Title
THE LEAP TO PRODUCE HEAVY NUCLEI AT THE LIMITS OF NUCLEAR STABILITY

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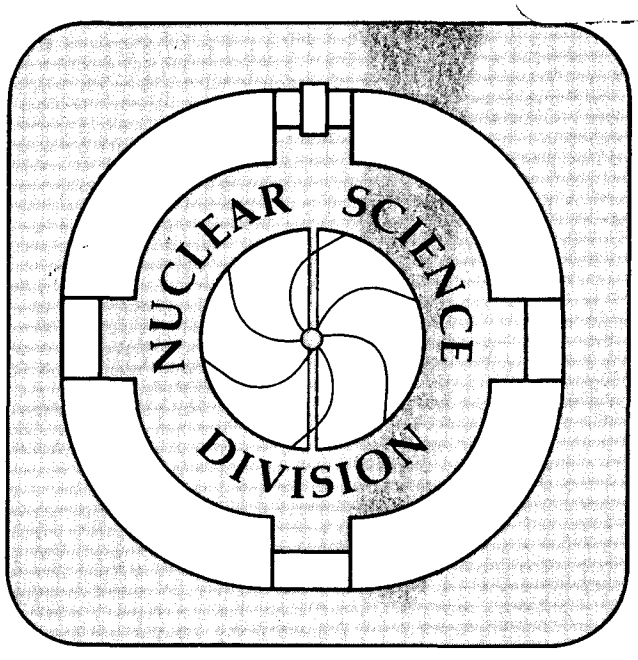

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THE LEAP TO PRODUCE HEAVY NUCLEI AT THE LIMITS OF NUCLEAR STABILITY

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Abstract

The Large Einsteinium Activation Program (LEAP) has been proposed by a consortium of four national laboratories. Central to the proposal is the preparation, for the first time, of a large target of Es-254 in order to accomplish a unique scientific program. Progress and activities to date and plans for the future will be discussed.

Introduction

The Large Einsteinium Activation Program (LEAP) was proposed [GHI84] in 1984 by four national laboratories--Lawrence Berkeley (LBL), Lawrence Livermore (LLNL), Los Alamos (LANL), and Oak Ridge (ORNL). This joint proposal was prepared in an effort to launch a major initiative to exploit the currently existing expertise in heavy element research and the unique potential for producing very heavy actinide target materials such as 285-day ^{254}Es at the High Flux Isotope Reactor (HFIR) at ORNL. It was also envisioned that major new techniques and instrumentation would be developed to enhance our ability to identify and study both chemical and nuclear properties of new neutron-rich heavy element isotopes at the limits of nuclear stability and even superheavy elements (SHE's). A workshop convened in 1983 by the National Research Council at the request of the U.S. Department of Energy had assessed the current status and future opportunities in research with transplutonium elements [BCS83].

In the area of nuclear research, the panel concluded that, "the exploration of the limits of nuclear stability is a prime motivation for studying nuclear species with the highest atomic numbers accessible. Improved understanding of nuclear-reaction mechanisms recently achieved gives great promise for reaching uncharted regions of nuclei at the upper end of and beyond the actinides, including presumably longer-lived isotopes of known elements than were previously available and possibly the long-sought superheavy elements of $Z \simeq 114$ with neutron numbers near 184."

In the area of chemical research, the panel concluded that, "From the chemist's point of view, the periodic table of elements is the most basic road map and to extend it to its farthest reaches is an obvious goal... Focus on the most basic chemistry for the transeinsteinium elements, including determination of properties of the metallic atoms, range and stability of oxidation states, ionic radii and complexation behavior, and simple binary molecular species." The LEAP proposal constituted an imaginative response to some of these challenges, a response which would expand, as well as utilize, our current capabilities.

The program proposes production of the largest ever target of ^{254}Es, at least 30 micrograms, which would permit preparation of a 400 $\mu g/cm^2$ target by depositing the ^{254}Es with a diameter of about 3 mm. The scientific goals of the LEAP are threefold: 1) to produce and identify neutron-rich isotopes of the heaviest elements by bombardment of the target with neutron-rich heavy ions in order to study nuclear properties, especially spontaneous fission, at the extreme limits of nuclear stability; 2) to prepare sufficient quantities of the heaviest actinides and transactinides for studies of their chemistry; 3) to produce superheavy

elements by irradiation of the Es-254 target with Ca-48 which gives a compound nucleus whose neutron number of 183 is nearer the predicted closed shell at 184 than has previously been achieved. These studies require access to an accelerator which can provide stable and relatively high intensities of neutron-rich light heavy ion beams such as ^{18}O, ^{22}Ne, and ^{48}Ca. These can be provided at the 88-Inch Cyclotron and SuperHILAC at LBL.

