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PROSPECTS FOR BARYON INSTABILITY SEARCH WITH LONG-LIVED ISOTOPES

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ABSTRACT

In this paper we consider the possibility of observation of baryon instability processes occurring inside nuclei by searching for the remnants of such processes that could have been accumulated in nature as rare long-lived isotopes. As an example, we discuss here the possible detection of traces of ^{97}Tc, ^{98}Tc, and ^{99}Tc in deep-mined non-radioactive tin ores.

Introduction

The possible experimental observation of baryon instability remains among the most fundamental problems of modern physics [1]. The observed "baryon asymmetry" of the Universe [2] and some Unification Theories including Supersymmetric Models [3,4,5] require either nucleon-decay (with baryon number change by $\Delta B=1$) or neutron-antineutron transitions (with baryon number change by $\Delta B=2$) with rates stretching the limits of present experimental capabilities. Future prospects for proton decay and neutron-antineutron transition searches by traditional detection techniques have been reviewed in a number of contributions to this Workshop [1,6,7,8,9]. These techniques are based, in general, on the direct detection in real time of the final products of baryon transformation leading to the disappearance of one or two nucleons inside the nuclei corresponding to $\Delta B=1$ and $\Delta B=2$, respectively.

In 1975 a geochemical approach to the search for baryon instability was proposed by Rosen [10]. This method is based on the observation of residual nuclei (noble gases) which are the remnants of intranuclear nucleon decay accumulated in ores during a period of time that is comparable to the age of earth crust. Later Evans and Steinberg [11], using existing data [12] on the concentrations of stable isotopes of xenon in a $\sim 2.5 \cdot 10^9$ year-old telluride ore and comparing these with the abundance of xenon isotopes in the atmosphere, obtained a lower limit for the nucleon life time of $1.6 \cdot 10^{25}$ years. The result is based on the assumption that the excess of ^{129}Xe concentration in the ore (as compared to different Xe-isotopes in the ore and in the atmosphere) can be attributed to proton decay. Although this limit is a few orders of magnitude lower than the limits obtained from exclusive nucleon decay modes in real-time decay experiments [1,6] it is often interpreted in the literature as being independent of the nucleon decay mode [13]. We believe that this is a misinterpretation of the results of Ref. 11. These results are applicable only to certain particular plausible nucleon decay modes which do not have hadrons in the final state such

as $p \rightarrow e^+ 6\nu$, $p \rightarrow e^+ \gamma$, $n \rightarrow 3\nu$, $n \rightarrow \gamma\nu$, etc. The results may also be related to other nucleon decay modes after a mode-dependent correction factor is applied. The correction factor can be obtained via nuclear theory calculations of the type described in Ref. 14 which take into account transmutation of nuclides resulting from interactions of nucleon decay products with daughter nuclei. Evans and Steinberg also pointed out in [11] that: "The possibility of further large improvement of the nucleon lifetime limit using geochemical techniques would seem unlikely because of uncertainties in the estimation of background effects over geological timespans."

New approach with long-lived isotopes

The new approach we discuss here relies on the observation of traces of rare long-lived nuclides in ancient ores which are remnants of intranuclear baryon-number-violating processes. The life time of such nuclides should be large enough to allow accumulation of the baryon-number-violating signal and small enough (as compared to the age of the ore) to exclude the primordial abundance of these nuclides. Nuclides with a life time in the region of $\sim 10^6$ years are good candidates for such a search. This life time region is also appropriate for the reliable reconstruction of the geological history (stability) of the initial ore. The ore should, preferably, include no traces of fissionable or radioactive elements and should be obtained from a large burial depth in order to avoid cosmic-particle-induced background yield of the nuclides of interest. This ore should be mined in as large amounts as possible and should allow simple processing for the extraction of the nuclides of interest. We assume that chemical extraction can be used for the initial separation of the base element and for concentration of the residues. Considerable progress made in laser selective photoionization spectroscopy during the last decade [15] should allow, in a final step, the detection of single atoms of chosen isotopes with a typical efficiency ~ 1% in milligram samples. For technetium isotopes a detection limit of $5 \cdot 10^6$ atoms and an efficiency of 0.4% have already been demonstrated [16]. The detection of single atoms of francium with a typical efficiency of ~ 1% was achieved in [17]. These examples suggest that a detection limit for technetium at the level of a few hundred atoms would be possible.

