Reports

Solar Neutrino Production of Technetium-97 and Technetium-98

Abstract. *It may be possible to determine the boron-8 solar neutrino flux, averaged over the past several million years, from the concentration of technetium-98 in molybdenite. The mass spectrometry of this system is greatly simplified by the absence of stable technetium isotopes, and the presence of the fission product technetium-99 provides a monitor of uranium-induced backgrounds. This geochemical experiment could provide the first test of nonstandard solar models that suggest a relation between the chlorine-37 solar neutrino puzzle and the recent ice age.*

A serious discrepancy exists between the solar neutrino capture rate measured in the ^{37}Cl experiment (*1*) and that predicted by the standard solar and weak interaction models (*2*). If, as many have suggested, this discrepancy is due to a misunderstanding of the physics of the solar interior, the implications for present theories of stellar evolution could be profound (*3*). Alternatively, if the sun does produce the expected neutrino flux, then some mechanism must be altering the character of these neutrinos before they reach the earth. This suggestion now seems particularly interesting in view of recent evidence for massive neutrinos (*4*) and neutrino oscillations (*5*).

These two classes of solutions to the ^{37}Cl puzzle can be distinguished. Proposed modifications of the standard solar model to accommodate the ^{37}Cl capture rate result primarily in a reduced flux of high-energy ^{8}B neutrinos, whose production depends most critically on the central temperature of the sun. Neutrino oscillations or decay would, except under unusual circumstances, affect all components of the solar neutrino flux equally. Thus there has been great interest in mounting new experiments to complete the spectroscopy of the neutrino sources shown in Table 1. Among those attracting serious attention are radiochemical experiments employing ^{71}Ga (*6*) and ^{81}Br (*7*) targets and a counter experiment with ^{115}In (*8*).

Several interesting proposals for geochemical experiments have also been made. These suggest measuring the concentrations of certain long-lived isotopes produced in natural ore bodies and salt deposits by solar neutrinos. The advantage of this approach over the laboratory experiments is that much larger concentrations of the daughter atoms can be produced over geologic times. The disadvantage is that one must be able to tolerate the backgrounds found in nature. Three isotopes that have been discussed are ^{205}Pb (half-life, $\tau_{1/2}$ = 14 million years) (*9*), ^{81}Kr ($\tau_{1/2}$ = 0.21 million years) (*7*), and ^{41}Ca ($\tau_{1/2}$ = 0.10 million years) (*10*), which are produced by the neutrino reactions on ^{205}Tl, ^{81}Br, and ^{41}K, respectively. At the moment, unresolved problems complicate each of these proposals. The capture cross section for ^{205}Tl is uncertain (*11*), and no ^{81}Br source has been found for which backgrounds from natural radioactivity and cosmic rays are acceptable (*12*). The technology for discriminating ^{41}Ca from ^{40}Ca at the necessary sensitivity has not yet been demonstrated (*13*) and the background problem is very troublesome.

In this report we discuss another geochemical possibility, the production of ^{98}Tc ($\tau_{1/2}$ = 4.2 million years) from ^{98}Mo (24.1 percent). This technetium isotope is of great interest not only in the context of conventional solar neutrino measurements, but also as a unique test of the steady-state sun hypothesis implicit in the standard solar model. The isotope ^{97}Tc ($\tau_{1/2}$ = 2.6 million years) is also produced by neutrinos but will be masked by the neutron capture product in ^{96}Ru unless the ruthenium content in the molybdenum ore is extremely low.

The threshold for the ^{98}Mo(ν,e)^{98}Tc reaction is 1.68 MeV. However, transitions from the 0^+ ground state of ^{98}Mo to the $(6)^+$ ground state and $(4,5)^+$ (23 keV) first excited state in ^{98}Tc are strongly hindered. Thus the effective threshold for neutrino excitation is $\gtrsim$1.74 MeV and only high-energy ^{8}B neutrinos can produce ^{98}Tc. The ^{97}Mo(ν,e)^{97}Tc reaction has a lower threshold, 0.32 MeV, and so in principle can be induced by the main component of the solar neutrino flux, the pp neutrinos. However, the ground state transition ($5/2^+ \rightarrow 9/2^+$) is again strongly hindered [log(ft) $\approx$ 13]. The second most plentiful neutrinos, from electron capture on ^{7}Be, can induce the Gamow-Teller (GT) transitions to the $7/2^+$ (216 keV) and $5/2^+$ (324 keV) excited states in ^{97}Tc, and of course the ^{8}B neutrinos can excite many levels.

