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Publication Date
1976-12-01

Submitted to Journal of Inorganic and
Nuclear Chemistry

LBL-5053
Preprint c.1

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December 1976

Prepared for the U. S. Energy Research and
Development Administration under Contract W-7405-ENG-48

DISCLAIMER

J. inorg. nucl. Chem., Supplement 1976. Pergamon Press. Printed in Great Britain

FURTHER SEARCH FOR SUPERHEAVY ELEMENTS IN NATURE WITH NEUTRON MULTIPLICITY SCINTILLATION COUNTER*

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Abstract—We have measured neutron multiplicities emitted by a large number of natural samples as an indication of the presence of superheavy elements. Such elements are expected to emit some 10 neutrons in their spontaneous fission compared to ≤4 for all known nuclides. We used a neutron multiplicity scintillation counter which had been placed in a tunnel under some 250 m of rock and dirt to minimize neutron multiplicities resulting from reactions of cosmic ray muons with heavy atomic nuclei. The samples included K, Cr, Fe, Ni, Cu, Zn, Sr, Ag, Ce, Pt, Au, Hg, Pb ores, ore concentrates, ore residues, as well as iron meteorites and some ultra basic rocks. We conclude that our results were negative within the accuracy of our measurements.

INTRODUCTION

THEORETICAL extrapolations[1, 2] suggest that there may be an "island of stability" with respect to all forms of nuclear decay in the atomic number Z range of about 110–124. These estimates involve the anticipated enhanced stability associated with the double-shell closure at $Z = 114$ and neutron number = 184. Further estimates indicate an average prompt neutron number/fission[1, 3] of $\simeq 10$ (for $Z \simeq 114$) compared to ≤ 4 for all known spontaneously fissioning nuclides. Superheavy elements (if they exist) or their daughters should decay primarily by spontaneous fission. Thus neutron multiplicity measurements should offer a good opportunity of definitive identification of superheavy elements in natural samples without the risk of chemical losses inherent in any chemical procedure involving unknown chemical properties.

Accordingly, we collected a large number of ores, ore concentrates, ore residues, flue dusts, etc. from Pb, Ag, Au, Zn mines in Colorado and from Cu, Ni, Pt mines in the Sudbury Region of Canada. We also collected ore samples from other parts of the United States and throughout the world. In addition, we examined some special samples, including some Ivigtut lead ore which has a relatively high ^{204}Pb content, indicating more primordial (compared to radiogenic) lead than most ores. All of these are described briefly in Tables 1a and b.

Some of these samples involved relatively large amounts of materials of higher Z. Such materials interact with the muon component of cosmic rays to give bursts of fission and spallation neutrons not distinguishable from spontaneous fission neutrons. The muons are highly penetrating and hence their flux decreases only slowly with shielding thickness.

The Lawrence Berkeley Laboratory neutron multiplicity scintillation counter was used in counting the above samples. It was located at that time in a new tunnel (not yet in use) of the (San Francisco) Bay Area Rapid Transit System under some 250 m rock and dirt. This afforded a muon background (and hence multiple neutron count) of some orders of magnitude less than at the surface of the ground for large samples of high Z.

EXPERIMENTAL

Normally, we put four 1·5 l. cans of sample in the 11·4 cm dia. × 105 cm high sample cavern of the counter. Because the time was limited that the counter would remain in the tunnel and because of manpower limitations, it was expedient to insert smaller amounts of up to four different materials at the same time in many cases (see Tables 1a, b and 2). The weights of samples were between 1 and 5 kg per can depending on the density and shapes of the sample material. During the measurement the counter was triggered either by a prompt γ-ray (in a fission or spallation reaction) or by gammas resulting from the first neutron captured by the scintillator. Then a gate was opened for 35 μsec and the number of additional events recorded during this time gave the multiplicity N. This time period was found (by using a ^{252}Cf source) to be sufficient to account for about 90 per cent of all neutrons captured[4]. The single neutron efficiency was found to be 60–70 per cent, depending on the self-shadowing of the sample itself and on the vertical position in the sample chamber of the main source of neutrons. A working efficiency of ~ 65 per cent was assumed.

*Research sponsored by the U.S. Atomic Energy Commission under contracts with the Union Carbide Corporation and the Lawrence Berkeley Laboratory.

