THE MOLYBDENUM SOLAR NEUTRINO EXPERIMENT

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Abstract

The present status of the molybdenum solar neutrino experiment is described.

In 1982 a proposal was made to determine the ^{8}B solar neutrino flux, averaged over the past several million years, from the concentrations of ^{97}Tc and ^{98}Tc in a deeply buried molybdenite deposit.[1] Progress on the experiment has been rapid. At this conference I would like to describe both the motivation for and present status of these geochemical measurements.

Davis's ^{37}Cl experiment[2] has established that the flux of ^{8}B neutrinos is less than that predicted by the standard solar model[3] and the standard model of weak interactions. One of the key constraints on solar models is the need to burn hydrogen at a rate consistent with the sun's luminosity. However, as photons require approximately 10^{7} years to migrate from the core to the sun's surface, this constraint is strictly true only in steady-state models. This observation has stimulated a number of nonstandard models in which secular variations in the solar core are postulated to explain the solar neutrino puzzle.[4] Clearly, a test of the standard model assumption of hydrostatic equilibrium and steady-state hydrogen burning is then of fundamental importance. The molybdenum experiment can provide such a test: a measurement of the ^{8}B neutrinos, our most sensitive probe of the sun's central temperature, over a period comparable to the Kelvin time of the solar core.

Over geologic times the ^{8}B neutrinos will produce measurable concentrations of ^{98}Tc ($\tau_{1/2}$ = 4.2 Myr) and ^{97}Tc ($\tau_{1/2}$ = 2.6 Myr) in a deeply-buried molybdenum deposit. As large quantities of ore are needed, one must find a deposit that is worked commercially. In the U.S. only one active deep mine exists, the AMAX Corporation's Henderson Mine in Clear Creek County, Colorado. This deposit extends from 1100 m to 1500 m below the surface[5], and was considerably deeper at the time of formation ($\sim$ 25 million years ago[6]). The present points of minimum overburden are due to glacial scouring that occurred only 10,000 years ago. Throughout most of the ore body the technetium concentration due to cosmic ray backgrounds is estimated to be less than 10% of the standard model solar neutrino signal.

A kiloton sample of this ore will contain approximately five tons of MoS_2 and, assuming standard model fluxes, somewhat in excess of 10^7 atoms each of ^{97}Tc and ^{98}Tc. The task of chemically isolating[7] this trace quantity of technetium for mass spectrometry is greatly simplified by the commercial processing of the ore. Flotation separation at the Henderson mine produces a concentrate that is 90 to 99% molybdenite. The concentrate is shipped to the AMAX roasting plant at Ft. Madison, Iowa, for conversion to MoO_3. The two roasters at this plant have a capacity of 50 tons/day, and thus process concentrate for a typical experimental run (approximately 10 tons) in a fraction of a day. In the roasting process elements such as rhenium and technetium form volatile oxides that pass into the gas stream, largely composed of SO_2, SO_3, and air, with high efficiencies. The gas stream is treated to remove dust, then cooled, scrubbed, and converted to sulfuric

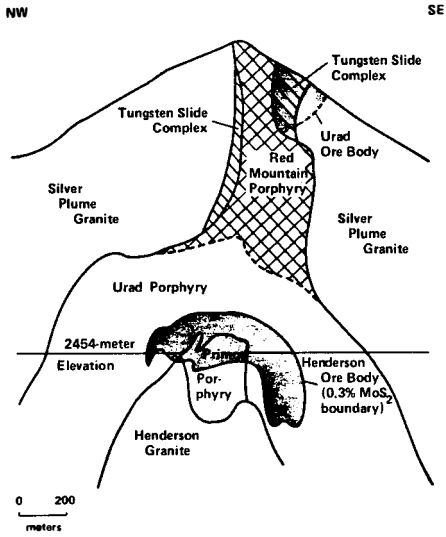

Fig. 1. Geologic section of Red Mountain showing the Henderson ore body (from Ref. 5).

acid. The scrubbing removes rhenium, an element chemically analogous to technetium, and unwanted fluorides, chlorides, and selenium before the gas stream enters the sulfuric acid plant. The initial experimental task is the quantitative recovery of technetium and rhenium from the scrub stream. Ion exchange columns for removing technetium from the scrub stream were first installed in the summer of 1985 and have been tested during runs of concentrate from the Climax mine (Colorado) and from Arizona mines. To improve the absorption of technetium, a pumping system was installed upstream to oxidize the scrub by the addition of NaOCl. These modifications of AMAX procedures cause only minimal disruptions in the commercial operation of the plant.