We have identified a parent-daughter combination of nuclides which represents a transmutation which is favorable from the point of view of these requirements. This is a transmutation of tin into three isotopes of technetium: ^{97}Tc with (half-life time) $\tau_{97} = 2.6 \cdot 10^6$ years; ^{98}Tc with $\tau_{98} = 4.2 \cdot 10^6$ years; and ^{99}Tc with $\tau_{99} = 2.1 \cdot 10^5$ years. The probability of a parent-daughter transmutation with $\Delta A \sim 20$ and $\Delta Z = 7$ is determined by final-state interactions [14], i.e. it is decay-mode dependent. Since none of the technetium isotopes exist in the nature (technetium was discovered in the remnants of cyclotron bombarded molybdenum [18]) its chemical extraction will not be obscured by the presence of stable isotopes.

Assuming that the initial extraction of the technetium can be performed on a large scale as a by-process in the mining industry [18], and assuming that background processes, which will be discussed later, are favorable, one can expect detectable amounts of technetium to signal the baryon instability at the level of $\sim 10^{33}$–10^{34} years. Although different decay modes will have different transmutation efficiencies, the limit obtained by

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this method will not permit identification of the particular decay mode involved or even to distinguish between $\Delta B=1$ and $\Delta B=2$ processes. Intranuclear neutron-antineutron transformations ($\Delta B=2$) in Sn with multi-pion final states will have nuclide transmutation efficiencies spread over a wide range of ΔA and ΔZ (Figure 1). Our choice of parent-daughter nuclides provides a higher sensitivity to $\Delta B=2$ processes than to those modes of nucleon decay with $\Delta B=1$ where ΔA and ΔZ values are expected to be lower.

A large ΔA gap between selected parent and daughter nuclides should be favorable from the point of view of suppression of background processes induced by low-energy cosmic rays, solar neutrinos, and neutrons and alpha particles from natural radioactivity since all of these processes result in smaller transmutations of the initial nuclide. However, interactions of high-energy muons, as discussed below, can, in some instances, produce rather large ΔA-transmutations.

Selection of tin (Sn) as a parent nuclide has some advantages. Tin is an abundant element with several stable isotopes with Z=50 and with an average mass of A=118.7. In nature the major source of tin is the mineral cassiterite (SnO_2). It consists of dense (~7 g/cm^3) hard granules. The world annual production of SnO_2 is $1.6 \cdot 10^5$ tons. The major deep mining areas for cassiterite are in Bolivia, Peru, Australia and England. Some of the cassiterites are found at a depth of more than a mile. As was pointed out in [18] it is likely that the industrial extraction scheme of tin from cassiterite can be modified in a simple and inexpensive way to allow the extraction of technetium with high efficiency from large quantities of tin ore as a by-process.

To estimate the yield of technetium isotopes we have calculated the spectrum of final-state nuclides resulting from neutron-antineutron transformations in tin nuclei followed by intranuclear cascades (the latter is described in Ref. 19). The results of these calculations are shown in Figure 1. The sizes of the boxes in this figure are proportional to the probability of the specific (A, Z) isotope production. Filled boxes correspond to the stable isotopes, and hatched boxes to long-lived isotopes of technetium. The arrows show the direction of natural radioactive transformations between nuclides. It is important to notice that isotopes with mass number of 97 and Z higher than 43 are unstable and have a relatively short life time. As a result of a chain of successive decays these isotopes are transmutated to ^{97}Tc and, therefore, essentially enhance its cumulative branching (ξ) to a total of 3.07 %. For the other two Tc isotopes the situation is not as favorable because the existance of stable isotopes of ruthenium (Z=44) terminates the corresponding decay chains earlier. Thus, the branching for ^{98}Tc is 0.56% and for ^{99}Tc it is 0.38%.

To estimate the sensitivity of our approach let us consider, for example, the extraction of the ^{97}Tc isotope from an initial sample of ore containing 1 ton of tin (N_n number of neutrons). We will ignore, for the time being, the processes which might lead to the background production of technetium in the ore. These processes will be considered in the next section. We will assume that the detection of single atoms by laser selective photoionization spectroscopy methods can be performed with an efficiency of 1% in the absence of a spurious background. This assumption corresponds rather closely to the presently-achieved level of detection efficiency [16, 17] but does require further refinement of the laser selective photoionization technique. Thus, assuming that the detection limit in laser selective photoionization spectroscopy (n_{min}) can be as low as 100