Table 1. Samples examined for multiple neutron events

Symbol	Description	Mass (kg)
(a)		
IVIG. Pb	PbS ore from Ivigtut, Greenland	10·4
$PbCO_3$	Cerussite from New Mexico	13·0
$BaSO_4$	Barite from Sweetwater, Tennessee	7·1
$CeFCO_3$	Bastnasite from Mt. Pass, California	10·0
Cu Conc.	Cu concentrate from Sudbury Region, Canada	11·9
Ni Conc.	Ni concentrate from Sudbury Region, Canada	9·4
Fe Meteor.	Canyon Diablo Iron Meteorites	18·2
PM Ore	Ultra basic rock containing precious metals, Sudbury Region	8·4
The following five are samples of ultra basis rock resembling Kimberlite in composition, from the edge of Norris Lake, Tennessee:		
KIM-1	Rock which had been brought to the surface 20 years ago	9·1
KIM-2	Rock from a vertical shaft about 20 m deep	9·0
KIM-3	Rock from a vertical shaft over 12–20 m deep	10·5
K3, M1	KIM-3 plus magnetically separated magnetite from KIM-1	
K3, M12	KIM-3 plis magnetically separated magnetite from KIM-1 and -2	
(b)		
A	Composite sample containing precious metals, Sudbury Region	1·7
B	Pt, Pd bearing rock, Transvaal	2·7
C	Composite chromite, S. Rhodesia	3·8
D	Composite strontium ore, Hamm, Westphalia	3·0
E	Quartz containing Au, S. Dakota	2·7
F	Galena, Cour D'Alene Region, Idaho	3·5
G	Galena, Cour D'Alene Region, Idaho	3·5
H	Cinnebar, Arizona	1·8
I	Chromite + Zn conc., S. Rhodesia	2·1
J	Ni conc., Sudbury, Ontario	2·4
K	Ag, Cu, Pb conc., Creede, Colorado	3·2
L	PbS conc., Silverton, Colorado	4·3
M	PbS conc., Silverton, Colorado	4·5
N	Pb, Ag, Zn sulfide, Creede, Colorado	2·3
O	Pb, Ag, Au, Zn sulfide, Telluride, Colorado	4·1
P	Cu, Ag, Pb, Au, Zn conc., Telluride, Colorado	2·8
Q	Unburned bag house fume dust, Tooele, Utah	2·2
R	Serpentinite from mid-Atlantic ridge at equator	2·2
S	Pb, Zn sulfide ore, Creede, Colorado	2·1
T	Pb, Zn sulfide ore, Creede, Colorado	2·4
U	Blast furnace dust, Sudbury, Ontario	1·5
V	Flue dust, Intern. Nickel, Sudbury, Ontario	1·8
W	CuS, mill conc., Intern. Nickel, Sudbury	2·9
X	Blast furnace dust, Intern. Nickel, Sudbury	1·3
Y	Smelter furnace dust, Intern. Nickel, Sudbury	1·7
Z	KCl ore, Hobbs, New Mexico	2·1
AA	Zn, Pb, Cu, Ag, Au sulfide ore, Silverton, Colorado	3·7
AB	Pb, Ag, Fe, Zn sulfide ore, Creede, Colorado	4·0
AC	Cu, Pb, Zn sulfide ore, Silverton, Colorado	4·1
AD	Pb, Zn, Ag sulfide ore, Creede, Colorado	2·3
AE	PbS ore containing Ag, Au, Zn, Telluride, Colorado	3·3
AF	PbS Ore, Telluride, Colorado	3·8

The counter background results from γ-rays from natural sources (e.g. U, Th and K) and cosmic rays and the products of their interactions with matter. These γ-rays in general appear as single random pulses, and accidental coincidences between these pulses yield a distribution which may be represented by a Poisson probability function. This means that a plot of log $[N!C(N)]$ vs N gives a straight line[4]. In actual practice a straight line is obtained for $N = 1$, 2 and 3. At $N = 4$ the random coincidences are of the same order of magnitude as those due to fission or cosmic-ray reactions in matter. At $N \geq 5$ neutrons from the latter sources predominate.

RESULTS

The results are presented in Table 2. Here we give the sample composition (see Tables 1a and b); the mass in kg; the counting time in hr ; the observed count $C(4)$ for

Table 2. Results on samples listed in Tables 1a and b

Samples	Mass (kg)	Time (hr)	$C(4)$	4 Ran	$C(\geq 5)$	Normalized cts./(50 kg × 250 hr)
IVIG. Pb	10·4	211·0	36	32 ± 0·5	4	27 ± 38
$PbCO_3$	13·0	91·3	3	3·2 ± 0·0	1	(−9 ± 23)
$BaSO_4$	7·1	120·0	7	2·7 ± 0·0	1	46 ± 45
$CeFCO_3$	10·0	166·0	337	(595)	2	(−1950 ± 2470)
Cu Conc.	11·9	152·0	8	2·6 ± 0·0	1	25 ± 23
Ni Conc.	9·4	171·0	9	8 ± 20	0	(−14 ± 44)
Fe Meteor.	18·2	231·5	11	2·2 ± 1·0	1	17 ± 12
PM ore	8·4	137·1	13	10·2 ± 0·2	2	26 ± 44
KIM-1	9·1	193·8	26	16·2 ± 0·6	1	52 ± 39
KIM-2	9·0	170·8	12	11·4 ± 0·4	2	(−6 ± 30)
KIM-3	10·5	262·6	35	30 ± 0·5	4	20 ± 30
K3, M1	(12)	113·0	20	15·3 ± 0·1	1	34 ± 43
K3, M12	(13)	86·5	27	14·4 ± 0·1	2	145 ± 61
A, B, D, E	10·1	94·0	6	3·5 ± 0·1	1	24 ± 36
C, H, K, R	11·0	93·4	6	3·5 ± 0·1	1	23 ± 34
F, G, O, P	13·9	120·0	12	6·1 ± 0·0	0	29 ± 27
I, J, W, X	8·7	272·7	5	2·9 ± 1·2	0	(−15 ± 18)
L, M, S, T	13·3	69·0	8	4·0 ± 0·5	0	38 ± 41
Q, U, V, Y	7·2	99·0	7	5·2 ± 1·6	2	36 ± 59
AA, AC, AE, AF	14·9	315·2	37	23·2 ± 0·3	3	32 ± 19
AB, AD, N, Z	10·7	281·0	48	117 ± 3500	2	(−303 ± 250)

