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THE ^{205}Tl EXPERIMENT

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ABSTRACT

^{205}Tl has been previously proposed as a geological detector for solar neutrinos, making use of the reaction $^{205}\text{Tl}(\nu, e^-)^{205}\text{Pb}$ with a neutrino threshold of only ≈ 43 keV. We report on an experiment performed to study the feasibility of detecting radioactive ^{205}Pb nuclei ($T_{1/2} = 15$ million years) at very low concentrations using the recently developed technique of accelerator mass spectrometry. Employing the high-energy ion beams of good quality from the UNILAC heavy-ion accelerator at the GSI Darmstadt, we are able to demonstrate a suppression of neighbouring isotopes to better than 1 in 10^{16} and of neighbouring elements to about 1 in 10^3 . While these results are very encouraging, the minimum number of atoms detectable is still severely limited by the efficiency of producing multiply-charged ions from present ion sources. Future improvements in ion-source performance are briefly discussed.

INTRODUCTION

^{205}Tl was proposed as a geological solar-neutrino detector eight years ago by M. Freedman and his collaborators at Argonne.¹ Its attraction comes from an extremely low neutrino threshold (≈ 43 keV) for the $^{205}\text{Tl}(\nu, e^-)^{205}\text{Pb}$ reaction,² and from the long half-life of the ^{205}Pb radioisotope ($T_{1/2} = 15$ million years). A partial level scheme is shown in Figure 1. The low neutrino threshold provides sensitivity to the low-energy part of the solar neutrino spectrum which is dominated by the well-known hydrogen burning reaction $p + p + d + e^+ + \nu + 430$ keV. This leads to a ν -capture rate up to a 100 times higher than that from the $^{37}\text{Cl}(\nu, e^-)^{37}\text{Ar}$ reaction.³ The long half-life has two attractive consequences: i) collecting radioactive ^{205}Pb nuclei over times comparable to their half-life allows accumulation of a large number of ^{205}Pb atoms which can be more easily detected. ii) The long collection time averages over possible short-time fluctuations in the solar neutrino flux.

These properties make ^{205}Tl a potentially interesting solar neutrino detector with features complementary to ^{37}Cl and ^{71}Ga . In particular, if the ^{71}Ga experiment were to reveal a solar neutrino

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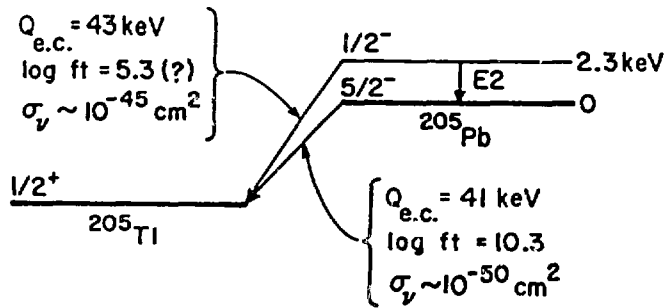


Fig. 1. Partial level scheme⁴ for ^{205}Tl and ^{205}Pb with electron-capture Q -values from reference 2.

deficit similar to the ^{37}Cl experiment, ^{205}Tl might help to distinguish between the possible causes of either short-term fluctuations in the solar neutrino flux or neutrino oscillations and decay.

However, a ^{205}Tl experiment has several severe problems which need to be solved before it can be useful. These regard i) questions of a suitable geological site with known history, ii) possible background nuclear reactions producing ^{205}Pb at such a site, iii) the unknown $\log ft$ value for the first excited state at 2.3 keV excitation energy in ^{205}Tl which, with high probability, dominates the neutrino absorption reaction, and iv) the difficulties in determining the ^{205}Pb content in a geological sample.

The first two, in particular, have been investigated in detail by M. Freedman.⁴ He has proposed what seems to be a suitable geological site, a mine in Macedonian, where the Tl mineral lorandite is rather abundantly found with low (few ppm) Pb contamination. For this particular location, background reactions were calculated to contribute less than 20% to the ^{205}Pb yield. Considerably more uncertainty arises from the unknown $\log ft$ value. Of some promise is the recent success in relating forward angle (p,n) cross sections at high incident energies to Gamow-Teller decay strengths,⁵ although for a $\Delta l = 1$, first-forbidden transition such a relation has not yet been established. The present experiment was directed at the last question, the detection and identification of the ^{205}Pb atoms. Our recent involvement in the technique of accelerator mass spectrometry (AMS) which allows the detection of radioisotopes at extremely low concentrations suggested to us the application of this technique to ^{205}Pb detection.