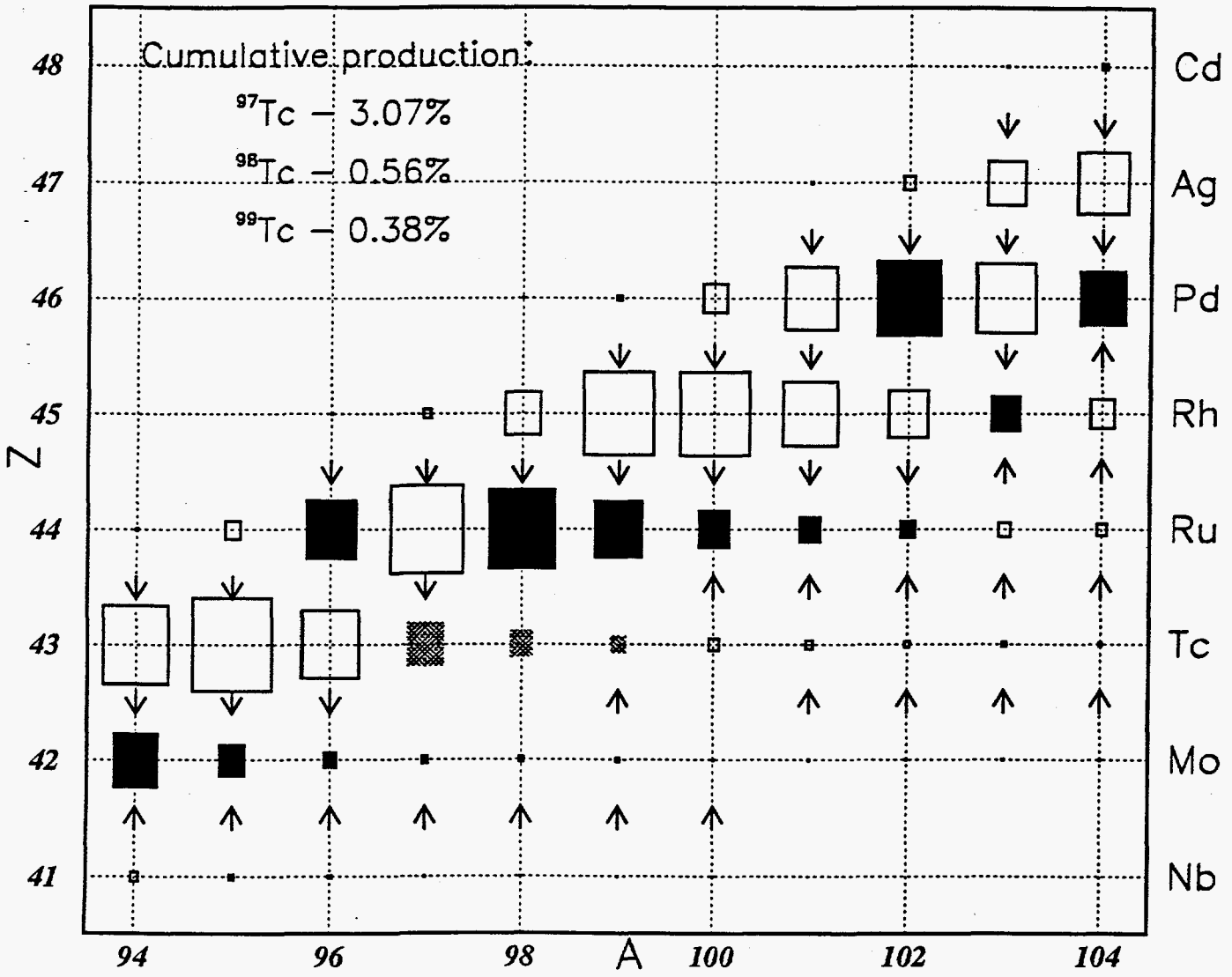

Fig 1. Yields of isotopes as a function of A and Z resulting from intranuclear neutron-antineutron transitions in tin, as calculated from the model of Ref. [19]. Open boxes indicate unstable isotopes, black boxes indicate stable isotopes, and hatched boxes indicate long lived technetium isotopes. The arrows show the direction of natural ß-decay. The sizes of boxes are proportional to the calculated isotope yields.

atoms, we can estimate the characteristic time for the case of neutron-antineutron transitions (^{97}Tc branching $\xi_{97} = 0.0307$) from:

$$\tau_{n-\bar{n}} = \frac{N_n \cdot \xi_{97} \cdot \tau_{97}}{n_{min}} = \frac{6.022 \cdot 10^{23} \cdot 68.69/118.69 \cdot 10^6 \cdot 0.0307 \cdot 2.6 \cdot 10^6}{100} \cong 2.8 \cdot 10^{32} (\text{years / ton})$$

Thus, the processing of an initial amount of ore corresponding to 10^3–10^5 tons of tin could, potentially, result (in case of favorable background) in a neutron-antineutron intranuclear transition limit of 10^{33}–10^{34} years. For comparison, the limit of 10^{33} years for neutron-antineutron intranuclear transitions can be obtained from a five-year operation of the Super-Kamiokande detector which has a total active mass of 50,000 tons [6].