Quantitative estimates of capture rates can be made by folding the GT and Fermi strength distributions in ^{97}Tc and ^{98}Tc with the various solar reaction neutrino spectra of Table 1. The model-independent Fermi contribution to the capture rate is relatively weak, as only those few ^{8}B neutrinos with energies $\gtrsim$ 11.4 MeV can excite the analog states. As these states decay by isospin-forbidden neutron emission (*14*) and as ^{99}Mo is unstable, only the Fermi transition in ^{98}Mo yields a product of interest, ^{97}Tc. The production rate, based on the standard model ^{8}B neutrino flux of Bahcall *et al.* (*2*), is 0.5 solar neutrino unit (1 SNU = 10^{-36} capture per target atom per second). However, we bear in mind that the ^{37}Cl experiment proves that today the ^{8}B flux is considerably weaker

Table 1. Neutrino sources and fluxes. Reactions (1) to (4) produce solar neutrinos with continuous distributions, while reactions (5) and (6) are line sources. E_ν^{max} is the maximum energy of the neutrinos for all reactions except (4), where it has been computed with respect to the center of the broad 2.9-MeV ^{8}Be resonance populated in the beta decay of ^{8}B. Fluxes are taken from the standard solar model calculation of Bahcall *et al.* (*2*).

Reaction	E_ν^{max} (MeV)	Flux (10^{10} cm^{-2} sec^{-1})
(1) $p + p \rightarrow {}^2H + e^+ + \nu$	0.420	6.1
(2) $^{13}N \rightarrow {}^{13}C + e^+ + \nu$	1.199	4.6×10^{-2}
(3) $^{15}O \rightarrow {}^{15}N + e^+ + \nu$	1.732	3.7×10^{-2}
(4) $^{8}B \rightarrow {}^{8}Be + e^+ + \nu$	14.02	5.85×10^{-4}
(5) $^{7}Be + e^- \rightarrow {}^{7}Li + \nu$	0.862 (89.6 percent) 0.384 (10.4 percent)	4.1×10^{-1}
(6) $p + e^- + p \rightarrow {}^2H + \nu$	1.442	1.5×10^{-2}

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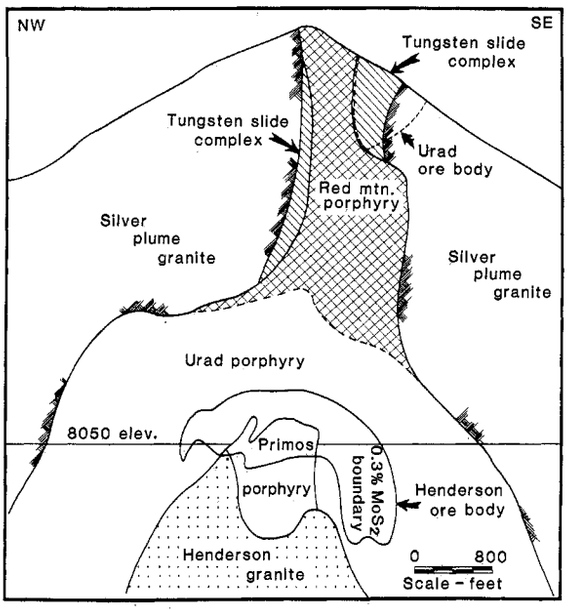

Fig. 1. Geologic section of Red Mountain showing the Henderson ore body, from Ranta *et al.* (*23*). The summit is at an elevation of 12,315 feet.

than predicted by the standard model.