Feasibility studies [BIG85] conducted at the HFIR at ORNL indicate that production of the ^{254}Es target material should be possible. The starting material will be up to 1 gram of ^{252}Cf which will be irradiated in the HFIR for 21 days in the standard configuration. After a 20-day cooling period to allow ^{253}Cf (T 1/2 = 17.8d) to beta decay to ^{253}Es, the material will be placed in a cadmium sheath to reduce the thermal neutron flux and reirradiated. The reduction of the thermal neutron flux minimizes prompt fission of the ^{254}Es which is produced without significantly reducing the neutron capture cross section for ^{253}Es and tens of micrograms of ^{254}Es should be produced. A schematic for the production bombardment is shown in Figure 1. A cooling period of several months will allow decay of 20.5-day ^{253}Es. At equal activities of ^{254}Es and ^{253}Es, the sample will contain about 3 x 10^{11} alpha decays per minute in 30 µg. Shielding from the alpha activity is relatively easy, but unfortunately the 3-hour ^{250}Bk daughter has ≈1 MeV gamma rays which will give about 10R/h at 5 cm.

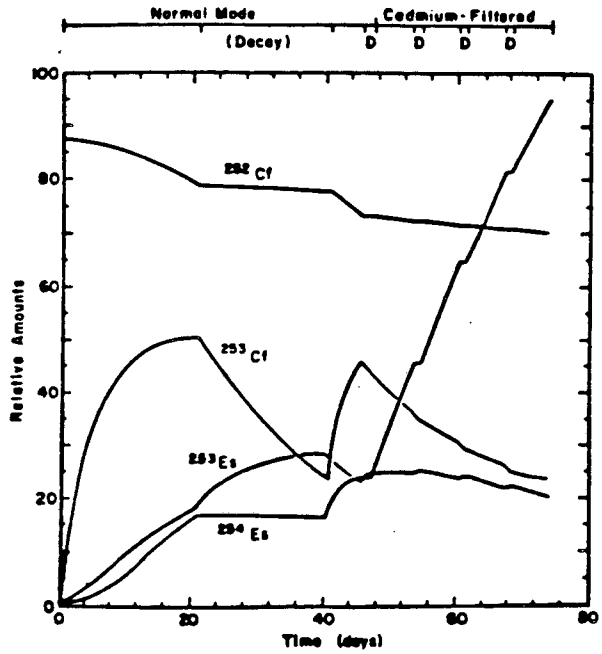

Fig. 1. Changes of Composition During Irradiation of ^{252}Cf to Produce ^{254}Es.

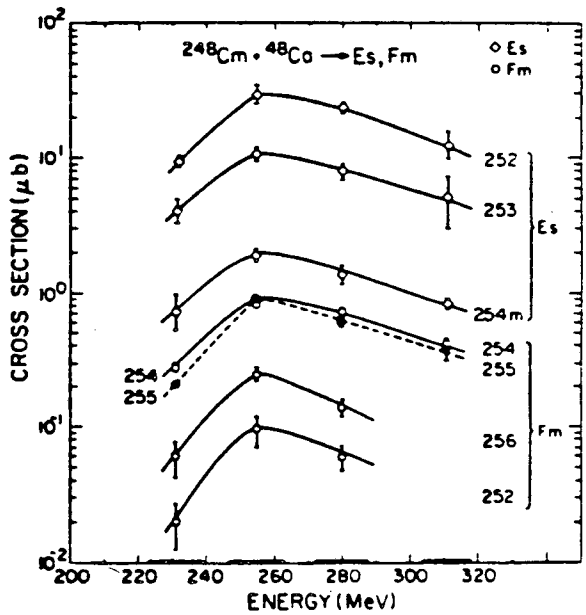

Fig. 2. Excitation Functions for Es and Fm Isotopes [HOF85.]