In practice, the experiment to search for technetium in tin ore would consist of two major stages: (a) efficient processing of a large amount of ore from deeply buried deposits including chemical separation of tin and concentration of technetium to a milligram-level sample, and (b) determination the concentration of technetium isotopes in the sample by laser selective photoionization spectroscopy methods.

Background

Neutron-antineutron transitions or nucleon decay are not the only mechanisms that might contribute to the production of isotopes of technetium in the tin ore. The following sources of background have to be considered: (a) natural radioactivity; (b) interaction of cosmic rays (mainly muons) with tin nuclei; (c) interaction of cosmic or solar neutrinos with possible admixtures of molybdenum present in the tin ore; and (d) technetium contamination during ore processing and sample analysis. We consider each in turn below.

(a) Natural radioactivity. If some amount of uranium is present in the ore, a background concentration of technetium will be produced due to spontaneous and/or induced fission processes. Nuclides with atomic masses of 90-100 are positioned at the maximum of the mass-yield curves for ^{235}U and ^{238}U fission, and have the yields of about 6%. It is known that primary fission fragments have the most probable Z-values that are lower than the Z-values for the stable isotopes of the same mass. As a result, the primary fission fragment yield of technetium is rather low. However, for nuclei with A=99, all isotopes with charge less then 43 are unstable with quite a short life time. They result in the chain of fast successive decays which lead to an accumulation of ^{99}Tc at the level of ~ 6% of fission products. For ^{98}Tc and ^{97}Tc isotopes the situation is quite different. Stable isotopes of ^{98}Mo, and ^{97}Mo terminate the decay chains and are the end products of unstable isotopes with lower Z which have been produced as primary fission fragments. As a result, the yield of ^{98}Tc and ^{97}Tc in uranium fission is the result of primary fission only. Unfortunately, there is only limited quantitative experimental information in the literature on the production of ^{98}Tc by fission and no data at all for ^{97}Tc.

In order to estimate the yield of ^{98}Tc and ^{97}Tc isotopes we used the usual Z-parametrization of the fission-fragment distribution [20] in the form $P(Z)=(c\pi)^{-1/2}\cdot\exp[-(Z-Z_P)^2/c]$ where parameter c is related to the width of the distribution ($c \sim 0.80 \pm 0.14$) and Z_P is the most probable value of Z of fragments with a given mass number. In figure 2a the