Pilot experiments monitored the rhenium recovery in these columns. Analysis of column effluents showed a rhenium concentration of 12 ppb, compared to 90 ppm in the scrub upstream of the columns, after four hours of operation. Thus the rhenium concentration in the acid scrub was reduced by a factor of 7500. Because technetium absorbs more strongly than rhenium, quantitative removal of technetium from the scrub stream is assured. These results indicate that four 12-inch columns operating at 7.5 gallons/min should remove ~ 90% of the technetium from the full scrub stream during a 12-hour run.

The experimentalists also established a mass balance for rhenium of 95% relative to the molybdenite feed. Because of uncertainties in flow rates, this value is uncertain to ± 10%. As hoped, it appears that the rhenium does report to the acid scrub stream and is not lost in a sink elsewhere in the plant.

A five-day run of Henderson concentrate is scheduled for February, 1986. Twenty-six ion exchange columns, six drums of NaOCl, and associated supplies have been delivered to the Ft. Madison plant in preparation for this run.

The recovery of technetium from the resin is done at Los Alamos. To remove the kilogram quantities of co-absorbed molybdenum, the resin is washed with a 2 M NaOH/0.5 M NaCl solution. The resin is then ashed in large Pyrex containers. The technetium loss from those procedures is less than 10%. The technetium in the ash is ≥ 95% soluble in 6 M HCl.

To separate technetium from 100 g quantities of rhenium, the technetium is precipitated out of HCl solutions containing 0.06 M $KReO_4$, 0.06 M Fe^{2+}, and 0.02 M Na_2SO_3. After the capped solution is permitted to undergo reduction for 90 minutes, 15 M NH_4OH is added to precipitate $Fe(OH)_2$ and TcO_2. The efficiency of the technetium removal depends on the acidity of the solution during reduction. Approximately 2% of the rhenium is carried with the precipitate; however, washing removes most of this contamination. Several precipitation cycles and washes should recover > 99% of the technetium. Work is continuing on the optimization of these procedures.

The final step is the mass spectrometry. When the experimentalists began their studies, state-of-the-art techniques would have required a technetium sample of 10^{12} atoms, rendering the experiment impractical. However, rather spectacular improvements have been made by the Los Alamos team. Currently TcO_4^- beams are being produced that are stable for long periods of time (0.5 to several hours) with no contamination from ubiquitous molybdenum, and with rates in excess of 10,000 cps for a 10^{10} atom technetium sample. These parameters permit one to measure $5 \cdot 10^7$ atoms to $\pm$ 0.5% and 10^6 atoms to $\pm$ 10%. The goal of the Los Alamos team has been quantitative mass spectrometry at the 10^7 - 10^8 atom level.

Thus the major technological hurdles appear to have been overcome. A number of questions, however, remain to be resolved before a claim for detecting solar neutrinos can be made:

1) The cross section for neutrino capture on ^{98}Mo has been determined from forward-angle (p, n) calibrations of the ^{98}Tc Gamow-Teller strength. The standard model cross section, due entirely to the capture of ^{8}B neutrinos, is 18 $\pm$ 5 SNU.[8] The 30% uncertainty in this number exceeds anticipated uncertainties arising from the chemistry and mass spectrometry. In principle, (p, n) calibrations of Gamow-Teller strength distributions can achieve an accuracy of 10 to 15%. An improved measurement is clearly needed.

The ^{97}Mo cross section is not known. The lowest Gamow-Teller transitions have thresholds of 536 keV and 644 keV, and can be excited by ^{7}Be, pep, and CNO-cycle

neutrinos. Of course, ^{8}B neutrinos excite many levels. To disentangle these contributions, high-resolution (p,n) measurements, or some direct calibration, will be necessary. With no present information on Gamow-Teller strengths, it is difficult to anticipate the extent of nuclear structure uncertainties for this isotope.

