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Optically Stimulated Luminescen Dating of Ancient Furnaces at Samaru-Westiron-Smelting Sites

M.K. Aliyu* and M.D. Oladipupo**

Abstract

Optically stimulated luminescence (OSL) dating was conducted on furnace samples obtained from four ancient iron-smelting site at Samaru, Zaria in Nigeria. Quartz grains extracted from the furnace samples contained accumulated radiation doses that were measured with OSL machine. The clay matrix also contained with long-live radioactive elements-Potassium, Thorium and Uranium-whose were determined with method of neutron activation analysis. The Concentration was converted to doserate. It was found that the values of equivalent doses ranged between 3046+300 and 5555+100 with average of 4749+258 while annual doserates ranged between 3.04+0.06 and 4.78+0.02, with average of 3.612+0.050. The average moisture content for each of the samples ranged between 0.043 and 0.077, with average of 0.0602. The ages of the samples ranged between 1002+119 and 1616+181, with average of 1324+128, which corresponds to a range of 388 AD and 1003ADand average of 680AD. This range compared well with those reported by Sutton (1978) thereby confirming that the charcoal dated by Sutton were the remnants of iron smelting. The wider range of age in this study is due

* *M. K. Aliyu, Department of Archaeology, Ahmadu Bello University, Zaria-Nigeria*
***M. D. Oladipo, Department of Physics, Ibrahim Babangida University, Lapai-Nigeria*

to Sutton were the iron-smelting sites compare to only one site dated with radiocarbon. A close look at all the ages shows that two groups of iron smelters occupied SF1 and SF2 around 400AD, and about four hundred years later, 800AD, another two groups occupied SF3 and SF4.

Keywords: *Furnace, Doserate, Equivalent dose, OSL, Samaru, Iron-smelting*

Introduction

Samaru-west-smelting site, located at 1191/2N, 7 37'E is a large ancient industrial site. There have been a number of research works to study the site, among which is the radiocarbon dating of some charcoal excavated from the site (Sutton, 1976), and an investigation, using neutron activation analysis to establish level of similarity between potsherds found at the site and some other nearby sites (Oladipo, et al 1992). It was reported that the charcoals used for radiocarbon dating were excavated from beneath some furnace believed to have been used in the past for iron smelting. The date obtained ranged between 660 AD and 940 AD. However, using radiocarbon age to deduce date of iron –smelting activity in the area is not conclusive, because there is no evidence that the sites were not occupied at any other time before or after the iron-smelting period. This implies that there is a non-zero probability that the charcoals dated could have been there many years either before or after the period of iron-smelting activities. There is need therefore to verify the radiocarbon date with an independent method.

It is certain that furnaces found at the site are remnants of those used for iron smelting. These are suitable to optically stimulated luminescence (OSL) dating. A furnace sample consists of sand grains (minerals, such as quartz, zircon, and feldspars) and clay matrix. In the clay matrix are radioactive elements (mainly uranium, thorium and potassium) with very long half-life, emitting ionizing radiations. The radiations create electron-hole par in crystals of the minerals, and because of energy absorbed, electrons are trapped at a higher energy state within the crystal lattices. The minerals are said to have received radiation dose. When stimulated

with light of suitable wavelength, the trapped electrons recombine with holes, and release light energy of shorter wavelength. The population of trapped electrons determines the intensity of light emitted. During usage of furnaces, heat evicts all electrons previously trapped in the minerals during geological ages, and the clock of the furnace reset to zero. When the iron-smelters abandon the furnace, the minerals in it continued accumulating new set of trapped electrons. The population of the trapped electron is proportional to radiation flux, and length of time of irradiation. By stimulating the minerals extracted from the samples with light in an OSL machine, and compare the intensity of light emitted with that due to a known quantity of artificial radiation dose, the natural dose in the samples can be determined. The doserate due to activity of radionuclide in the samples need to the determined also, to calculate how many years ago was the last time the furnace was used.

