Schmitz (*1*) states that the prehistorical study of the Brazilian southern coastal plains is largely based on the so-called sambaqui culture. A sambaqui is a shell mound, to which eating refuses are added and less frequently human skeletons. These sambaquis indicate that the southern sea coast of Brazil was peopled as early as 3340 ± 70 years ago.

Guarani and Tupi are two large tribes of natives that initially were separated and occupied northern region of South American Continent. They gradually drifted to the south after giving rise to Tupi-Guarani people by mixing those two tribes, then reaching Bolivia, Paraguay and southern Brazil. Particularly, in this work we are concerned with native Indians in the Rio Grande do Sul State. By radiocarbon dating in the Jacuri river Valley and Taquani Valley, Brochado (*2*) tentatively established the following time periods for Tupi–Guarani settlement: (a) 500 AD – beginning of the settlement; (b) 500–900 AD – Old period; (c) 900–1300 AD – Intermediate period; (d) 1300–1500 AD – Late period; (e) 1500–1800 AD – Colonial period.

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ISSN 1042-0150 print/ISSN 1029-4953 online

http://dx.doi.org/10.1080/10420150.2012.702114
http://www.tandfonline.com

Bona et al. (*3*) carried out archaeometric analyses using energy-dispersive X-ray fluorescence of 120 pottery fragments collected from seven archaeological sites in the central region of the Rio Grande do Sul state. Using principal components analysis of chemical elements found in the investigated samples, these authors pinpointed clay sources used in the production of ceramics and possible interaction between natives.

The thermoluminescence (TL) method has been used in the present study to date pottery fragments, in spite of the considerable development of the dating method based on the optically stimulated luminescence (OSL) technique.

Dating by TL is a particular application of TL dosimetry in which there is a source of constant irradiation (the natural radioactivity of the ceramics), the activity of which can be independently determined. The first successful application of luminescence to the dating of archaeological material was extensively studied by a group at Oxford University headed by Aitken (*4*), but it took several more years and considerable research and development for this method to achieve the status of a reliable dating tool (*5*)–(*8*).

The duration of irradiation is taken to be the same as the age of the ceramic, and this is proportional to the amount of the TL signal. Of course, it is essential to have an initial 'zeroing of the TL signal', this generally being provided in the making of the ceramic itself: the high temperature of about 500 to 600 °C reached by the furnace during the manufacture of the object erased the previously induced TL signal by emptying all of the electronic traps.

In the case of TL dating, ceramics can be considered to consist of a number of crystalline inclusions embedded in the ceramic matrix, mainly quartz and feldspar. The inclusions act as dosimeters of the irradiation arising principally from the natural radioactivity of the ceramic material. The natural radiation responsible for creating the conditions for luminescence has four primary sources: ^{40}K and the decay chains of ^{238}U, ^{232}Th and cosmic radiation (*9*). This natural radioactivity is the source of the annual dose rate (D_{an}) in the ceramics.

The accumulated dose (D_{ac}) was obtained by the additive method through TL dosimetry of the crystalline inclusions in the ceramics (*10*, *11*).

In the present study, pottery fragments of four archaeological sites of the Taquari Valley were dated and analyzed by the technique of TL. By dating pottery fragments of this region, we hope to contribute to find different periods of occupation of these native people in this region of Brazil.

2. Materials and methods

Figure 1 is a map showing the location of the Taquari Valley, where the four archaeological sites are considered. Figure 2 shows some samples of ceramics studied. These and other samples investigated in the present work have been characterized by archaeologists (*12*).

To obtain pure quartz grains for TL readings, each pottery fragments was subjected to the following procedures: the sunlight effect is eliminated by removing a surface layer of approximately 1 mm of the pottery (*4*). After crushing delicately, the powder was sieved to retain grain sizes between 0.080 and 0.0180 mm in diameter. The powder was then immersed in a 20% solution of hydrofluoric acid (HF) for 45 min. The expected effect of HF is to corrode a thin layer of the surface of quartz grains in order to both clear the surface and to eliminate the α-particle effects (*13*). After rinsing in distilled water, the sample was treated with 20% solution of hydrochloric acid for 60 min, to eliminate as best as possible the carbonates and organic materials, to be finally washed again in distilled water.