Model-dependent calculations of the GT strength distributions in ^{97}Tc and ^{98}Tc can be made, but it is difficult to estimate the accuracy of such theoretical predictions. We proposed instead to measure these distributions by using forward-angle (p,n) reactions off molybdenum, as it was demonstrated recently that this technique can map known GT transitions with a typical accuracy of 5 percent (*15*). Presently, however, we need a rough estimate of the capture rates. Shell model calculations (*16*) predict that the transitions to the first $7/2^+$ and $5/2^+$ states in ^{97}Tc are quite weak, resulting in a small capture rate for ^{7}Be neutrinos (*17*). Thus the ^{97}Tc concentration in molybdenite (MoS_2) may also monitor the high-energy ^{8}B neutrinos. To crudely estimate the ^{8}B capture rates we scale a recent calculation (*18*) of the ^{81}Br^{8}B cross section by the ratio of neutron excesses, as the naïve single-particle GT sum rule is proportional to the neutron number minus the proton number ($N - Z$), and by a factor accounting for the Z dependence of the Coulomb distortion of the final-state electron. We find GT ^{8}B capture rates of 5.0 and 4.6 SNU for ^{98}Mo and ^{97}Mo, respectively. Taking into account the ratio of abundances for ^{98}Mo and ^{97}Mo, we obtain a total production rate for ^{97}Tc of 5.9×10^{-36} atom per ^{97}Mo atom per second.

The most difficult aspect of geochemical experiments is the demonstration that backgrounds from natural radioactivity and from cosmic rays are tolerable. In the present case, the most common mineral containing molybdenum, molybdenite (MoS_2), is found primarily as an accessory mineral in altered granitic rock. The uranium content of granitic rocks averages 3.7 ppm (*19*) and may be 20 ppm in molybdenite deposits (*20*) (although the thorium content is quite low). This discouraged our interest in molybdenum in an earlier investigation (*10*) of geochemical experiments. We will now sketch the results of careful background calculations (*16*) which show that we were unduly pessimistic. Critical to this discussion is the occurrence locally of molybdenite in macroscopic crystals in which the metal-to-sulfur atomic ratio is essentially 1:2. While details of these compound nucleus statistical calculations will be left to (*16*), we point out these techniques have been tested against the ^{37}Cl background calibrations (*12*) (α,p) followed by ^{37}Cl(p,n)^{37}Ar in C_2Cl_4 and ^{34}S(α,n)^{37}Ar in CS_2 and found to agree well.

Although both ^{97}Tc and ^{98}Tc can be produced by direct (α,p) reactions on Mo, the unfavorable thresholds, 6.052 and 6.142 MeV, and the associated Coulomb barriers greatly suppress the charged-particle channels. Much more serious are the two-step reactions S(α,p)Cl followed by ^{97}Mo(p,n)^{97}Tc and ^{98}Mo(p,n)^{98}Tc. Assuming that the alpha particles initiating these reactions are produced in the uranium sites distributed uniformly in the ore outside the molybdenite crystals, we find a ratio of solar neutrino–induced to background-induced Tc of $R\langle d\rangle/3.75U$. Here U is the typical uranium content (parts per million) in the veinlets where the molybdenite is concentrated, R is the solar neutrino capture rate (SNU), and $\langle d\rangle$ is the mean thickness of the molybdenite crystal (micrometers). We take $U = 20$, although we do not know that the veinlet uranium content will mimic that of the full ore body, and $\langle d\rangle = 100$, yielding the signal-to-background ratio of 6.7 for ^{98}Tc. However, this estimate may be pessimistic. The uranium is concentrated in grains of zircon, aeschynite, and rare-earth oxides typically 50 to 100 μm in diameter (*20*), so that the average alpha-particle energy on leaving the grain is significantly lower than that used in our calculations.

Another important two-step background is due to fission neutrons, S(n,p)P followed by Mo(p,n)Tc. As the range of the neutrons is large compared to $\langle d\rangle$, we treat this reaction as if the uranium were dispersed throughout the molybdenite. We find a signal-to-background ratio of 510 R/U, or about 130.