ACCELERATOR MASS SPECTROMETRY OF ^{205}Pb

In a geological ^{205}Tl experiment rather involved physical and chemical procedures will be used to separate the Tl mineral lorandite (~10-100 kg) from the mining ore (a few tons), and then to extract the Pb fraction, which, anticipating a few ppm Pb contamination in

the lorandite, will result finally in a sample of up to 1 gm Pb containing 10^6 - 10^7 ^{205}Pb atoms. Thus, we expect a Pb sample with a $^{205}\text{Pb}/\text{Pb}$ concentration of the order 10^{-13} to 10^{-14} and possibly a $^{205}\text{Pb}/^{205}\text{Tl}$ concentration of 10^{-3} to 10^{-5} . The latter is expected if chemistry can reduce Tl to ppb levels in the Pb sample. The use of 99.9% enriched ^{203}Tl as a dilutant in a second stage of chemical processing should help in reducing the ^{205}Tl isotope concentration to the required low level.

The recently developed technique of AMS is now routinely used to measure very low concentrations ($<10^{-14}$) of light radioisotopes in various applications e.g., using ^{10}Be , ^{14}C and ^{36}Cl .⁶ However, for the heaviest radioisotopes this is much more difficult and has not yet been achieved. Very briefly, in AMS a high-energy heavy ion accelerator is used to accelerate the radioisotope to energies of the order of MeV/nucleon at which point ions can be unambiguously identified and individually counted by mass and, in particular, by nuclear charge Z. The identification is greatly facilitated by using standard instrumentation and established techniques from nuclear physics heavy ion research or by expanding them. The field of AMS has grown rapidly over the past few years. An overview can be obtained from the proceedings of the three international symposia held on this topic.⁶

To attempt identification of ^{205}Pb atoms by AMS, a large heavy-ion accelerator is necessary in order for the ion to be accelerated to sufficiently high energies where nuclear charge determination via differential energy loss becomes possible. The only such accelerator presently in operation and capable of accelerating the heaviest

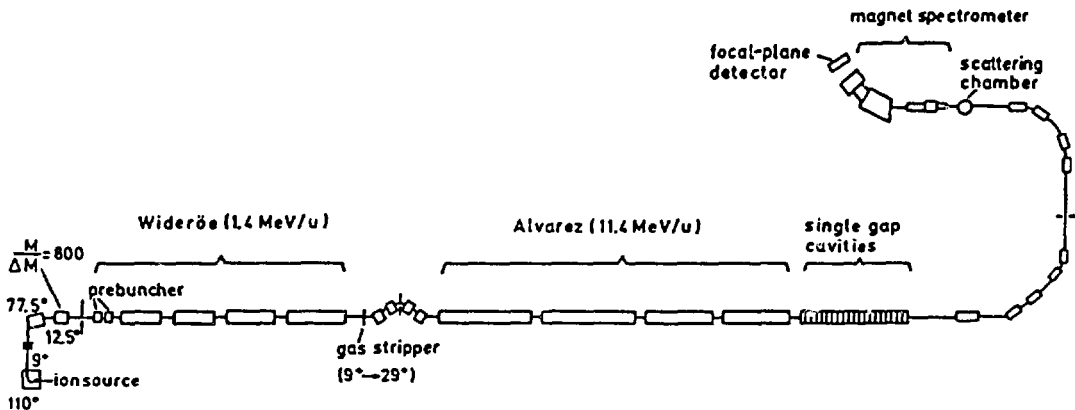
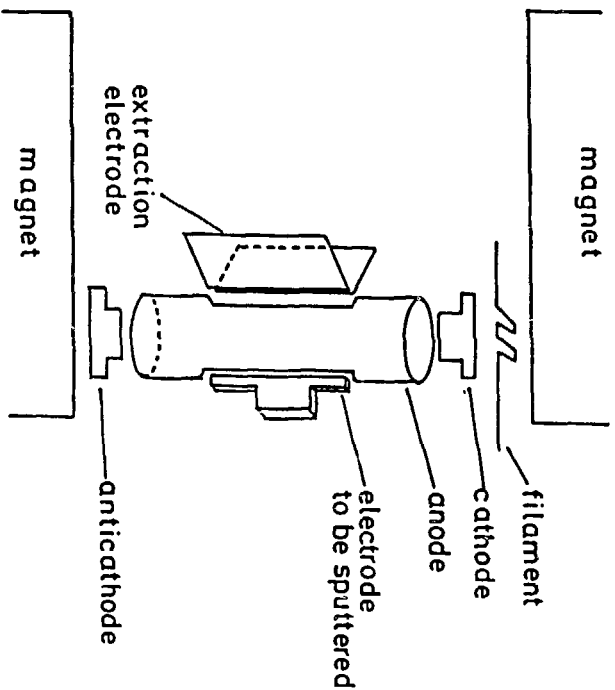