The D_{ac}-value was evaluated using the additive method (alternatively the regeration method has also been used in some cases). The annual dose value was estimated from uranium, thorium

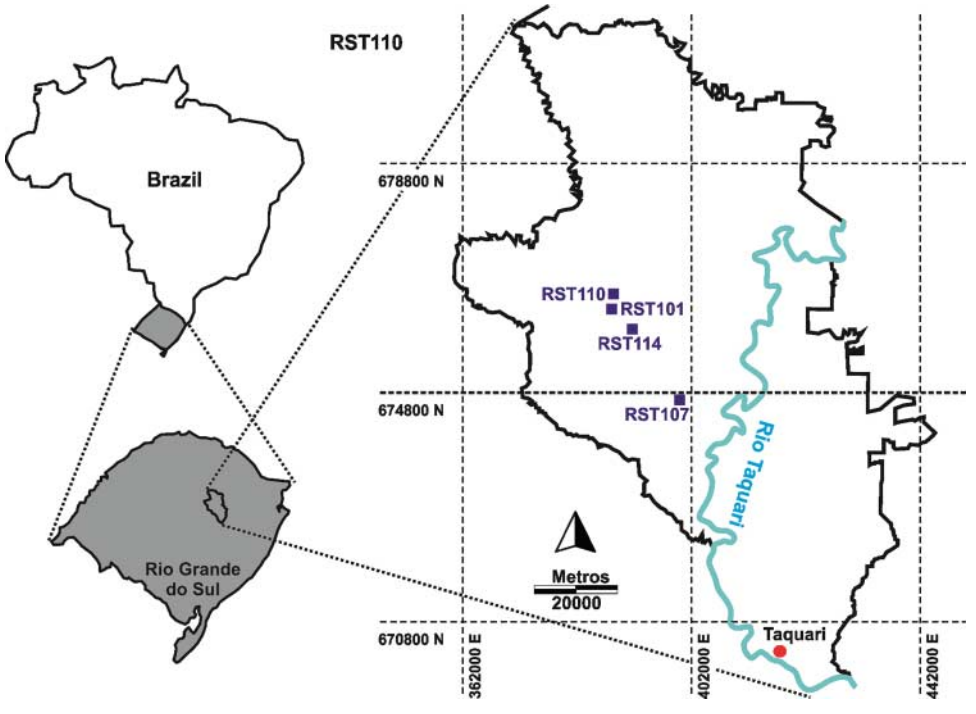

Figure 1. Map showing the location of the Taquari Valley in the state of Rio Grande do Sul and the four archaeological sites RST110, RST101, RST114 and RST107.

Figure 2. Some ceramics fragments from RST114.

and potassium concentrations obtained by ICP-MS mass spectrometer. D_{ac} here is the dose value absorbed by ceramics during the time it has been buried. In the estimation of annual dose D_{an} both internal and external sources should have been considered besides cosmic ray contribution. Now the age of ceramics is given by the quotient of D_{ac} by D_{an}.

