

CRN 93-41

1



CRN
STRASBOURG

CRN - 93-41

FERMILAB
APR 27 1994
LIBRARY

Activity limit for radioactive break-up of a primordial S isomer

A. Pape, M. Debeauvais, and C. Heitz

**Centre de Recherches Nucléaires et Université Louis Pasteur,
23, rue du Loess, 67037 Strasbourg Cédex 2, France**

NAME	ID NO	M	S.

CENTRE DE RECHERCHES NUCLEAIRES
STRASBOURG

RETURN TO FERMI LAB LIBRARY

IN2P3
CNRS

UNIVERSITE
LOUIS PASTEUR

Activity limit for radioactive break-up of a primordial S isomer

A. Pape, M. Debeauvais, and C. Heitz

**Centre de Recherches Nucléaires et Université Louis Pasteur,
23 rue du Loess, 67037 Strasbourg Cédex 2, France**

Following a recent suggestion that the symmetrical break-up of long-lived isomers could produce the characteristics observed for the class of pleochroic halos known as the dwarf halos, we have looked for coincident O ions that might issue from S using solid state nuclear track detectors. No events could be unambiguously identified, giving activity limits of $A \leq 1.7 \times 10^{-10}$ Bq/ μg for naturally abundant S, and $A \leq 4.7 \times 10^{-10}$ Bq/ μg for S enriched principally in ^{34}S (23x) and ^{36}S (36x).

PACS numbers : 23.70. + j (Heavy-particle decay)
27.30. + t ($20 \leq A \leq 38$)

Measurements have been made on thin CdS films sandwiched between Lexan detectors to look for eventual O (or other heavy) ions emitted in the break-up of a primordial S isomer. A radioactivity of this type has been suggested¹ to account for the class of pleochroic halos known as the dwarf halos.^{2,3} In the over three-quarters of a century since these structures were first observed, no other explanation has ever been offered that might account for their three distinctive features, namely their 1.5 - 11 μm radii, their overexposed disks and their single halo rings. In a day when low-energy nuclear research has slowed to a standstill precisely because of the advanced state of knowledge of nuclear systematics, this state of affairs is at the least disquieting. While the third above-mentioned feature, the single rings, is particularly indicative of a symmetrical break-up with the nuclear structure consequences that follow, any such radioactivity must be very weak. There is still the question of the atomic number of the precursors, although correlations found in widely dispersed old data suggest that one precursor might be found with the element S.

Films of 25 and 50 $\mu\text{g}/\text{cm}^2$ of CdS were evaporated onto one Lexan detector and covered by a second. CdS forms stable homogeneous films with the S being a 22 % constituent. (Films of S in elemental form are composed of microscopic spheres following evaporation that spontaneously migrate and crystallize.) The two disks of Lexan forming a detector array on either side of a CdS film were pinned to a

board, sealed in a plastic envelope under vacuum to ensure close proximity, and stored in the dark at 0°C. After exposition for some weeks, spatially coincident trajectories were searched for by a combination of chemical etching (CE) and electrochemical etching (ECE). The etching conditions were chosen such that α -particle trajectories in Lexan were not revealed. Thus an α -particle cascade following Rn decay, for example, which can produce spatial coincidences in more sensitive detectors by either etching technique, is not observed.

CE⁴ of a detector depends on there being a faster chemical attack along an ion path than on the bulk surface, with the result that a visible track is etched along the ion trajectory. For low-energy ions, however, the cone-like tracks are very small and localizing a few randomly distributed tracks by microscope scanning would be a herculean undertaking. Sometimes they can be difficult to identify reliably. On the other hand, once found, the track diameters and lengths furnish low resolution information on the identity and energy of the impinging ions. Conditions for CE : 5N NaOH, 60°C. A CE of 2h is optimal for developing 0.680 - 2.000 MeV ¹⁶O ions. The minor track diameters are \approx 0.5 - 2.0 μ m. The ranges⁵ of 1.41 - 2.72 μ m are diminished by the 0.45 μ m/h of Lexan etched from the bulk surface. Trajectories of ions incident at 30° (or greater) to the plane of the detector were visualized.

ECE⁶ uses an oscillating high voltage discharge to create an arborescence around the tip of an etch cone as it is formed along an ion path. The resulting so-called trees are readily localized under low magnification, but their diameters are not characteristic of the ion responsible for their formation. Conditions for ECE : 6N KOH, 45°C, 2.9 V/μm or 500 V for the 175 μm thick Lexan, 1 cm cell radius, 50 Hz for 2h followed by 2 kHz for 45 min. ECE produces trees from 0.680 - 2.000 MeV ¹⁶O ions incident at angles of 45° (or greater) to the plane of the detector. Trajectories incident at 30° were not visualized. We take a threshold angle for ECE of 37.5°.