Fig. 2. Schematic layout of the UNILAC accelerator at GSI Darmstadt, with emphasis on those components that are of particular importance in the present accelerator mass spectrometry measurements.

nuclei with good beam characteristics is the UNILAC at the GSI research center in Darmstadt. In the following we briefly describe our AMS measurements using this system.

Figure 2 shows schematically the major components of the UNILAC and the beam transport system used in the present studies. The present system can be viewed as consisting of 3 major components, each of which contributes in a specific way to suppression of unwanted ion species at the final detector: i) the ion-source and injector system with charge-state and mass selection reduce the neighbouring stable isotopes ^{204}Pb , ^{206}Pb , ii) the rf accelerator and beam transport system act as an efficient mass and ion-optical filter in addition to accelerating ions to the necessary kinetic energy, and iii) the heavy-ion detection system allows mass and nuclear charge measurements.

The ion source is a positive-ion penning source pulsed at 50 Hz with a sputter target for the production of the desired ion species. Figure 3 shows a schematic of the source which was originally developed at Dubna.⁷ Ar was used as discharge gas during our experiments. The source was mounted in a 110° magnet. The magnetic field was used for both the penning discharge and the separation of the 9^+ charge state. After preacceleration to 2 MeV the beam was analyzed by a 77.5° and 12.5° magnet with a mass resolution of $M/\Delta M = 800$ at mass 200.



DISCLAIMER

Fig. 3. Positive-ion penning source with sputter target.

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After injection into a system of four Widerøe tanks an energy of 287 MeV was reached. In the following section the ions were stripped in a gas stripper, and an achromatic charge analyzing system selected the 29^+ charge-state for further acceleration. A final energy of 2.3 GeV was reached in the Alvarez tanks. One of the 20 single gap resonators was used as a debuncher, resulting in a reduction of the energy spread from 0.3% to 0.1%. A highly dispersive 180° beam-line system ($p/\Delta p \approx 1000$) transported the beam to the target chamber of the GSI magnetic spectrometer.⁸

For mass and nuclear charge identification the magnetic spectrometer was used in conjunction with a gas absorber positioned in the target chamber in front of the spectrometer and a high resolution detector system as shown in Fig. 4. The flight time of the ions through the spectrometer (≈ 6.4 m path length) was measured between a channel plate start detector at the spectrometer entrance slits and a scintillator foil stop detector in front of the focal plane ionization chamber. The ion chamber provided energy loss and position signals. The elemental (Z) resolution was mainly achieved by a residual energy measurement of the ions following their traversal of the absorber. The residual energy was determined independently from both time-of-flight and magnetic rigidity measurements, with auxiliary gating on the other detector signals. The gas cell (14 cm long) was filled with a few hundred mbar of CH_4 or isobutane. The expected energy-loss difference between ^{205}Tl and ^{205}Pb was calculated from the Northcliffe-Schilling tables⁹ to be about 18 MeV for an energy loss of ~ 1 GeV at an incident energy of 2.3 GeV.