Several additional backgrounds differ fundamentally from those discussed above in that technetium is produced from elements residing in host minerals other than molybdenite. The commercial processing of the raw ore begins with repeated flotation, yielding an ore concentrate containing 85 to 90 percent molybdenite. Thus, to the extent that this flotation process separates the host minerals from the molybdenite, these backgrounds are removed, assuming that these minerals retain technetium over 4.2 million years.

The isotope ^{98}Tc can be produced by the (n,p) reaction induced by fission neutrons on ^{98}Ru. The production rate can be estimated from the systematics of fission-averaged (n,p) reactions, taking the total neutron cross section in the host mineral equal to that for molybdenite. We find a signal-to-background ratio of 2.3×10^4 $R/U \times$ Ru, where Ru is the ruthenium content of the molybdenite concentrate in parts per million. For $U = 20$ and $R = 5$ the signal-to-background ratio will exceed 10 for Ru $\lesssim 580$, so we do not anticipate that this background will be troublesome.

In contrast, background problems for ^{97}Tc may seriously impair the usefulness of this isotope as a monitor of solar neutrinos. The most important background is likely to be thermal neutron capture by ^{96}Ru followed by the electron capture transition ^{97}Ru $\rightarrow$ ^{97}Tc (note, in contrast, that ^{98}Tc is a shielded nucleus). The signal-to-background ratio is 3.9 $R/U \times$ Ru, so that a ruthenium content of 0.11 ppm in the concentrated ore generates a 10 percent background. Whether such efficient separation of ruthenium-bearing minerals occurs in the flotation process is doubtful. Also, ^{97}Ru can be

produced by $^{94}Mo(\alpha,n)^{97}Ru$ induced by long-range alpha groups in the decay of ^{214}Po, although the finite molybdenite grain size probably renders this background tolerable. The signal-to-background ratio corresponding to a third reaction, $^{93}Nb(\alpha,\gamma)^{97}Tc$, is 170 $R/U \times$ Nb, where Nb is the niobium content of the concentrated molybdenite ore in parts per million. Thus Nb = 4.2 ppm will generate a 10 percent background. However, this result was calculated assuming the uranium and niobium are dispersed in the host minerals. Thus significantly higher Nb levels may be acceptable, depending on the grain sizes of the niobium- and uranium-bearing minerals.

Spontaneous fission of uranium can also produce technetium. From systematics (*16*) we find a signal-to-background ratio of 9.3×10^4 $R/\tilde{U}$ for ^{98}Tc, with $\tilde{U}$ denoting the uranium content of the molybdenite ore concentrate in parts per million. If $U = 20$ ppm and we assume that all uranium in the raw ore follows molybdenite in the flotation process, this background is less than 1 percent of the solar neutrino signal. Fission production of the more neutron-poor ^{97}Tc is presumably less.

Finally, evaporation protons are produced in molybdenite by high-energy cosmic-ray muons. We estimate from (*12*) that an overburden equivalent to 4100 m of water will be necessary to reduce cosmic-ray–induced technetium to 10 percent of the solar neutrino signal. A density of 2.9 g/cm^3 is appropriate for the host rocks of the molybdenite ore, so the required depth is 1410 m (*21*).

We know of one commercial molybdenum source that satisfies this criterion, the Henderson ore body at Red Mountain, Clear Creek County, Colorado (*22, 23*). A geologic section of Red Mountain, from Ranta *et al.* (*23*), is shown in Fig. 1. The ore contains 0.49 percent molybdenite on average, is currently being mined at a depth in excess of 1132 m, and extends to more than 1500 m below the surface (*23*). Furthermore, for the present geologic experiment, the effective depth is somewhat greater, as the ore body at time of formation (~25 million years ago) lay 1500 to 1800 m below the surface (*22*). The present minimum depth of overburden is at the valley floor through which the deposit is entered. This floor resulted from geologically recent glacial scouring (~10,000 years ago).