Production of Heavy Element Isotopes

In order to study the chemical or nuclear properties of the heaviest elements we must first devise suitable production reactions. Traditionally, compound nucleus reactions involving complete fusion of projectile and target nuclei, followed by particle emission have been used. However, because of the high excitation energies of these compound nuclei and their

high fissionability, losses due to prompt fission and neutron emission are excessive. Furthermore, it is difficult to produce the neutron-rich heavy element isotopes which we predict may have longer half lives. Fortunately, recent studies [LEE82, 83; ESK84; HOF84] have shown that binary transfer reactions between neutron-rich, light heavy-ion projectiles and neutron-rich actinide targets can be used to produce neutron-rich products of elements up to four protons heavier than the target in relatively high yields. Because the Q values for these reactions are negative, the desired products can be produced with relatively low excitation energy and the excitation functions are quite broad as shown in Fig. 2 for production of Es and Fm isotopes from reactions of ^{48}Ca projectiles with ^{248}Cm [HOF85].

Comparison of effective transfer of Be fragments for several different systems indicates comparable cross sections for these rather diverse systems. Based on systematics of this type and assuming similar cross sections for transfer of the same fragments to ^{254}Es, we estimate the production rates shown in Table I for isotopes of interest for chemical studies. Hulet et al. [HUL85] have now measured production cross sections for many of these isotopes and indeed the comparison for ^{254}Es and ^{248}Cm targets given in Table II shows that much large quantities of these actinide isotopes can be produced using ^{254}Es with ^{22}Ne projectiles. Currently, we are trying to measure cross sections for Z = 5 and 6 transfers to lighter actinide targets in order to predict what they may be for ^{254}Es.

Table I. Production Rates for Chemical Studies (400 μg/cm^2 ^{254}Es, 1p μA beam).

NUCLIDE	REACTION	$T_{\frac{1}{2}}$	ATOMS/MIN.
^{256}Md	(^{18}O or ^{22}Ne)T	1.3 h	8 x 10^5
^{259}Md	"	1.6 h	4 x 10^4
^{259}No	(^{22}Ne)T	58 m	2 x 10^3
^{260}Lr	"	3 m	3 x 10^2
^{261}Rf	(^{11}B, 4n)	1 m	(20)
^{262}Ha	(^{12}C, 4n)	34 s	(8)

Table II. Ratio of Yields for ^{254}Es Relative to ^{248}Cm Target.

		Transfer
Fm: (254)	$\frac{3 \times 10^3 \mu b}{3 \mu b} \approx 10^3$	p vs. Be
Md: (256)	$\frac{10^3 \mu b}{.01} \approx 10^5$	He vs. B
No: (259)	$\frac{4\text{-}6 \mu b}{<.03 \mu b} \gtrsim 10^2$	Li vs. C (or α, 3n)
	(only 1 pt.)	
Lr: (259,260)	1μb	Be vs. N

Chemical Studies

Studies of the chemical properties of the heaviest elements are of particular interest because of the strong relativistic effects caused by the electric field resulting from the high nuclear charge. For example, calculations [DES80] show that because of relativistic stabilization of the p orbitals the electronic configuration for Lr (Z = 103) could be $5f^{14}$ $7s^2$ $7p_{1/2}$ rather than the $5f^{14}$ $6d^1$ $7s^2$ expected on the basis of simple extrapolation from the lighter actinides. Lr might then have a stable +1 oxidation state, although earlier studies [SIL70] did show its extraction behavior was consistent with that of a trivalent actinide. We have installed a helium jet transport system at the 88-Inch Cyclotron at LBL and have successfully transported reaction products recoiling out of the target and attached to KCl aerosols to a chemistry laboratory some 80 meters

away. The products are collected on aluminum foil, picked up in water and adsorbed on a tiny cation exchange resin column to investigate the elution position of 3- min. ^{260}Lr by elution with ammonium alpha-hydroxyisobutyrate. From the position of Lr relative to added rare earth tracers, the ionic radius can be deduced. A small ^{254}Es target (20μg/cm^2) is being used and it is obvious from the low production rates that the large target will be required for more extensive studies. Plans to use other techniques such as thermochromatography, preparation of volatile fluorides of the transactinides using the powerful oxidizing and fluorinating agents FOOF and KrF_2, continuous liquid-liquid extractions, and various other systems to study the chemical properties of the transactinides are also being made. Separations based on the difference in chemical properties between actinides and transactinides will also be useful for separations prior to studies of nuclear properties.