Materials and Method

A survey of Samaru west showed that four distinct slagheaps are still present in the area. Beside each of them are crumbles of furnaces. Some of these were collected and labeled. Moisture content of the samples was determined. Each of them was then treated as follows: About 150g of furnace sample was selected and allowed to dry at room temperature. Under subdued red light, the dry sample was thoroughly scratched to remove the outer layer that has been exposed to sunlight. The solid residue was gently crushed, minding not to allow large grains of sand to break into smaller pieces. Nose-cover was used because of clay dust. This was sieved with a 500um sieve. The residue inside the sieve was joined to those initially scratched. The quantity that passed through the sieve was a mixture of sand and clay. This was further sieved to select sand size in the range of 250um and 90 um. Most of clay pass through the 90um sieve and was joined to the other residues. All the residues were kept aside for dose-rat determination. Still under subdued red light, the send was put into a 100ml plastic beaker, and placed in fume chamber. Nose cover and thick hand glove were worn for protection against

poisonous hydrofluoric acid. About 50ml of 10% hydrochloric acid was added to remove carbonates. The mixture was left for ten minutes and then decanted. The addition of the acid was repeated once, decanted and rinsed with distilled water. In like-manner, 10% hydrogen peroxide was added to remove organic materials. The residue sand was rinsed with distilled water and then treated with 50ml of 10% hydrofluoric acid. This dissolves the clay and feldspars. After about thirty minutes of reaction, ten thick, brownish liquid was decanted and the left for another ten minutes. The residue sand in the beaker was brownish white. It was thoroughly rinsed with distilled water to get rid of clay and dissolved feldspars. About 10ml of 40% hydrofluoric acid was then added, to etch the quartz grains and dissolve away any residue feldspar. This was left for 45 minutes. The clean quartz grains was rinsed and placed in oven to dry at 60c for a day. It was finally sieved to obtain grain sizes in the range of 90um to 180um. This was labeled and kept in lightproof bag, for onward transfer to OSL laboratory.

The OSL machine, TL/OSL system, model TL-DA-15 from Riso National Laboratory, Roskilde, Denmark has a turntable panel attached to module containing switches for lifting and lowering lid of the panel. In the turntable panel are facilities for optical illumination of the samples, a heater and a photomultiplier tube for detection of light emitted by the samples. Its activity at time of supply in year 2000 was 1.49GBq. The doserate per second to a sample from this was checked, using quartz grains that were irradiated with standard source of radiation in Denmark. Based on 28 year half-life of strontium 90, the present dose rate D at a time, from the date of measurement was calculated. The lifting and lowering of strontium source in the machine is done through pressurized air from a compressor, which pressure was usually set at 2 bars. The control of the turntable panel, which includes turning of carousel tray, heating and optical illumination of samples, as data acquisition through the photomultiplier tube, is controlled by a computer mini-system. This receives instructions from a host computer in which there are software programmes – The SEQUENCE EDITOR and ANALYST.

Under subdued red light, the samples were sprinkled on to five aluminium discs that only fabricated sprinkler. The discs were carefully

shaken to remove excess grains on them, so that only one layer of the grains is formed on each of the discs. These were positioned on the carousel tray in such a way that they are remote from each other. The positions used are 1,4,7,10 and 13. This is to prevent cross-talk-a situation where part of artificial irradiation given to an aliquot is absorbed by those near it. The single aliquot regenerative (SAR) dose protocol developed for dating of geological sediments (Botter-Jensen, 2000: Murray, et al, 2000) was adopted for running these samples. A SAR sequence was written on the sequence editor programme, as shown in table 1

Table 1 Sequence of Programme Used to Run the OSL Machine. (Botter-Jensen, 2000)

