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Search for superheavy elements in monazite from beach sands of South India

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Abstract. Monazite minerals obtained from beach sands of South India were examined for the presence of superheavy elements with photon-induced x-ray fluorescence method. The accumulated data of a number of runs each of several days duration do not show any convincing peaks above the background at the expected locations for superheavy elements which are above the present sensitivity of detection of about 10 ppm by weight for element 126. However, some intriguing features pertaining to structures in the x-ray spectra around 27 keV were observed, which are of interest for further investigations.

Keywords. Superheavy elements; search in monazite; photon-induced x-ray fluorescence.

1. Introduction

There have been a number of experimental studies in the past for the search of superheavy elements in nature motivated by the theoretical prediction of existence of an island of relatively stable superheavy elements much beyond the present periodic table (see for example reviews on the subject by Thompson and Tsang 1972 and Kapoor 1972). Earlier experimental work to find out superheavy elements in nature around the theoretically predicted island of elements with atomic numbers 101-114 did not result in any positive finding. However, a recent communication by Gentry *et al* (1976) reported x-ray spectra of some Madagaskar monazite inclusions in biotite mica induced by low energy protons, which were interpreted to imply the presence of superheavy elements of atomic numbers 116, 124 and 126 in those monazite inclusions which are accompanied by giant halos in mica. The concentration of superheavy elements in these manozite inclusions was estimated to be as high as 10^2 to 10^3 ppm. These surprising findings have also led to a theoretical re-examination of whether an island of stability could indeed be in the neigbhourhood of Z=126 rather than Z=114 as shown by earlier calculations (Anderson *et al* 1976, Wong 1976, Moller and Nix 1977, Petrovich *et al* 1976). Further, following Gentry *et al*'s (1976) communication, a number of other workers have examined similar types of monazite inclusions in biotite mica as well as bulk monazite samples drawn from the same Madagaskar location but have not found any evidence for the presence of superheavy elements at concentrations down to 1-2 ppm (Bosch *et al* 1976, Ketelle *et al* 1976, Sparks *et al* 1977).

In this paper we report results of our examination of bulk monazite samples from beach sand deposits in South India, by photon induced x-ray, fluorescence method with regard to the presence of any superheavy elements. The photon induced x-ray analysis has some advantages over the proton induced analysis. Firstly, there are no uncertainties due to the γ-rays excited in the Ce(p, $n\gamma$) reaction on Ce present in the inclusion which have nearly the same energy as expected for the L_α line of element 126. Secondly, the x-ray excitation cross-sections of superheavy elements relative to that for thorium are about two orders of magnitude higher for photons (40-60 keV) than for protons (5-6 MeV), resulting in an increased detection capability with photon induced x-ray analysis.

2. Experimental

A 5 mm diameter collimated beam of electromagnetic radiations from a 100 mci ^{241}Am source filtered through a 1 mm thick copper disc was used for x-ray excitation of the sample. The filtration ensured that the emerging photon beam constituted only the 59·57 keV γ-rays, since other radiations of 26·36 keV and lower energies were attenuated by 10^7 or more relative to 59·57 keV γ-rays. In this investigation, we have examined monazite minerals obtained from the Chavara (CH) and Manavalakurichi (MK) beach sands of South India. Samples of thickness of about 25 mg/cm^2 were prepared by sandwiching the powdered monazite samples between two thin adhesive tapes. The samples were mounted at 45° to the incident photon beam direction, and the fluorescent x-rays were energy analysed by a cooled 30 mm^2 $\times$ 3mm Si(Li) x-ray spectrometer of about 230 eV energy resolution. In the chosen geometry the Si (Li) detector was kept at a distance of 3·0 cm from the sample to detect fluorescent x-rays at right angles to the incident beam. The experimental geometry and the shielding arrangement for sample excitation and fluorescent x-ray detection were optimized to ensure maximum count rates for fluorescent radiation with a minimum of background counts resulting from the scatter of direct beam from the sample holder and environment. The energy calibration of the x-ray detector system was carried out through the use of standard sources and on line energy calibration during the run was also provided by the known elements occurring in monazite.