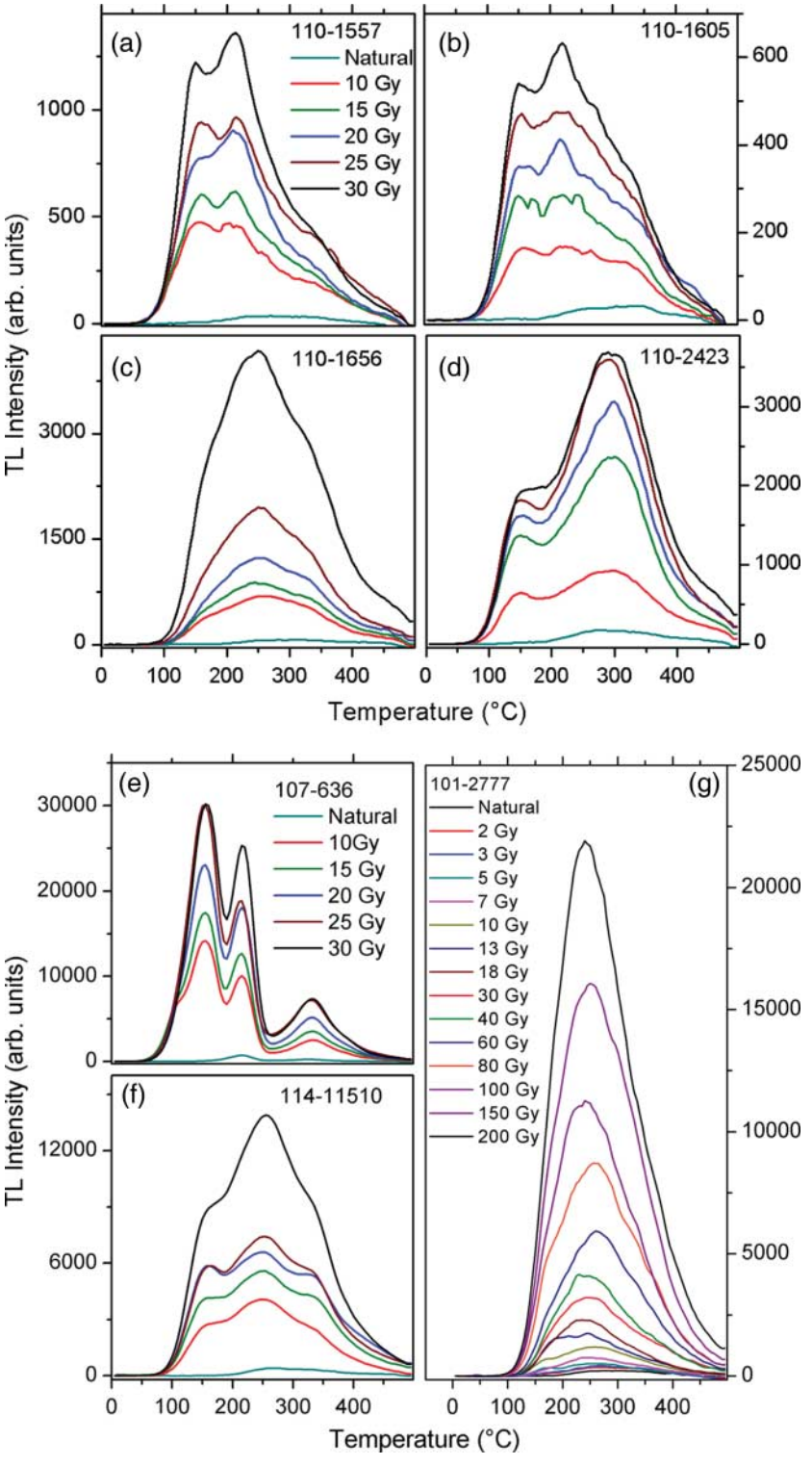

Figure 3. Glow curves of quartz grains from pottery fragments: (a) 110-1557; (b) 110-1605; (c) 110-1656; (d) 110-2423; (e) 107-636; (f) 114-11510 and (g) 101-2777.