Nuclear Properties

The use of a large ^{254}Es target (400μg/Cm^2) and transfer reactions from neutron-rich light heavy ions ($Z \leq 20$) should allow production of a large number of new neutron-rich heavy element isotopes for study. Those predicted to be produced with cross sections greater than a nanobarn are shown as cross-hatched areas in Fig. 3. Compound nuclei for reactions of ^{18}O with ^{248}Cm, ^{252}Cf and ^{254}Es are given in parenthesis.

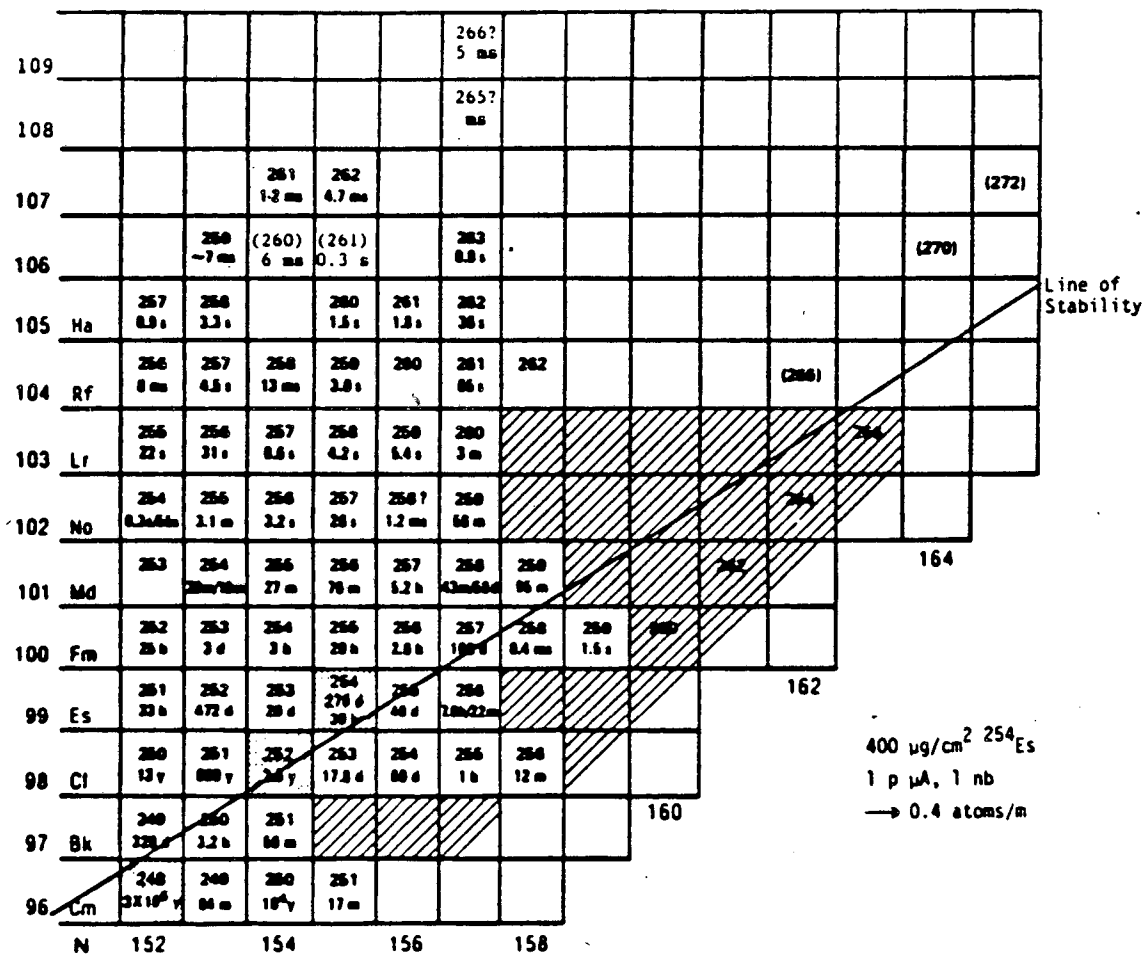

Fig. 3. Portion of table of isotopes showing isotopes potentially produced from light heavy ion reactions with ^{254}Es.

