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FURTHER ATTEMPTS TO ISOLATE SUPERHEAVY ELEMENTS IN THE METEORITE ALLENDE

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The search for superheavy elements /SHE/ in the carbonaceous chondrite Allende was continued. This time we carried out a thermochromatographic separation at 1050 °C in O_2-, resp., H_2-gas flow. The volatile fraction /mostly Pb/ was collected on a Pt-foil or trapped in KOH-solution. The heavy element probes were investigated for spontaneous fission activities. We observed zero spontaneous fission decays during 421 days. This yielded an upper limit of $\leq$2.9 fission x kg^{-1} x $year^{-1}$ /95% confidence limit/. We were unable to confirm the existence of a spontaneous fission activity in the meteorite Allende.

INTRODUCTION

During the past 15 years a wide variety of samples has been investigated for the occurrence of superheavy elements /SHE/ in nature. One of the samples investigated

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was the meteorite Allende. It was considered a very suitable probe, as this carbonaceous chondrite was formed during the early period of our solar system some 4.5×10^9 years ago; ever since it has remained unchanged until it fell onto the earth during 1968. Any confirmed observation of SHE in this meteorite would make it very likely, that SHE should also be present in some observable quantity on our earth. We do not want to discuss in this note the general aspects of looking for SHE in nature, as this was recently done by Kratz[1]. We want to concentrate on the experimental work studying the meteorite Allende.

Flerov et al.[2] presented evidence for the observation of SHE in the meteorite Allende. They used a neutron multiplicity counter, which investigates the bulk sample. They found an unknown spontaneous fission activity of about 1 fission/kg x month. Recently, this result has been confirmed by another group[3].

Furthermore, Zvara and coworkers[4] tried to isolate SHE as a volatile fraction from the meteorite Allende. They were able to isolate with thermochromatographic methods a volatile fraction, showing an unkonwn spontaneous fission activity of about 1/2 fission/kg x month. Continued work on this isolated volatile fraction showed that there remains some spontaneous fission activity in the samples[5]. However, its spontaneous fission activity had decreased to such an extent, that the discussion of the statistical significance of the results was difficult.

An independent attempt to isolate SHE from the meteorite Allende was started by Lund et al.[6] using wet-chemical methods. They separated from the bulk material those elements /Pb, Cd, etc./ which can be dissolved in strong acids and precipitated at pH $\simeq$ 1.5 with H_2S. No spontaneous fission activity was observed in the sulfide fraction /<1/20 fission/kg x month/. This result is obviously dependent on the chemical enrichment of SHE, which was applied, i.e. that SHE should have followed the sulfidic group element separation in aqueous solution.

Therefore, our chemical enrichment experiments have been repeated and modelled according to the procedures used by Zvara et al.[4] using their thermochromatographic method. It shall be noted, that it was not possible to use exactly the same procedure.

EXPERIMENTAL

A sample of 1804 g Allende meteorite was finely grained and divided into several portions. Then these portions were placed into our thermochromatographic apparatus, as shown in Fig. 1.

The material was filled in ceramic boats in layers of 5-6 mm thickness. Pre-experiments showed that after 2 h heating at 1050 ^{o}C the material was oxidized in O_2 /air/ flow, and successively reduced for 2 h in H_2 gas flow. During the same time Pb /as SHE-thermometer/ escaped quantitatively from the matrix.

The Allende meteorite was heated successively at 1050 ^{o}C in O_2 /air/ and H_2-gas flow for /in total/ 6-8 h. In each experiment we heated 25-100 g material. All material escaping the matrix was condensed on a cooled Pt-foil /150 $^{o}C < T < 1000$ ^{o}C/ or trapped in two 10% KOH-traps; an exception are, of course, the gaseous components.

After the completion of the heating procedure the KOH-solution was made acidic by adding HCl /pH $\sim$ 2/ and