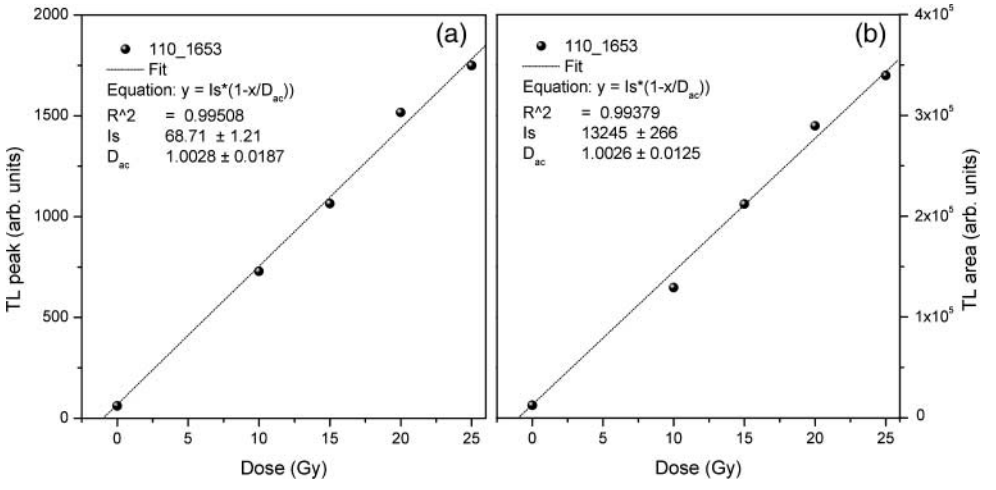

Figure 4. TL response curve as a function of dose: (a) using peak height, (b) using area under the curves, for the sample 110-1653.

TL glow curves were recorded with TL/OSL automated systems, model 1100-series Daybreak Inc, with optical filters Schott BG-39 (300–700 nm) and kopp 7-59, and the heating rate was 4°C/s.

3. Results and some comments

For dating of pottery by TL, usually the 325 °C and 375 °C peaks of quartz are used. In the present case, while the quartz grains from RST107 presented glow curves similar to that of a common alpha-quartz, with distinct high-temperature peaks, those from RST110, RST101, RST114 are characterized by strong and broad peak in the 180–250 °C region such that the high-temperature peak, if present, appears as a shoulder. Therefore, the prominent peak around 220 °C has been used. Figure 3 shows the glow curves of four samples from RST110, one from each of RST101, RST114 and RST107. One extremely broad peak extending from 130 °C to 400 °C in the samples 110-1557, 110-1605; 130 °C and broad 280 °C peaks in the sample 110-1656; 130 °C and very broad 200–400 °C peaks in the sample 110-2423. The glow curve of quartz grains from RST101 has only a very large and broad peak around 240 °C, which extends from 120 °C to close to 400 °C. In the glow curve of quartz grains from RST114, we can observe the broad peak that occurs around 230–250 °C.

As mentioned above, the quartz grains from RST107 presented a superposed 325 °C and 375 °C peak, since most of RST107 samples possess a prominent 220 °C peak and since this peak has long enough mean half life, this peak has been used for the accumulated dose, D_{ac}, evaluation; however, the samples numbers 107-464 and 107-2200 presented a strong high temperature peak, hence this peak has also been used for D_{ac}-value evaluation. The quartz grains from RST101, RST110 and RST114 sites presented prominent peaks around 220 °C and 250 °C; therefore, this peak was used to obtain D_{ac}-value. In few other samples as in 110-2423, the prominent peak at 280 °C and 300 °C was used.

Due to such unusual behavior of quartz grains from sites RST101, RST110 and RST114, we also used area under glow curve to evaluate D_{ac}-values. The comparison of the two D_{ac}-values has shown that the difference between these two values is small (Figure 4). In Tables 1 and 2, the results of dating 63 pottery fragments are shown. The correlation depth-age might be somewhat

Table 1. Accumulated dose D_{ac} in Gy, annual dose rate D_{an} in mGy/a and age obtained.