3. Results and discussion

A number of runs with different samples, were taken during the course of this investigation. Figure 1 shows a typical spectrum observed for a monazite sample, which is similar to the spectra seen by Gentry *et al* (1976). The x-ray peaks of thorium, uranium and rare earths which are the primary constituents of monazite dominate the spectrum. In some of the runs like that shown in figure 1, where Ta and Pb were present in the collimator placed in the path of fluorescent x-rays, the x-ray lines of these elements were also seen. As expected, a low background window exists in the spectrum in the energy region from 22-31 keV which is free from the emission x-ray lines of the known constituents of monazite. This is also the energy region of interest in the search for superheavy elements, as x-ray emission lines of a range of these superheavy elements are expected to fall in this window, as can be inferred from figure 2 which shows calculated energies of L_{α_1}, L_{α_2}, L_{β_1} and L_{β_2} x-ray lines of elements of atomic numbers 100-130. The primary source of background counts

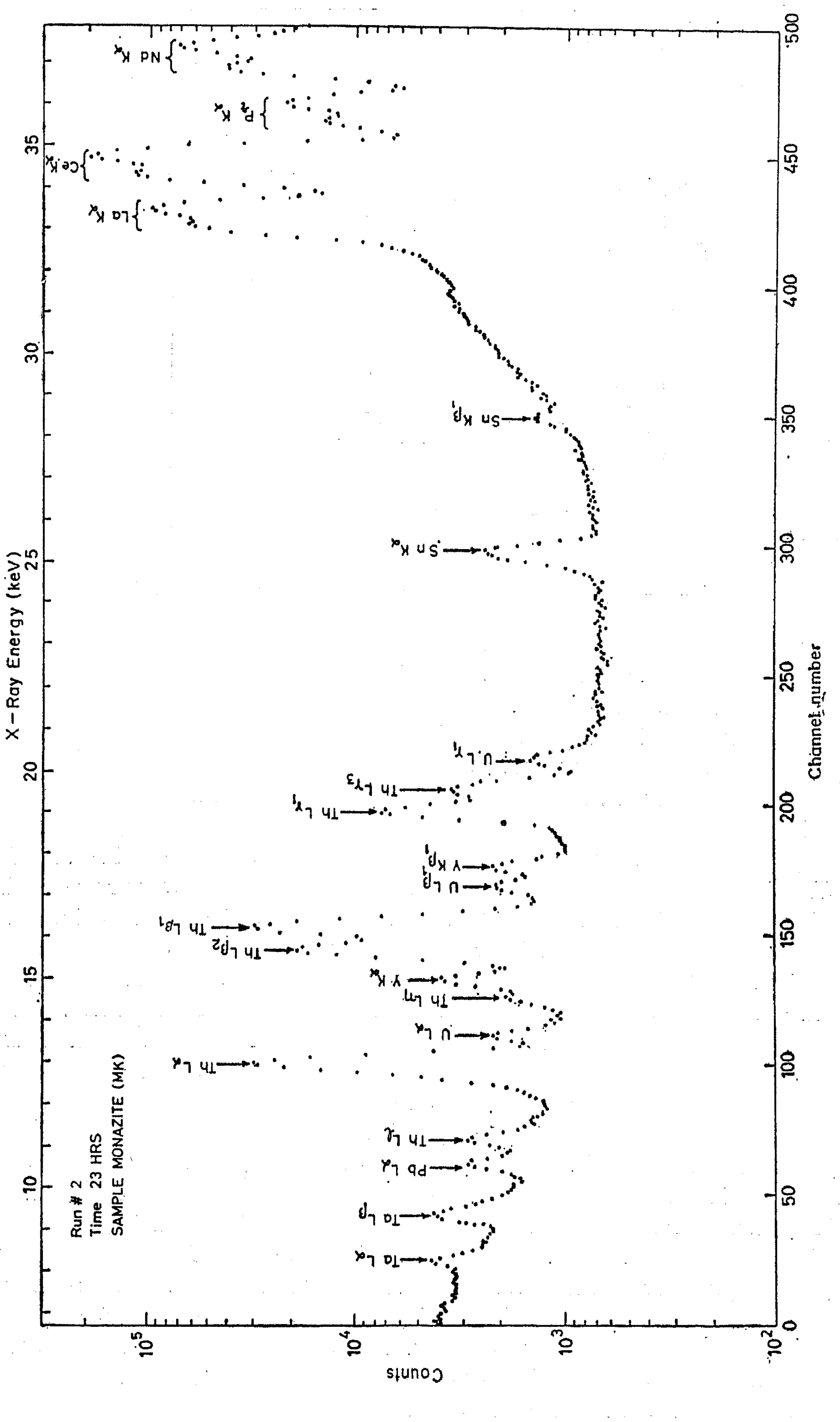

Figure 1. A typical x-ray spectrum of a monazite sample.