The efficiency of ECE in revealing ¹⁶O ion trajectories above a threshold angle is probably near unity. This value was deduced by the approximate agreement between the number of ECE trees and the number of CE tracks for the same ¹⁶O irradiation conditions.

After exposition, the two detectors of a sandwich array were separated and the CdS was removed with dilute HCl. One leaf of the array was then developed by ECE. The trees produced delimit regions in the facing detector where a recoil ion trajectory might be located. The facing detector was given the 2h CE to visualize any O ion tracks. Upon reuniting the detector array, coincident emission of two O ions would be visualized under the microscope as an ≈1 μm long CE track facing an ≈ 80 μm diam tree. Tests using ²³⁵U + n fission fragments showed that the shift upon remounting the detector array to look for spatial coincidences was usually less than 20 - 25 μm.

We could not unambiguously identify any spatial coincidences where a CE track in one detector lay within 100 μm of the center of an ECE tree in the facing detector. With the assumptions of a 100% detection efficiency within $\Delta\Omega = 61\%$ and a 95% confidence interval, the activity limit found for naturally abundant S, from 8 films of $25 \mu\text{g}/\text{cm}^2$, $\Delta T = 9 - 17$ weeks ; 2 films of $50 \mu\text{g}/\text{cm}^2$, $\Delta T = 21$ and 43 weeks ; 3.14 cm^2 visualized for each sandwich, is

$$A \leq 1.0 \times 10^{-4} \frac{\text{coinc}}{\text{week} \cdot \mu\text{g}} = 1.7 \times 10^{-10} \frac{\text{Bq}}{\mu\text{g}}.$$

With enriched S ($^{32}\text{S} : ^{33}\text{S} : ^{34}\text{S} : ^{36}\text{S} = 1.13\% : 1.01\% : 97.1\% : 0.72\%$), the activity limit from 9 films of $25 \mu\text{g}/\text{cm}^2$, $\Delta T = 9 - 15$ weeks ; 3.14 cm^2 visualized for each sandwich, is

$$A \leq 2.8 \times 10^{-4} \frac{\text{coinc}}{\text{week} \cdot \mu\text{g}} = 4.7 \times 10^{-10} \frac{\text{Bq}}{\mu\text{g}}.$$

These results confirm what one suspected already, namely that any break-up radioactivity from a primordial S isomer into heavy ions must be low. However, at the present time, there is no reason to alter our previous suggestion that the dwarf halos give evidence for an unrecognized symmetrical nuclear break-up. The conclusion of the present trials is that the dwarf halos themselves are probably the best indicators of a new radioactivity (rather than attempts at direct counting which have always been non-reproducible) because the mica is capable of integrating extremely low disintegration rates. From the

value of $10^8 - 10^9$ α -particles necessary to form a U halo,⁷ α -particle and O stopping powers,⁵ and inverse squares of radii, one calculates that $10^5 - 10^6$ O ion producing decays could form the smallest dwarf halos. With the age of $\sim 10^9$ yr^{2,8} for the mica in which these structures were found, one deduces that the mica can register an *average* decay rate as low as one disintegration per $10^3 - 10^4$ yr. The experiments described above would have to be scaled up considerably if one expects to detect any radioactivity in S using dielectric detectors. Other approaches would probably be more telling.

We would like to recognize the efforts of Mme A. Méens in the preparation of the CdS and its films, L. Oberlé for varied logistical aid, and B. Escoubès for discussions on the statistical treatment of low count rates.

References

1. A. Pape and M. Debeauvais, Centre de Recherches Nucléaires Report CRN 93-23, 1993.
2. J. Joly, *Nature* *114*, 160 (1924).
3. R.V. Gentry, *Science* *173*, 727 (1971).
4. R.L. Fleischer, P.B. Price, and R.M. Walker, *Nuclear Tracks in Solids* (Univ. California Press, Berkeley, 1975).
5. J.F. Ziegler, J.P. Biersack, and U. Littmark, in *The Stopping and Ranges of Ions in Matter*, edited by J.F. Ziegler (Pergamon Press, Oxford, 1985) Vol.1 and program TRIM.
6. L. Tommasino and C. Armellini, *Radiat. Eff.* *20*, 253 (1973).
7. R.V. Gentry, in *Annu. Rev. Nucl. Sci.*, edited by E. Segrè, J.R. Grover, and H.P. Noyes (Annual Reviews, Palo Alto, 1973), pp. 347-362.
8. R.V. Gentry (private communication).