$N = 4$; the calculated random count (4 Ran) for $N = 4$; the observed count $C(\geq 5)$ for $N \geq 5$; and the "normalized" count for $N \geq 4$ after correcting for (4 Ran), the Empty Chamber Counts, and normalizing to a sample weight of 50 kg and a counting time of 250 h[4]. Thus

$$\text{"Normalized" count} = \left\{\frac{[C(4) - (4\,\text{Ran}) + C(\geq 5)] \times 250}{\text{Time (sample)}} - \frac{\text{Empty chamber count} \times 250}{\text{Time (empty)}}\right\} \frac{50}{\text{weight}}, \quad (1)$$

where

Empty chamber count $= [C(4) - (4\,\text{Ran}) + C(\geq 5)]$ for the empty chamber. We took the values from a previous paper for 844 total hours of counting [Ref. [4], Table 1]: $C(4) = 32$, $(4\,\text{Ran}) = 22$, $C(\geq 5) = 5 \pm 4$.

(4 Ran) was determined by fitting

$$\ln [N!\, C(N)] = p_1 + p_2 N \quad (2)$$

to the data for $N = 1$, 2 and 3 by the method of least squares on an IBM 360/91 computer, p_1 and p_2 being the parameters of fit, and then evaluating $(4\,\text{Ran}) = C(4)$ calculated from Eqn (2).

The parentheses around the masses of samples $K3$, $M1$ and $K3$, $M12$ indicate that the differences between these masses and that of KIM-3 are approximate. Parentheses were put around the value of (4 Ran) for $CeFCO_3$, because it is so much larger than the value of $C(4)$. We put parentheses around all negative values of "normalized" counts although all of these are zero within the statistical uncertainties.

There may be a slight hint of excessive higher multiplicities in some of the Tennessee "Kimberlites". This cannot be explained by the 4·5 ppm of uranium in the samples nor by the few per cent of iron. Elements of high Z are present in very small amounts. We feel that the statistical nature of a very low count plus the fact that the background changes with time are adequate to explain our observations. The apparent slightly high count in the samples AA, AC, AE, AF probably results from the high Z materials present. Thus we conclude that our results in Table 2 show no evidence for the presence of superheavy elements.

Acknowledgements—We wish to thank the directors of the Bay Area Rapid Transit System for allowing us the sole use of one of the cross passages inside their tunnel leading from Berkeley to Orinda; Dr. Carleton B. Moore, Director of the Center for Meteorite Studies, Arizona State University, for loaning us the Canyon Diablo iron meteorites; Dr. David E. Jensen, Vice-President of Ward's Natural Science Establishment, Inc., Rochester, N. Y., for loaning us the New Mexican cerussite; Drs. Norman N. Schiff and Paul R. Kruesi of the Molybdenum Corporation of America for the bastnasite ore; Mr. H. W. Miller of the Rocky Flats Division, Dow Chemical Co., Golden, Colorado, for a number of samples including the cinnebar ore from Arizona and the galena ore from The Cour D'Alene Region, Idaho; Dr. E. Skougaard Hansen of the Kryolitselskabet Oresund A/S, Copenhagen, for the sample of Ivigtut PbS ore; Dr. Stuart W. Maher, Chief Geologist, Tennessee Division of Geology (Knoxville) for telling us where to get the Tennessee "Kimberlite", barite and other samples; Mr. W. P. Bowen, General Manager of the National Lead Company's plant at Sweetwater, Tennessee, for giving us the barite and other samples; B. M. Benjamin, B. H. Ketelle, and F. E. McKinney of Oak Ridge National Laboratory and R. H. Ketelle of Oak Ridge for getting the Tennessee "Kimberlite" from a vertical shaft near the edge of Norris Lake.

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This report was done with support from the United States Energy Research and Development Administration. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the United States Energy Research and Development Administration.

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