	Site 101					Site 110			
Samples	Depth (cm)	D_{ac} (Gy)	D_{an} (mGy/a)	Age (years)	Samples	Depth (cm)	D_{ac} (Gy)	D_{an} (mGy/a)	Age (years)
2606	39	1.869	1.14	1636	2272	61	3.101	1.58	1960
2753	57	3.017	2.14	1411	2290	48	2.292	1.88	1222
2776	32	2.272	1.98	1147	2279	57	1.912	1.59	1204
2764	22	3.004	2.68	1121	3753		1.615	1.81	893
2788	19	2.267	2.18	1040	1558	48	1.496	1.82	822
2559	70	2.583	2.63	981	3745	45	1.76	2.15	820
3124	42	3.127	2.85	950	1559	63	1.852	2.29	808
2565	71	1.45	1.68	864	2405	58	1.837	2.50	736
3154	59	2.807	3.28	856	3766	46	1.615	2.21	731
2609		1.424	1.70	838	2364	61	1.116	1.62	690
3065	86	1.923	1.88	829	2439	63	1.212	1.79	678
3174	29	2.057	2.63	782	2435	47	0.906	1.35	670
2955	45	1.342	1.88	714	1605	62	1.108	1.87	593
2512	24	1.917	2.88	667	2423	57	1.481	0.58	574
2517	15	2.336	2.41	653	2425	67	1.59	1.26	569
2563		1.342	2.19	613	2416	56		1.35	531
2959	22	0.796	1.30	612	2366	61	1.383	2.50	554
2607	77	1.418	2.53	554	1557	51	3.393	1.35	448
2561	35	1.500	2.86	525	1656	58	0.874	1.97	444
2745	30	1.245	2.48	503	2350	55	2.371	2.28	394
2747	44	1.123	2.79	402	1653	45	1.003	2.50	402
2763	47	0.618	2.09	295	2277	53	0.724	1.91	379
3094	23	0.364	2.50	146	3751	55	0.774	2.19	351

Table 2. Accumulated dose D_{ac} in Gy, annual dose rate D_{an} in mGy/a and age obtained.

	Site 114					Site 107			
Samples	Depth (cm)	D_{ac} (Gy)	D_{an} (mGy/a)	Age (years)	Samples	Depth (cm)	D_{ac} (Gy)	D_{an} (mGy/a)	Age (years)
12,776	27	1.108	1.42	779	669	98	1.252	1.72	727
10,988	31	1.307	2.1	622	446	127	0.963	1.35	712
13,344	5	1.402	2.3	609	464	119	3.935	1.06	592
10,356	51	1.254	2.5	503	634	135	0.712	1.21	588
12,709	14	0.874	2.03	431	448	155	1.028	1.88	547
11,810	28	0.631	1.87	338	636	129	1.011	1.90	531
					649	142	0.672	1.48	453
					2183	132	0.971	2.50	389
					659	127	0.850	2.41	353
					2200	103	0.825	2.74	302
					2181	119	0.647	2.50	259

changed. These archaeological sites are close to farmland and although no large disturbance might have been introduced some influence cannot be ignored.

4. Conclusions

The most northerly situated sites, namely RST110 and RST101, were peopled about 1400 years ago. 1960 years of age found for the sample 110-2272 seemed too old; however, archaeologists find it still within expected time of arrival of first settlers. The sites RST114 and RST107 have been populated much later, about 800 years and later.

Ages such as 146 and 295 years of number 101-3094 and 101-2763, respectively, are in agreement with the fact that European (mainly German) immigrants arrived in the years 1700 and later encountered many Tupi–Guarani Indians.

The fact that quartz grains from RST101, RST110 and RST114 have different behavior compared with that from RST107 indicates that clay used by people from RST101, RST110 and RST107 is different from clay used by people from RST107. Possibly, we can say that the interaction between the people from these two groups was very small. This conclusion was also reached in statistical analysis of chemical elements contained in pottery fragments as well as in few clays and soil related to the present work, result that will be published elsewhere.

Acknowledgements

The authors thank Ms. E. Somessari and Mr. C. Gaia, Instituto de Pesquisas Energeticas e Nucleares (IPEN), Brazil, for kindly carrying out the irradiation of the samples. This work was carried out with the financial support of the FAPESP and CNPq.

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