Estimated production rates and half lives for some of these isotopes are given in Table III. Spontaneous fission (SF) will ultimately limit the production of the heavy element isotopes and the half lives for SF can still not be predicted accurately, especially for nuclides containing an odd proton or neutron. It has been shown [RAN73] that the odd particle can greatly inhibit SF. Hindrance factors, particularly for high spin particles, can be as high as 10^7 as shown in Fig. 4. Half lives for odd-odd isotopes are expected to be especially hindered.

Table III. Estimated Production Rates for New N-Rich Isotopes (400 $\mu g/cm^2$ ^{254}Es, 1 pμA beam).

NUCLIDE	ESTIMATED $T_{1/2}$	REACTION	RATE ATOMS/m
Md-260	1-10h β	(^{6}He)T	800
261	2-200 μs, SF	(^{7}He)T	80
262	≈0.2 s, SF	(^{8}He)T	8
No-260	< ps SF	(^{6}Li)T	1000
261	1.1 h α	(^{7}Li)T	80
Lr-261	5 m α	(^{7}Be)T	400
262	14 m α	(^{8}Be)T	120
263	0.7 h α	(^{9}Be)T	20
264	2.3 h α	(^{10}Be)T	8
Rf-262	ms, SF	(^{8}B)T	4
263	1.5 m α	(^{9}B)T	4
Ha-263	0.6 s α	(^{12}C, 3n)	4
264	4 s α	(^{10}C)T	0.4

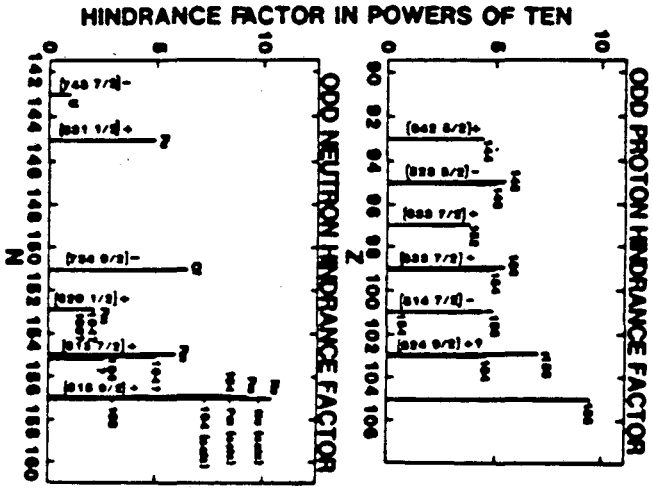

Fig. 4. Odd Proton and Neutron Hindrance Factors

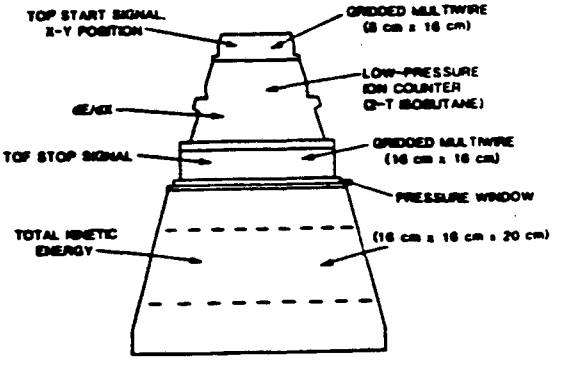

Fig. 5. Detector Module to be used in Heavy Element Fission Tracker (HEFT) System

Recently, ^{260}Md, which has both an odd proton and neutron, has been produced [HUL85] and shown to have an SF half life of at least 30 days, confirming this extra hindrance. This makes it hopeful that longer-lived isotopes of No(102) and especially of Lr(103) can be produced. Studies of their SF properties will be particularly interesting since SF properties change very rapidly in the region of Z = 100 and N = 158. The most probable mass division is symmetric in this region and unusually high total kinetic energies which approach the Q value for fission have been observed. There is also some indication of two different kinds of symmetric mass division, one with high and one with "normal" total kinetic energy. Production of longer-lived isotopes of these elements should also make studies of chemical properties much more viable if the cross sections are large enough.