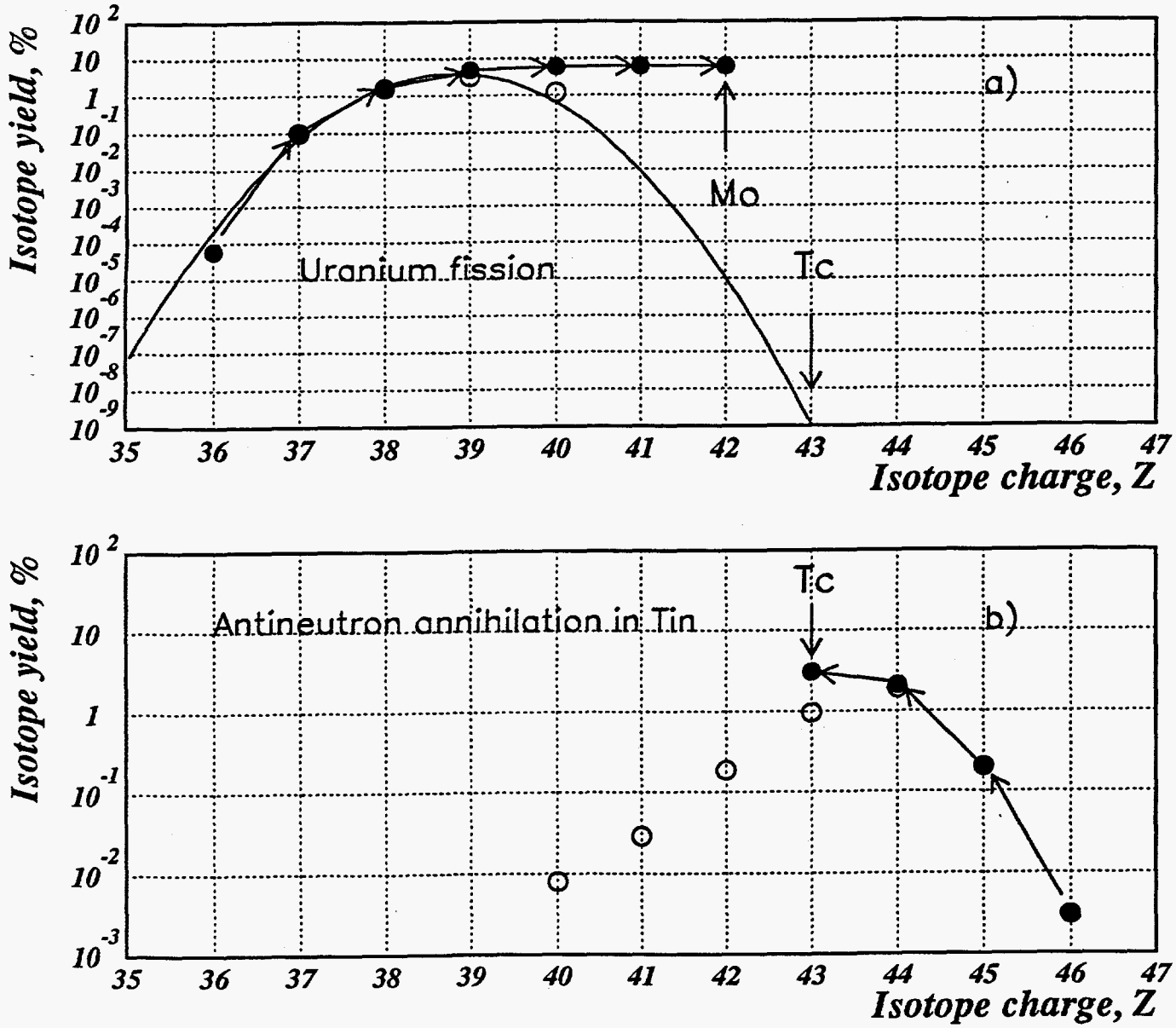

Fig 2. Cumulative yields of isotopes with mass number 97 produced by a) uranium fission [20], and b) antineutron annihilation in tin [19]. Open circles are for primary-produced isotopes; solid circles are for cumulative yields.

cumulative yield of isotopes (primary + decay) with mass number 97 produced by uranium fission is shown by the filled circles. The open circles in the figure represent the experimental data for primary-produced isotopes. The curve depicts the parametrization function with parameters c=0.8 and Z_p=38.8. Using this and similar parametrization for mass 98, we obtained the yield per fission event of $1 \cdot 10^{-11}$ for ^{97}Tc and of $7 \cdot 10^{-10}$ for ^{98}Tc isotopes. Shown in figure 2b is the yield of nuclides with mass 97 resulting from assumed neutron-antineutron intranuclear transformations in a natural mix of tin isotopes as calculated [19] from the model mentioned earlier [14]. Open circles are for primary-produced isotopes; solid circles and arrows indicate cumulatative yields.

Typical uranium concentrations in the earth crust are of the order of one ppm [21] and the uranium concentration in deeply buried tin ores has not been measured. The spontaneous fission life time of ^{238}U is $8 \cdot 10^{15}$ years. For ore with 1 ppm uranium concentration (by number) this should result in technetium concentrations of ~ 17 atoms of ^{97}Tc, 1,880 atoms of ^{98}Tc, and $8 \cdot 10^9$ atoms of ^{99}Tc isotopes per ton of tin. If the concentration of uranium in the initial material is known (the rate of induced uranium fission can be estimated from the known concentration of uranium and from the measured neutron flux in the deposit) the corresponding uranium background contributions can be calculated and subtracted from the measured amounts of technetium isotopes. Simultaneous measurement of all three technetium isotopes should provide additional constraints on the subtraction of the contributions of uranium fission, both spontaneous and induced. This would, in turn, require that the yields of ^{98}Tc and ^{97}Tc isotopes per uranium fission be determined experimentally with an appropriate accuracy. In Table 1 below we show estimated yields of Tc isotopes due to spontaneous uranium fission for two uranium concentrations of 1 ppm and 10 ppb representing pessimistic and optimistic levels.

(b) Cosmic rays. The main component of cosmic rays that penetrate to large underground depths consist of muons. The flux of muons is quickly attenuated with depth. After the first 500 meters of water equivalent it is ~ 1 muon$\cdot$m$^{-2}\cdot$s^{-1}. At a depth of ~ 4 km of water equivalent (~1 mile of rock), it is reduced to ~ $5.5 \cdot 10^{-5}$ muon$\cdot$m$^{-2}\cdot$s^{-1} [22]. At this depth the number of muon-nucleus inelastic interactions with a transferred energy E_{tr} > 100 MeV is about 0.11 per year in 1 ton of rock, as can be calculated from Ref. 23. The spectrum of these muons and their energy transfer in inelastic collisions with nuclei can be calculated from the known sea-level muon spectrum [24] and from the muon energy losses [23].