RUN ON	ACTION TO PERFORM BY OSL MACHINE
1	Prepheat at 260c for 10 seconds
2	OSL (Blue) for 40 seconds (at elevated temperature of 125c
3	Give test dose (irradiation with Sr-90)for 20 seconds
4	Preheat at 160c for 10 seconds
5	OSL (Blue) for 40 seconds (at elevated temperature of 125 c
6	Give regeneration dose (irradiation with Sr-90) for 30 seconds
7-41	Repeat step 1-6 with regenerative does of 50,80,120, and 0 second

OSL Data collection

For each of the run in the sequence above, the computer automatically downloaded data and plotted a decay curve. Integration limits are selected manually from these decay curves, and all the decay curves of an aliquot are combined together to plot a single aliquot regenerative growth curve, using a programme called ANALYST in the computer system. The average paleodose for the five aliquots was obtained for each sample, and for each aliquot of a sample, different curves were analyzed. This includes

linear, cubic and exponential curves. The curve that produced best fit was accepted for each aliquot, to obtain the equivalent dose. The equivalent dose values are obtained as raw values in units of seconds. This was converted to real dose in Gray, using the value of dose-rate per second obtained with calibrated standard. This was combined with annual dose-rates to obtain the age, as:

$$\text{Age} = \frac{\text{Equivalent dose}}{\text{doserate}} \qquad (1)$$

Dose Rate Measurement

Potassium-40, Thorium-232 and Uranium-238 are the major source of naturally occurring radiations. To determine their concentration in the samples, neutron activation analysis method was employed. The source of neutron was the Nigerian nuclear reactor (NNR-1) at Center for Energy Research and Training, Ahmadu Bello University, Zaria. Dried samples were powdered in agate mortar. About 0.3 mg was carefully weighed and sealed as a capsule in polythene leather. This was put into plastic vial and packed with cotton wool before sealing with cellotape. Two sets of capsule were prepared for each of the samples. One is for short irradiation, to analyze the concentration of potassium, and the other for long irradiation, to analyze the concentration of thorium and uranium. For short irradiation, samples were subjected to two minutes of irradiation in a neutron flux of 1011 n/cm2sc. After removal from the reactor, the samples were allowed to cool till radiation dose fall to a level save for handing (about 30umSv). For long irradiation, samples were placed in a neutron flux of 5 x 1011n/m2sce for six hours. When removed from the reactor, they were left to cool for four days before counting. The samples were mounted near a germanium detector for counting. The resulting energy spectrum was analyzed with a multi-channel analyzer. This has a built-in library of radionuclide, and displayed a list of radionuclide detected in the sample with their concentrations. Three

standards – soil-7, fly ash and IAEA-312 – were included in the irradiation and analysis, for quality assurance test. Table 3.2 shows the energy peaks used to analyze elements of interest.

Table 2 Energy peaks used for identifying uranium, thorium and potassium in neutron action analysis

ELEMENT	ISOTOPE	HALF-LIFE	ENERGY (KeV)
Uranium	Np-239	2.36 day	277.6
Thorium	Pa-233	27.0 day	312.0
Potassium	K-42	12.36 hour	1524.6

The concentrations of the elements were obtained in part per million (ppm). These were used with values of doserates (per ppm) in table 3, to calculate annual doserates.

Calculation of doserates from values of elemental concentration

The values of concentrations (ppm) obtained were converted to dose, using conversion factors provided by Aitken (1985). This comprise of the total energy of all alpha particles, beta particles and gamma rays involved in the decay scheme of a particular decay series, when there is elemental concentration of one part per million (ppm). The formula for obtaining the dose is:

D(Gy) = Engergy (J) x specific activity (BqKg-1) x 3.15 x 107 (2)

For concentration of 1 ppm, the dose rates are as shown in the table 3.3.

Table 3: The dose rate (uGy per annum) and specific activity (Bqkg-1) corresponding to concentration of 1 ppm. (Aitken, 1985)

Element	Dose – rates (uGy per annum)		Specific ativity (Bqkg1)	
	Alpha particles (uGy/a)	Beta Particles (uGy/a)	Gamma rays (uGy/a)	
Potassium	-	0.083	0.0244	0.0317
Thorium	738	28.60	51.40	4.08
Uranium	2779	146.10	114.9	13

Result and Discussion

The result of equivalent dose, doserate, moisture content and the corresponding age of each of the furnaces are presented in table 5.