One of the problems with transfer reactions is that they are not specific and a wide variety of products results. Identification of the isotopes which spontaneously fission is especially difficult since the fission process rather effectively destroys the information on the Z and A of the fissioning species. A new detector system, the Heavy Element Fission Tracker (HEFT) is being developed to measure half lives as short as nanoseconds and simultaneously identify the fissioning isotope. It consists of an array of modules each consisting of four elements (Fig. 5) to characterize the energy deposited by the fission fragment. The first records the x-y position of the fragment and provides a start signal. The second gives the energy loss. The third again provides an x-y position and a timing signal. The fourth element stops the fragment and gives information concerning its residual kinetic energy. The mass of the fission fragment is then determined from the kinetic energy and velocity and the atomic number from the rate of energy loss. An array of perhaps five pairs of these modules would permit efficient detection of coincident fission fragments. By summing information on the mass and charge of coincident fragments, it should be possible to make definitive assignments from some 10 to 100 recorded events.

Superheavy Elements

Use of the ^{254}Es target and ^{48}Ca projectiles would give a compound nucleus with 183 neutrons, closer to the postulated stable shell at 184 neutrons than has previously been achieved. In addition to fast chemical separation techniques, SASSY-II, a higher efficiency version of the Small Angle Separating System will be used to search for Superheavy Elements (SHE's) formed in such compound nucleus reactions. Half lives as short as microseconds can be detected with crude mass resolution. Final identification will depend on observation of alpha decay chains resulting in previously identified nuclides.

Summary

Preliminary experiments indicate that the proposed large amount of ^{254}Es can be produced. Experiments with small ^{254}Es targets show production of much larger amounts of trans-Es isotopes than is possible with ^{248}Cm, but the large target will be required for studies of chemical properties. It appears hopeful that there will be longer-lived, neutron-rich isotopes with odd proton and neutron number. New instrumentation for identification and study of new SF isotopes and SHE's is being developed.

References

[BCS83] Board of Chemical Sciences, National Research Council, National Academy Press, Washington, D.C., 1983, "Opportunities and Challenges in Research with Transplutonium Elements."

[BIG85] J.E. Bigelow, C.W. Alexander, and L.J. King, Am. Inst. Chem. Eng. Summer Natl. Meeting, Aug. 25-28, 1985, Seattle, Wash. Paper 28B.

[DES80] J.P. Desclaux and B. Fricke, J. Physique 41, 943 (1980).

[ESK84] K. Eskola, P. Eskola, M.M. Fowler, H. Ohm, E.N. Treher, J.B. Wilhelmy, D. Lee, and G.T. Seaborg, Phys. Rev. C 29, 1160 (1984).

[GHI84] A. Ghiorso, D.C. Hoffman, E.K. Hulet, O.L. Keller, and G.T. Seaborg, Lawrence Berkeley Laboratory Report PUB-5118 (1984).

[HOF84] D.C. Hoffman, Accts. Chem. Res. 17, 235 (1984).

[HOF85] D.C. Hoffman, et al., Phys. Rev. C 31, 1763 (1985).

[HUL85] E.K. Hulet, private communication.

[KEL84] O.L. Keller, Jr., D.C. Hoffman, R.A. Penneman, and G.R. Choppin, Physics Today, March 1984, p. 35.

[LEE82] D. Lee, H. von Gunten, B. Jacak, M. Nurmia, Y.-f. Liu, C. Luo, T.T. Seaborg, and D.C. Hoffman, Phys. Rev. C 25, 296 (1982).

[LEE83] D. Lee, K.J. Moody, M.J. Nurmia, G.T. Seaborg, H.R. von Gunten, and D.C. Hoffman, Phys. Rev. C27, 2656 (1983).

[RAN73] J. Randrup et al., Nucl. Phys. A 217, 221 (1973).

[SCH84] M. Schädel et al., LLNL, Nuclear Chem. Div. FY84 Annual Report, UCAR 10062-84/1, p. 6-26.

[SIL70] R. Silva, T. Sikkeland, M. Nurmia, and A. Ghiorso, Nucl. Chem. Letters 6, 733 (1970).

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