We pointed out earlier that the large ΔA of selected parent-daughter combination should be helpful for the suppression of background technetium production initiated by muon interactions with tin. This, however, is true only for low-energy muons. If the energy transferred to the tin nuclei by scattered muons is sufficiently large (~ > 200 MeV) it might result in inelastic interactions with a wide spectrum of ΔA losses. Since no direct experimental data on isotope production by high-energy muons are available, we estimated this effect by using the Rudstam formula [25] with parameters from [26]. In this way we calculated the concentration of technetium isotopes produced by muons per ton of initial tin at a depth of one mile of standard rock (ρ=2.65 g$\cdot$cm^{-3}) as $2.08 \cdot 10^3$ for ^{97}Tc, $0.63 \cdot 10^3$ for ^{98}Tc, and 22 for ^{99}Tc. We hope that the accuracy and reliability of these estimates can be further improved if some model (for example, [27]) that fits all the existing

experimental data on the yield of isotopes in high-energy collisions for all particles can be employed and if measurements of isotope yields resulting from interactions with virtual photons can be performed (for example, in the electron beam at SLAC). In Table 1 below we give the estimated yields of technetium isotopes due to cosmic muon interactions with tin for two burial depths of 1 mile and 2 miles, corresponding to pessimistic and optimistic background levels.

(c) Solar neutrino background. If molybdenum is presented in the ore the following reactions $^{97}Mo(\nu,e)^{97}Tc$ and $^{98}Mo(\nu,e)^{98}Tc$ might also produce technetium isotopes. In Ref. 28 the authors estimated the total production rate for ^{97}Tc due to this reaction to be $5.9 \cdot 10^{-36}$ atoms per ^{97}Mo-atom per second. Thus, assuming 1 ppm level (by number) of ^{97}Mo in the initial tin ore (abundance of ^{97}Mo is 9.55%) we obtain a concentration of ^{97}Tc at the level of ~ 2.5 per ton of initial tin. For the ^{98}Tc, due to the higher threshold of the $^{98}Mo(\nu,e)^{98}Tc$ reaction, the corresponding concentration is much smaller (see Table 1). The same authors estimated the contribution of another source of background associated with the presence of molybdenum, the process $^{97}Mo(p,n)^{97}Tc$. Their conclusion was that the yield of technetium for this process will be below the level of solar-neutrino production. If the concentration of molybdenum does not exceed the few-tens of ppm level, this source of background can be neglected.

(d) Technetium contamination during processing and analysis. It is very often quoted in the literature that technetium "was never found in nature". This is not quite correct. For example, tiny amounts of ^{99}Tc were found in uranium ores in the Belgium Congo [29]. Moreover, starting in 1945, technetium began to be produced in large quantities "artificially" in nuclear weapon tests and in nuclear reactors. To date, the total amount of technetium produced in nuclear weapon tests in the atmosphere is estimated to be 250 kg [30]. This includes a total of ~ 3 mg of ^{98}Tc, or ~ $3.5 \cdot 10^4$ atoms per square meter of the earth's surface. The distribution of technetium obviously is not uniform. The highest concentration should be found in the top layers of soil near the nuclear explosion sites. Measurements [31] of ^{99}Tc concentration in the soil in Japan have shown the presence of ~ $6 \cdot 10^{11}$ atoms per 1 kg of soil at the surface. This concentration falls quickly with increasing depth. At a depth of ~ 25 cm below the surface it is already lower by a factor ~100. The concentration of ^{98}Tc should be lower by factor of ~10^8 and concentration of ^{97}Tc by another factor of ~ 70. Thus, we expect the artificial technetium contamination of tin ores from the deeply buried deposits to be negligible. We also anticipate that all necessary precautions can be taken in order to avoid contamination during the ore processing, transportation, and during analysis of the samples.

Discussions and conclusions

We now can include background effects in the estimates of the possible achievable limits in the search for baryon instability via the detection of long-lived isotopes of technetium in nature. The baryon instability signal can be treated as a statistically significant excess over the background in concentrations of technetium isotopes in deep deposits of tin ores. We use the assumptions discussed above and summarized here as follows: (a) large volumes (up to 10^5 tons) of tin ore can be processed industrially with technetium extraction as an inexpensive by-process; (b) tin ore deposits from depths

between 1 and 2 miles (4-8 km of water equivalent) can be used; (c) the concentration of uranium in the tin ore is in the range of 10 ppb to 1 ppm and can be determined; (d) the chemical extraction efficiency of technetium will be ~ 25%; and (e) the minimum detection level of laser selective photoionization spectroscopy of technetium isotopes is 100 atoms, corresponding to an efficiency of ~ 1% with no ion detection background. Under these assumptions we have calculated the background contributions from different sources for the three isotopes of technetium considered above. These calculations are summarized in Table 1 below and are also presented in Figure 3 in term of the estimated discovery potential of the proposed method for ^{97}Tc and ^{98}Tc isotopes, assuming the optimistic and pessimistic background scenarios from table 1.