2) The most difficult aspect of geochemical experiments is the demonstration that natural backgrounds are tolerable. The high uranium content of Henderson ore (11 ppm) produces many neutrons and alpha particles. Although (n,γ) and $(n,2n)$ reactions induced by fission neutrons generally overwhelm solar neutrino signals, these reactions don't occur in the present case because of the absence of stable technetium. (This property also greatly simplifies the mass spectrometry.) Molybdenite occurs as exceptionally pure macroscopic crystals in which the metal-to-sulfur ratio is essentially 1:2. Therefore uranium was assumed to reside only outside these crystals in the original background calculations.[1] This assumption is consistent with subsequent measurements of the uranium content of the Henderson molybdenite concentrate.

The principal background reaction producing ^{98}Tc is believed to be $S(\alpha,p)$Cl followed by ^{98}Mo$(p,n)^{98}$Tc. For the standard model ^{8}B flux the predicted signal-to-background ratio is 40. This reaction also occurs for ^{97}Tc, but other backgrounds producing ^{97}Tc may prove more worrisome. Thermal neutron capture on ^{96}Ru followed by the electron capture transition ^{97}Ru$\dashrightarrow{}^{97}$Tc will produce a 10% background (assuming a 10 SNU solar neutrino capture rate) if the Ru content of the concentrate is 0.4 ppm. It remains unclear whether such efficient separation of ruthenium-bearing minerals occurs in the flotation process. The reaction ^{93}Nb$(\alpha,\gamma)^{97}$Tc may be troublesome if the Nb content of the concentrate exceeds 15 ppm. We are concerned that these backgrounds may make the ^{97}Tc results difficult to interpret.

Long-lived ^{99}Tc ($\tau_{1/2}$ = 0.21 Myr) is produced principally by spontaneous fission of ^{238}U residing in the ore. Thus the ^{99}Tc in the ore concentrate should track the radioactivity backgrounds discussed above. A demonstration that ^{97}Tc and ^{98}Tc are not correlated with ^{99}Tc could be viewed as evidence that uranium-induced backgrounds are not significant.

(Presumably the ^{99}Tc content of the concentrate will be determined by local variations in the uranium content of the ore body and by the efficiency of separation for uranium-bearing minerals in flotation.)

Cosmic ray backgrounds were discussed previously. It would be attractive to establish a correlation between cosmic ray production of ^{98}Tc and depth within the ore body. The Henderson ore may be too deep to make this feasible.

3) The geochemical stability of technetium in the reducing environment of the Henderson ore body must be carefully studied. (Technetium is quite mobile in oxidizing environments.) If technetium is lost on time scales of a million years, the experiment is not credible. A check on migration over $\sim 10^5$ years may be possible by demonstrating that ^{99}Tc is in secular equilibrium with the very small amounts of uranium that may exist in the molybdenite.

4) The roaster may retain a "memory" of previous ore. In particular, ore from shallow mines, with elevated levels of cosmic ray-induced technetium, could contaminate the Ft. Madison plant. Both the extent and duration of such contamination must be studied. The AMAX Corporation is willing to schedule ores from successively deeper molybdenite mines while the experimentalists monitor the technetium in the scrub stream.

In summary, the experimentalists are cautiously optimistic that the major technical roadblocks to this experiment have been removed. Before the end of this year the technetium content of Henderson ore may be known. Clearly, the interpretation of this measurement requires careful attention to questions of chemical mobility, dependence of the background on depth and on the geochemical environment, memory effects in the Ft. Madison plant, and nuclear physics uncertainties in the capture cross sections. As these questions can only be answered by careful experimentation, the program of technetium isolation may continue for some time.

I have not mentioned one very interesting sidelight, the suggestion by Cahn and Glashow[9] that chemical isolation of technetium might provide powerful constraints on the abundance of unknown heavy charged particles. Such a heavy "lepton" could bind to

Ru to form a stable nucleus that would exhibit the chemistry of technetium or rhenium while retaining the isotopic distribution of ruthenium. The principal commercial source of rhenium is molybdenum ore. (ReS_2 is often the primary impurity in molybdenite crystals.) Presumably, stable "technetium" would also concentrate in molybdenite. If 10^7 atoms could be detected in Henderson ore, relative abundances of stable "technetium" to Re of 10^{-17} would be measurable. This implies a sensitivity to an average abundance in the earth's crust of one part in 10^{26}. Of course, without some prior knowledge of the mass of the heavy lepton, the mass spectroscopy is significantly more complicated. Nevertheless one hopes that the availability of powerful techniques for performing the large scale chemistry will motivate clever solutions to the remaining analytical problems.

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