An experiment may require 13 tons of molybdenite extracted from 2600 tons of ore, yielding 10^7 atoms of ^{97}Tc and ^{98}Tc, respectively (*24*). In commercial processing the molybdenite is concentrated by repeated flotation of finely ground ore, and molybdenum is then separated by roasting with excess oxygen. Under these conditions rhenium, an element chemically analogous to technetium, forms volatile oxides at the controlled furnace temperature (~700°C) and passes into the gas stream, largely composed of SO_2 and air, with efficiencies that can exceed 90 percent (*25*). Under certain conditions, calcium and potassium perrhenates may form and remain in the ash. We believe that, under current operating conditions, most of the rhenium and, presumably, the accompanying technetium can be recovered from a gas-scrubbing operation before conversion of the SO_2 to sulfuric acid. After additional processing to recover a pure rhenium-technetium fraction, it will be necessary to prepare a pure sample of technetium appropriate for mass spectrometric analysis. We believe state-of-the-art tandem mass spectrometry could provide a discrimination factor of 10^8 between masses 98 and 99, with sufficient sensitivity and a low enough background to permit quantitative measurements at the 10^7 atom level.

A "standard" sample for analysis may contain ~10^7 atoms of solar neutrino–induced ^{98}Tc associated with ~2×10^{12} atoms of ^{99}Tc ($\tau_{1/2} = 0.21$ million years), which would satisfy these spectrometric specifications. The actual content of ^{99}Tc, which is produced principally by spontaneous fission of ^{238}U in the ore, is given by $^{99}Tc/^{98}Tc = 2.8 \times 10^4$ $\tilde{U}/R$. If the uranium content of the concentrated molybdenite ore mimics that of the raw ore, $U = \tilde{U} = 20$ ppm, this ratio is ~10^5.

Proportionate retention of ^{99}Tc in processing or addition of a tracer may be necessary to the overall analysis. The ^{99}Tc concentration, a direct measure of the uranium content, should track the radioactivity backgrounds we have discussed. A demonstration that ^{97}Tc and ^{98}Tc are not correlated with ^{99}Tc could be viewed as evidence that such backgrounds are not significant (*26*). In addition, to the extent that these isotopes remain together, it should be possible to measure the chemical efficiency for recovering technetium by monitoring the uranium and ^{99}Tc throughout the process.

A final point requiring careful study is the mobility of technetium over geologic times in the reducing environment of the Henderson ore body. (Technetium is quite mobile in oxidizing environments.) We must ascertain that the loss of technetium is low. A demonstration that the ^{99}Tc is close to secular equilibrium with uranium is a prerequisite for a successful neutrino flux measurement.

Clearly, a number of points we have raised will require careful experimental and theoretical study before the discussion begun here can be considered complete. However, we are cautiously optimistic. Part of our enthusiasm for undertaking this project is due to our belief that a unique test of suggested periodic variations (*27, 28*) in the sun's energy production might result. Dilke and Gough (*28*), in their discussion of the "solar spoon," suggest that an overstability causes the sun's core to mix every few hundred million years. There would follow a temporary reduction in the solar luminosity, which could induce a terrestrial ice age of about 2 million years duration, and a somewhat more extended depression in the solar neutrino flux. As the Pleistocene Epoch began about 3 million years ago, we may be living in such a special time. Mixing would have occurred about 4 million years ago, and the sun would not presently be in thermal equilibrium, thus accounting for the ^{37}Cl result. The most sensitive neutrino monitor of this mixing is the ^8B flux. To our knowledge, the technetium experiment discussed here is the only proposal to test this flux on time scales characteristic of changes in the solar core.

In particular, clear memory of the steady-state phase prior to mixing should be retained in the concentration of ^{98}Tc ($\tau_{1/2} = 4.2$ million years). We estimate that this concentration would be 65 percent of the steady-state value, considerably larger than that obtained by directly extrapolating from today's ^{37}Cl experiment. About 79 percent of the ^{98}Tc would have been produced prior to mixing.