Table 1. Expected concentrations of technetium isotopes (atoms per ton) in deep-deposition of tin ores.

Isotope:	^{97}Tc	^{98}Tc	^{99}Tc
Expected Tc yield due to baryon instability:			
$\tau_{isotope}$, years	$2.6 \cdot 10^6$	$4.2 \cdot 10^6$	$2.1 \cdot 10^5$
$n \rightarrow \bar{n}$ branching, ξ	0.0307	0.0056	0.0038
Yield atoms/ton for $\tau_{n\bar{n}}=10^{32}$ yr	278	82	2.8
Pessimistic background yield:			
Yield atoms/ton due to cosmic μ (at depth of ~1 mile of rock)	2077	626	22
Yield atoms/ton due to 1 ppm of ^{238}U	17	1,880	$8 \cdot 10^9$
Optimistic background yield:			
Yield atoms/ton due to cosmic μ (at depth of ~2 mile of rock)	2.6	0.8	0.03
Yield atoms/ton due to 10 ppb of ^{238}U	0.17	19	$8 \cdot 10^7$
Yield atoms/ton due to solar ν	2.5	<1	–

The discovery potential in Figure 3 is expressed in metric tons of initial tin needed to be processed in order to establish the excess technetium concentration signal over the background level as 2.3 sigma (90% CL) of statistical fluctuation of the background. Additional requirement imposed on the estimated discovery potential limit is that the