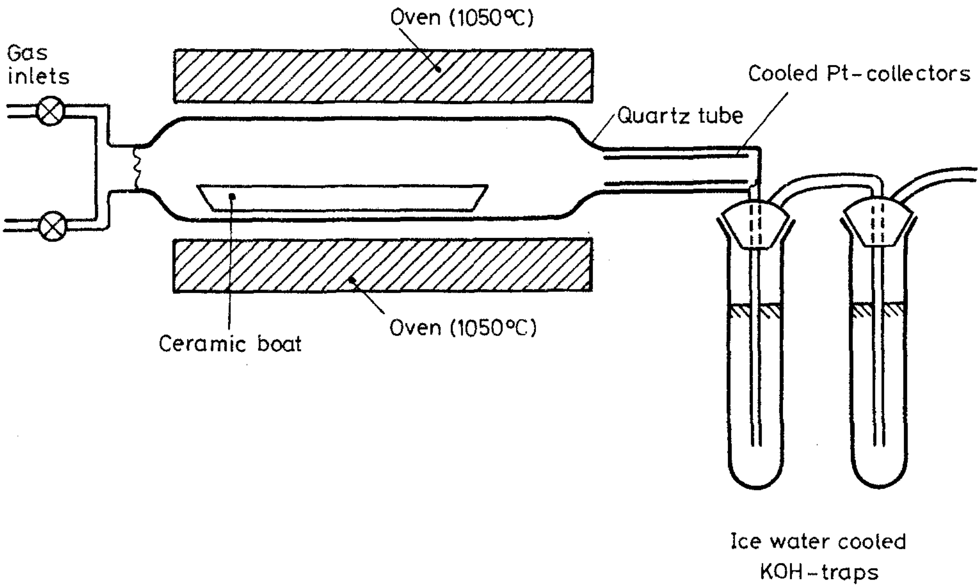

Fig. 1. Thermochromatographic column used to isolate SHE from Allende meteorite

the dissolved material was precipitated as sulfide. The sulfide precipitate was placed as a layer on mica.

Both the Pt-foil /Sample 1/ and the precipitate /Sample 2/ were examined for spontaneous fission events with Makrofol. The thickness of each sample was ≤ 1 mg cm^{-2}.

Makrofol /10 µm thickness/ is a plastic track detector, sensitive to fission fragments. In order to study the spontaneous fission activity of our sample, we employed the "spark-jump" technique. Here, a spontaneous fission event yields a visible hole in an aluminized plastic foil. This technique had been used by us previously in the investigation of a voletile fraction of Atlantis II-samples. Here also, we had looked for spontaneously fissioning SHE[7].

RESULTS AND DISCUSSION

At first, we studied our two samples in the laboratory for some time and obtained the following results:

Sample 1 /Pt-foil/: 1 event/88 days,
Sample 2 /Sulfides/: 0 events/46 days.

Such a low activity is to be expected in the laboratory due to cosmic-ray induced fission in heavy elements and it has also been observed previously[7]. Then we carried out the exposures in a tunnel under 25 m rock and found:

$$\text{Sample /1 and 2/: 0 events/421 days} \qquad /1/$$

The chemical yield for Pb is 90%, therefore, we assumed a chemical yield for SHE E_{chem} = 70%. According to Ref. 7, the counting efficiency for fission fragments with the Makrofol/"spark-jump" technique is ε_{det} = 0.7. We have calculated the spontaneous fission activity with ≤ 3 events, this gives a confidence level of 95% for zero events observed. The upper limit for a spontaneous fission activity $A_{S.F.}$ in Allende meteorite - chemically enriched by our method - is found to be:

$$A_{S.F.} \leq 2.9 \text{ fission/kg x year} \qquad /2/$$

It has been very difficult for the authors of Refs 2-5 to quote an exact spontaneous fission activity in their samples, including exactly formulated uncertainties. This is due to the very faint nature of the effect observed by those authors. It is interesting to note, that in Ref. 2. there is written: Taking into account the interval $4 < \nu < 10$, we obtain for the Allende samples 10-20 spontaneous fissions per year /i.e. per kg sample, note added by authors/.

Taking a value of 1 fission/kg x month, we should have seen 12 events with our technique. It is obvious, that in this work we find no evidence for the existence of SHE in the Allende meteorite. However, we are cautious to make any conclusions outside the stated limit of our measurements.

Finally, it shall be noted, that the same experimental data can be treated in two further ways:

1. Assuming a half-life of 10^9 years for SHE and a 100% spontaneous fission decay branch, we find /95% confidence level/ the following upper limit N /SHE/ for SHE-atoms:

$$N\ /SHE/ \leq 4\times10^{9}\ \text{atoms/kg.}$$

   In Ref. 2. is quoted a content of 6×10^9-6×10^{10} atoms per kg of material.

2. Assuming further a mass of 300 u for SHE we find /95% confidence level/ the following upper limit C /SHE/ for the concentration of SHE in Allende meteorite:

$$C\ /SHE/ \leq 2.5\times10^{-15}\ \text{g/g.}$$

In Ref. 2. is quoted a concentration of 3×10^{-15}-3×10^{-14} g/g for the various meteorites.

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