Finally, Cahn and Glashow (*29*) recently suggested that chemical isolation of Tc could have important implications for unified theories of the strong and electroweak forces in which particles in the mass range 10 GeV to 100 TeV are present. If some abundance of integrally charged superheavy particles $X^{\pm}$ was created in the early days of the universe, superheavy nuclei may now exist. In particular, the X^- could bind electromagnetically to Ru to form a stable nucleus that would have the charge of Tc and be chemically similar to both Tc and Re. The principal commercial source of Re is molybdenum ore, as ReS_2 is often the primary impurity in molybdenite. Presumably, if RuX^- exists, it should also concentrate in molybdenite. Al-

though the Henderson ore is relatively poor in Re, we expect that the Re-Tc fraction obtained from our standard ore sample will contain 100 g of this metal. Thus, if 10^7 atoms of RuX^- can be detected in the pure "Tc" sample prepared from this fraction, relative abundances $RuX^-/Re = 10^{-17}$ can be measured. This implies a sensitivity to an average RuX^- concentration in the earth's crust at 1 part in 10^{26}.

Note added in proof: Measurements of the uranium content of samples of Henderson ore and ore concentrate give $U = 11.8$ ppm and $\tilde{U} = 1.8$ ppm. Our background estimates in this report should be reduced accordingly.

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Search for Interstellar Superheavy Hydrogen

Abstract. *Models for fundamental physical interactions allow for the existence of stable or nearly stable elementary particles much heavier than the proton. Stellar spectra were searched for a positively charged superheavy particle, X^+, which, with a bound electron, should appear as apparently superheavy neutral hydrogen in the interstellar medium. An upper limit for the abundance of X relative to normal hydrogen in the line of sight toward the bright star γ Cassiopeiae is 2×10^{-8}.*

Cahn and Glashow (*1*) described how models for unified fundamental interactions might allow for the existence of essentially stable superheavy elementary particles. If a positively charged superheavy, X^+, exists, it will appear chemically as very heavy hydrogen when an electron is bound to it. Smith and Bennett (*2*) placed an upper limit in terrestrial water samples for the fractional abundance of X relative to H of $n(X)/n(H) < 10^{-21}$ if the mass of X lies between 6 and 350 proton masses; apparently this limit can be reduced by orders of magnitude in the near future. Here we report a search for X in the interstellar medium. Our upper limit is much greater than the terrestrial bound, but our search may nevertheless be useful since we examine a different portion of the universe and our result is valid for any particle mass greater than about four times that of the proton.

Our search procedure is similar to that used to discover interstellar deuterium (*3*, *4*); that is, we searched for resonance absorption lines from the ground electronic state of hydrogen-like X atoms in the spectrum of background stars. Because both hyperfine splitting (*5*) and the energy shifts which result from the finite size of the nucleus are likely to be small (*6*), the electronic isotope shift of X is expected to be dominated by the difference in the reduced mass between X and H. Therefore, we expect the electronic spectrum of superheavy hydrogen to be similar to that of hydrogen only with an isotope shift that corresponds to an apparent Doppler motion of -160 km sec^{-1} (for an infinitely heavy nucleus) instead of -80 km sec^{-1} for deuterium.

Interstellar Lyman α absorption is generally so broad that it is usually impossible to detect lines only 160 km sec^{-1} from the line center, and it is necessary to use higher order Lyman lines. Consequently, the only suitable data are from the Copernicus satellite, which was sensitive to wavelengths shortward of 1200 Å. Since this satellite is no longer operational, we must use data from previous sensitive searches for interstellar deuterium. We require observations of stars (i) that are rapidly rotating so that blends with stellar lines are not important, (ii) that are sufficiently bright that a high signal-to-noise ratio was obtained, (iii) that are sufficiently nearby that there is no high-velocity normal hydrogen in the line of sight, and (iv) yet are sufficiently far that the amount of hydrogen in the line of sight to the star is appreciable. From the results given in (*7*), the most suitable star for our search is γ Cassiopeiae, where the interstellar medium in the line of sight is reasonably well understood (*8*).

From the figure published in the paper of Vidal-Madjar *et al.* (*9*) and the standard procedure for placing upper limits from Copernicus data (*10*), we estimate a 3 standard deviation upper limit to the equivalent width of the Lyman β line of superheavy hydrogen toward γ Cassiopeiae of 1.5 mÅ. Using the same oscilla-

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