Table 5. The moistures contents, doserates, equivalent dose, and ages of Samaru-west furnaces

N/S	Sample	Equivelent Dose (mGy)	Doserate (mGy/a)	Moisture Content	Age Years	AD
1	SF 1a	3046 + 300	3.04 + 0.06	0.070	1002 + 119	1003 AD
2	SF 1b	5172 + 167	3.20 +0.03	0.077	1616 + 181	388 AD
3	SF 2	5396 + 373	3.40 +0.09	0.54	1587+ 202	418 AD
4	SF 3	4577 + 350	3.64 +0.05	0.057	1162 +26	843 AD
5	SF 4	5555 + 100	4.78 + 0.02	0.057	1162 + 26	843 AD
	Average	47492.2 + 258	3.612 + 0.059	0.0602	1324.8 + 128	680 AD

The values of equivalent dose ranged between 3046+300 and 5555+100 with average of 4749+258, while annual doserates were found to be between 3.04+0.06 and 4.78+0.02, with average of 3.612+0.050. The SF4 site location has relatively high background radiation, which resulted in high equivalent dose and doserate. The average moisture content for each of the samples ranged between 0.043 and 0.077, with average of 0.0602. This implies that average moisture content of the samples is usual about 6% the bulk mass of a sample. This depends on seasonal variations in the area. It is unlikely however that the value will ever fall below 4% or rise above 8%, considering the fact that the annual rainfall of area under study is between 46.4cm and 66.7cm per annum (Encarta, 2008). According to Aitken (1985), the upper limit of moisture content for a sample that is wet throughout the year is about 10%.

The ages of the samples ranged between 1002+119 and 1616+181, with average of 1324+128, which corresponds to a range of 388AD and 1003AD and average of 680 AD. This range is wider than but includes the range of radiocarbon date of 660AD and 690AD reported by Sutton (1976). The average age obtained here, 680AD also full within the of radiocarbon date range. These shows that the charcoal dated by Sutton were the remnants of iron smelting. The wider range

of age in this study is expected, because samples were obtained from four different locations, unlike charcoals that were found at only one of the site, which was labeled as SF3 in this work. The result of SF3 in this study,748AD fall within 660AD 940AD radiocarbon date. This confirms that iron smelting took place at the site around 700AD. There is similarity between values of SF1b and SF2. Similarly, the values of SF3 and SF4 are almost equal. The sites of SF1 and SF2 are about twenty meters part, and so also are SF3 and SF4. Each of this group are however about one kilometre apart. There is however a significant difference in the ages of SF1a and SF1b. SF1b lies by side of a footpath and it is possible that someone has made fire on top of it sometime ago. This could be reason for the lower value of equivalent dose the resulted in a top of it a relatively lower age. A close look at all the ages show that two groups of iron smelters occupied SF1 and SF2 around 400AD, and about four hundred years later, 800AD, another two groups occupied SF3 and SF4.

Conclusion

Optically stimulated Iuminescence dating was conducted on furnace samples obtained from four ancient iron-smelting site at Samaru, Zaria in Nigeria. Quartz grains extracted from the furnace samples contained accumulated radiation doses that were measured with OSL machine. The clay matrix also contained long lived radioactive elements: Potassium, Thorium and Uranium. Their concentrations in the samples were determined with method of neutron activation analysis. The concentration was converted to doserate. It was found that the values of equivalent doses ranged between 3046+300 and 5555+100 with average of 4749+258, while annual doserate were found to be between 3.04+0.06 and 4. 78+0.02, with average of 3.612+0.050. The average moisture content for each of the samples ranged between 0.043 and 0.077, with average of 0.0602.

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