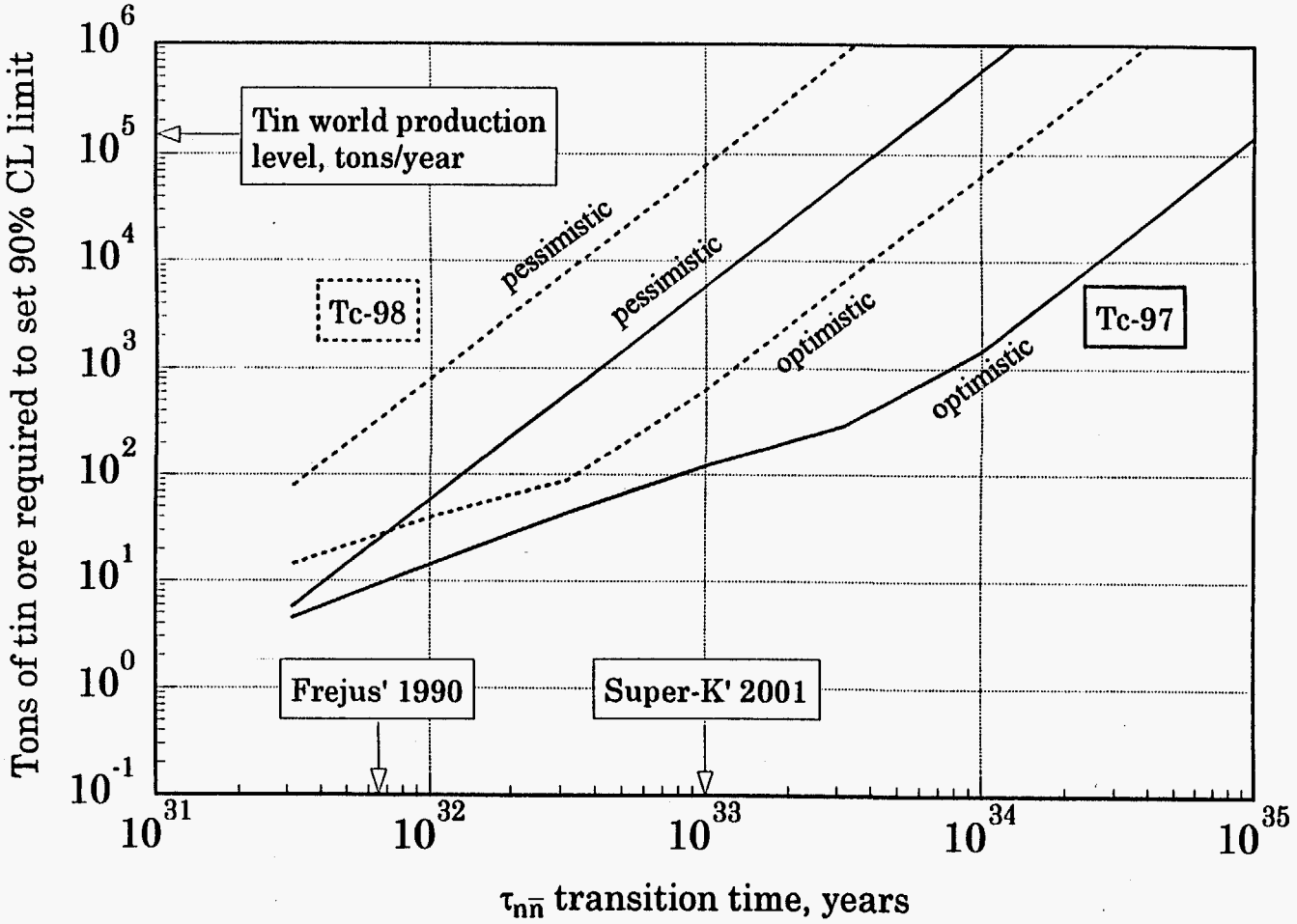

Fig 3. Amount of initial tin ore to be processed in order to establish, at 90% confidence level, the excess concentration of technetium isotopes due to intranuclear neutron-antineutron transitions in tin relative to background concentration of technetium. "Optimistic" and "pessimistic" assumptions used for background calculations are explained in the text.

minimum detected number of atoms be higher than 10. It is also assumed that the background can be estimated reliably and that systematic errors can be minimized as a result of direct measurements, related to the technetium production processes, made at different depths and extrapolated to the working depth.

Simultaneous detection of all three isotopes, together with direct measurements of uranium concentrations in the ore, muon flux measurements, and determination of the yield of technetium isotope production in uranium fission should provide additional constraints on the determination of contributions of different backgrounds. The efficiency of chemical extraction of technetium should be measured in the industrial environment with samples of known concentration entered at the beginning of the separation process. A minimum detection level of 100 atoms of technetium by the laser selective photoionization spectrometry methods needs to be demonstrated. Also, it will be very important to locate tin mines with a burial depth of deposits of more than a 4,000 m of water equivalent.

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References:

1. M. Goldhaber, these proceedings.

2. A. D. Sakharov, JETP Lett. **5** (1967) 24.

3. J. C. Pati, these proceedings.

4. R. N. Mohapatra, these proceedings.

5. P. Nath, these proceedings.

6. J. Stone, these proceedings.

7. A. Mann, these proceedings.

8. D. Gibin, these proceedings.

9. Yu. A. Kamyshkov, these proceedings.

10. S. P. Rosen, Phys.Rev.Lett. **34** (1975) 774.

11. J. C. Evans, Jr., R. I. Steinberg, Science **197** (1977) 989; also R. I. Steinberg, J. C. Evans, Jr., E. L. Fireman, in Proceedings of the International Conference on Neutrino Physics, Elbrus, USSR, June 1977.

12. E. W. Hennecke, O. K. Manuel, D. D. Sabu, Phys.Rev. **C 11** (1975) 1378.

13 Review of Particle Properties, Phys.Rev. **D 50** (1994) 1675.

14 A. S. Il'inov, M. V. Mebel', and S. E. Chigrinov, Yad.Fiz. **37** (1983) 18-26.

15 V. S. Letokhov, V. I. Mishin, A. A. Puretzky, Prog. Quant. Electr. **5** (1977) 139-303.

16 H.-U. Hasse et al., in proceedings of the Seventh International Symposium on the Resonance Ionization Spectroscopy 1994, pp 499-502 (1995).

17 S. V. Andreev, V. S. Letokhov, V. I. Mishin, Phys. Rev. Lett. **59** (1987) 1274.

18. G. W. Parker, these proceedings.

19. Ye. S. Golubeva et al., these proceedings.

20. R. Vandenbousch, J. R. Huizenga, "Nuclear Fission", Academic Press, NY 1973.

21. B. Mason, "Principles of Geochemistry", Wiley, NY 1966.

22. Ch. Berger et al. (Frejus Collaboration), Phys. Rev. **D40** (1989) 2163.

23. W. Lohman, R. Kopp, R.Voss, "Energy Loss of Muons in the Energy Range 1-10,000 GeV", CERN 85-03 (1985).

24. L. V. Volkova, G. T. Zatsepin, L. A. Kuz'michev, Yad. Fiz **29** (1979) 1252-1265.

25. G. Z. Rudstam, Naturf. **21a** (1966) 1027.

26. G. G. Jonsson, K. Lindgren, Physica Scripta Vol.**15** (1977) 308-310.

27. J. Jastrzebski et al., Phys.Rev. **C 47** (1993) 216.

28. G. A. Cowan, W. C. Haxton, Science, Vol. **216** (1982) 51-54 .

29. B. T. Kenna, P. K. Kuroda, J. Inorg. Nucl. Chem. **23** (1961) 142.

30. K. Yoshihara, Topics in Current Chemistry **176** (1996) 17.

31. S. Morita, K. Tobita, M. Kurabayashi, Radiochim Acta **63** (1993) 63.