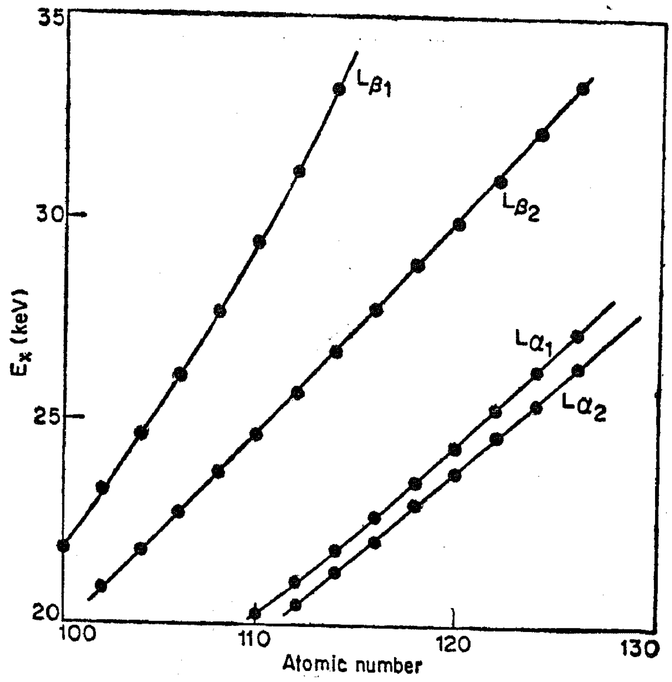

Figure 2. Energies of some prominent L x-ray lines of superheavy elements.

in the window region is the low energy tailing background of the *K* x-ray peaks of rare earth elements, and the spectra recorded in the present experiments generally yielded about 250 as the ratio of Ce peak channel counts to average background counts per channel. In the window region, we also see distinct peaks due to Sn *K* x-rays primarily resulting from the fluorescent excitation of the solder material present in the vicinity of the detector, by the *K* x-rays of the rare earths incident on the detector.

Independent runs each of several days duration, were taken to examine a number of CH and MK monazite samples. Figure 3 shows portions of the spectrum covering the window region for CH monazite and MK monazite samples. The position of *K* lines of various elements which fall in the window region are also indicated. There are noticeable indications of peaks in the spectra which can be attributed to the presence of Ag and Cd in trace amounts. It is likely that these elements also have their origin in the solder material. We have also shown in figure 3 a spectrum taken for a synthetic sample (S) of Ce, La and P_2O_5 mixed in the right proportion to simulate the background producing constituents of monazite. This spectrum is also seen to have peaks similar to the monazite spectrum in the window region of 22·0 to 31·0 keV supporting the above explanation for the origin of these peaks due to *K* x-ray lines of trace impurities of Sn, Ag and Cd present in the detector environment.

Since the spectrum for CH monazite has better counting statistics, this can be examined in more detail for any indication of the presence of *L* lines of superheavy elements, in particular of elements 116, 124 and 126 which were claimed to be indicated in the giant halo inclusions studied by Gentry *et al* (1976). It is seen from figure 3 that there is no convincing evidence of peaks clearly standing out of background at the calculated x-ray energies of superheavy elements, although a small structure at the location of element 126 is not ruled out. The sensitivity of detection of superheavy elements which depends on the background was calculated as follows:

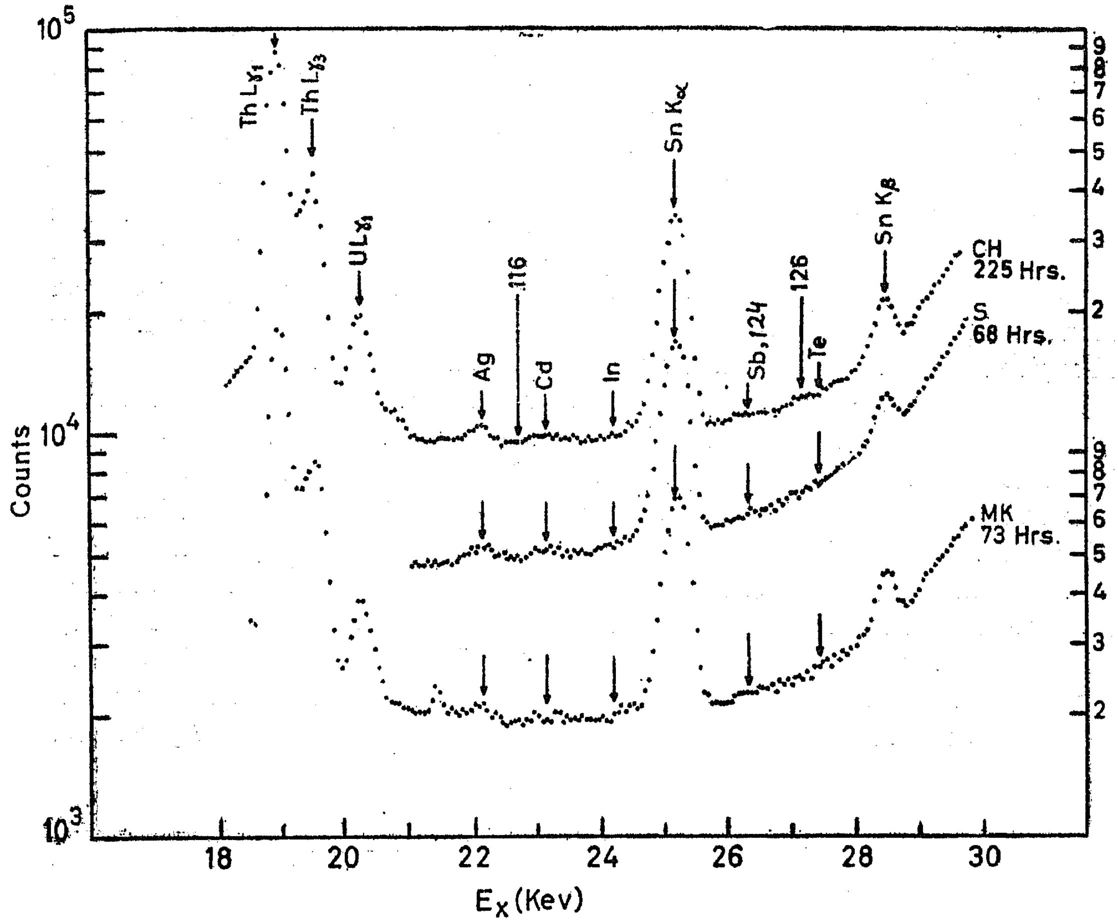

Figure 3. Portions of x-ray spectrum for CH monazite, MK monazite, and synthetic sample.

The number of fluorescent radiation events from an element *i*, which belong to the energy of interest can be written as

$$N_i = I_0 t \, n_i \, \epsilon_i \, T_i \, \sigma_i$$

where I_0 is the number of incident photons/cm²—sec, *t* the counting time, n_i the number of atoms of element *i*, σ_i is the fluorescence cross-section of element *i*, ϵ_i the product of detector efficiency and solid angle of detection, and T_i is a correction factor for self absorption of the fluorescence radiations in the sample.

It then follows that the detectable number of $Z=126$ atoms, relative to that of thorium atoms in the sample is given by

$$f = \frac{N_1}{E_1 T_1 \sigma_1} \times \frac{E_2 T_2 \sigma_2}{3\sqrt{N_B}}$$

where suffixes 1 and 2 refer to the Th $Lr_{1,2,3}$ peaks and 27·27 keV energy respectively, N_1 integrated counts under Th $Lr_{1,2,3}$ peaks and N_B is the integrated background counts in an energy spread of 800 eV (2·3 FWHM of the detector) centred at 27 KeV. With the values of σ_1/σ_2 taken from the paper of Sparks *et al* (1976) and experimentally measured values of ϵ_1, ϵ_2, T_1, T_2, the sensitivity of detection of element 126 is calculated to be about 10 ppm by weight for the CH monazite spectrum shown in figure 3.

We therefore do not find definite evidence for the presence of superheavy elements in beach sands of South India in concentration exceeding our sensitivity of detection of about 10 ppm by weight for element 126.

There are, however, some intriguing features observed in this work which need to be pointed out. As mentioned earlier, the spectra shown in figure 3 are the sum of several independent runs. Figure 4 shows spectra for some of these individual runs taken for CH monazite samples. A close examination of the individual spectrum shapes around 27 keV does indicate that these shapes are not completely smooth and the background appears to be modulated with some interesting structures; Although the estimation of intensities of these structures or peaks sensitively depends on the assumed shape of the background, their existence outside counting statistics appears real. Small structures or peaks in the spectrum can originate from a process of inelastic resonance scattering (Sparks 1974) of the fluorescent radiations generated in the sample from the constituents of the sample itself. In fact, the K lines of lanthanum and cerium undergoing inelastic resonance scattering by the L edges of the same elements can give a number of peaks in the energy region around 27 keV some of which can interfere with the L x-ray lines of superheavy elements. However, we do not see any noticeable structure around 27 keV in the background for the case of synthetic sample and this implies that the probability of this second order process is negligibly small and the effects of this process need not be considered. Appearance

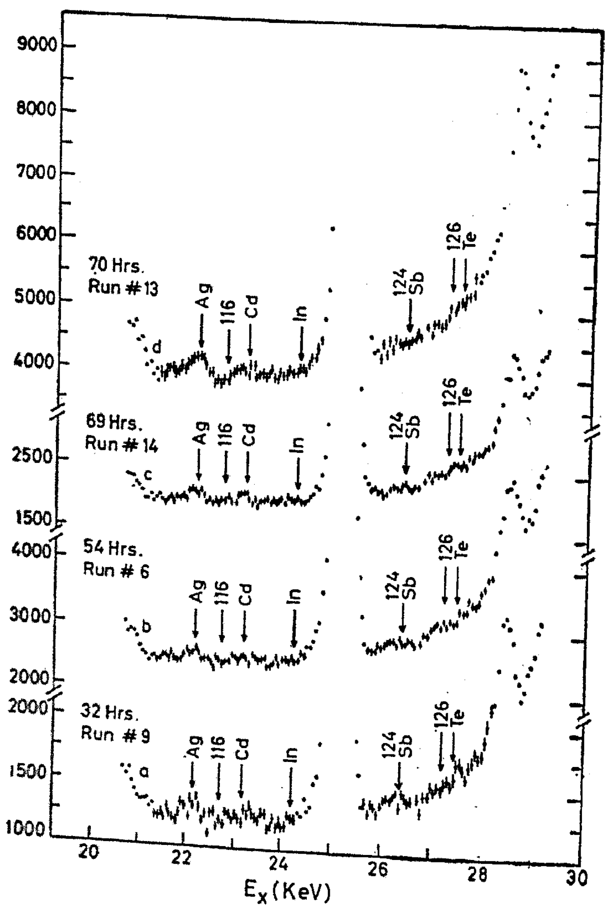

Figure 4. Portions of x-ray spectra of individual runs for CH monazite samples.

of any noticeable structures in the background around 27 keV for CH monazite sample therefore appears to be of some significance. There is, however, another intriguing feature that structures seen in figure 4 in the individual spectra around 27 keV do not repeat in all the runs. For example, a structure similar to the doublet observed by Gentry *et al* (1976) in the energy range of 27-28 keV for the giant halo inclusions is indicated in figure 4(b), although it is not so apparent in other runs. Considering that different runs correspond to different samples and sample positions as seen by the collimated incident photon beam, the question does arise as to whether there are any pockets of superheavy elements in the samples, even though such a possibility is difficult to imagine for powdered monazite samples. The present results are of sufficient interest to continue further investigations along these lines.

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References

Anderson G *et al* 1976 (Preprint)
Bosch F *et al* 1976 *Phys. Rev. Lett.* **37** 1515
Gentry R V *et al* 1976 *Phys. Rev. Lett.* **37** 11
Kapoor S S 1972 *Sci. Cult.* **38** 22
Ketelle B H, O'Kelley G D, Stoughton R W and Halperin J 1976 *Phys. Rev. Lett.* **37** 1734
Moller P and Nix J R 1976 *Phys. Rev. Lett.* **37** 1461
Nix J R 1972 *Phys. Today* April 30
Petrovich F *et al* 1976 *Phys. Rev. Lett.* **37** 558
Sparks C J Jr 1974 *Phys. Rev. Lett.* **33** 262
Sparks C J Jr *et al* 1977 *Phys. Rev. Lett.* **38** 205, 617
Thompson S G and Tsang C F 1972 LBL 665
Wong C Y 1976 *Phys. Rev. Lett.* **37** 664