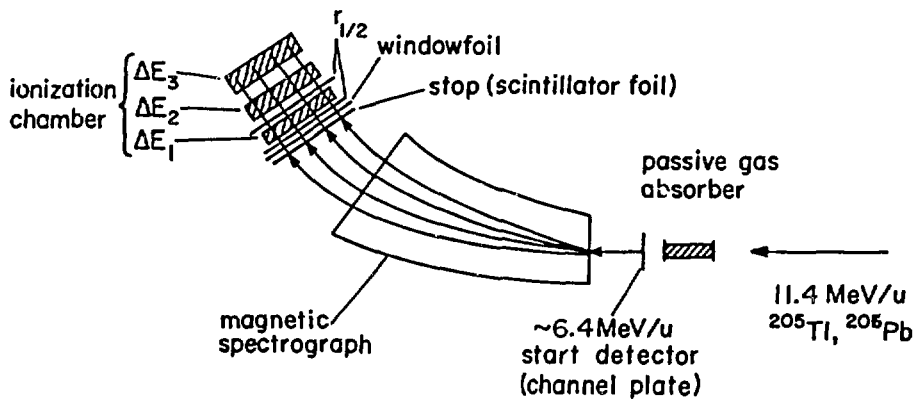


Fig. 4. Schematic of the heavy-ion detection system used in the present measurements. Isobaric ^{205}Pb and ^{205}Tl ions of identical energy experience different energy losses in a gas absorber due to their different nuclear charges. This energy-loss difference is determined by the magnetic spectrograph in a high-resolution measurement of the remaining energy.

RESULTS

In two initial experiments at 2.3 GeV incident energy we investigated the isotopic separation between ^{205}Pb and its stable neighbouring isotopes $^{204},^{206}\text{Pb}$, as well as the elemental separation between ^{205}Pb and ^{205}Tl . Figure 5 shows a spectrum of time of flight vs. magnetic rigidity as measured in the spectrograph. During this run a mixture of natural thallium and lead was used in the source. First the full accelerator system plus heavy ion spectrograph and detectors were carefully tuned for the ^{205}Tl isotope. Then the extraction magnet at the ion source and the injection system for the UNILAC was tuned to mass 206, leaving the rest of the accelerator and beam line system optimized for mass 205. The source currents during these measurements were about 40 nA $^{206}\text{Pb}(9^+)$ and 40 nA $^{205}\text{Tl}(9^+)$, respectively. For the second setting, we measured a count rate for ^{206}Pb ions of 1 Hz and for ^{205}Tl ions of 45 Hz in the spectrograph focal plane detector. Using the previously determined transmission through the UNILAC ($\sim 10^{-3}$) and comparing these count rates the following suppression between neighbouring isotopes is obtained: injection system $\sim 2 \cdot 10^{-6}$, accelerator and beam-line system $\sim 4 \cdot 10^{-8}$. This results in a total suppression between neighbouring masses of $\sim 8 \cdot 10^{-14}$. Time-of-flight and magnetic-rigidity measurement in the

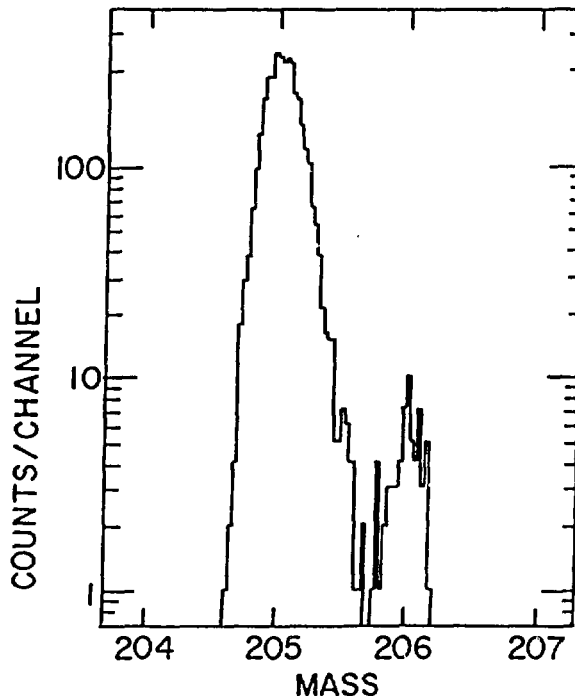


Fig. 5. Mass Spectrum of ^{205}Tl and ^{206}Pb ions as determined from the time-of-flight and magnetic-rigidity signals from the spectrograph focal plane detector.

spectrograph provided for another factor of $\sim 10^{-3}$ resulting in a total suppression between neighbouring masses near $A \approx 200$ of 1 in 10^{16} .

In the second experiment a source sample containing ^{205}Pb and ^{205}Tl in a ratio of $\sim 1:10$ was used (the actual sample composition was $\text{nat}_{\text{Pb}}:\text{nat}_{\text{Tl}}:^{205}\text{Pb} = 1:10^{-3}:10^{-4}$). Taking into account that ^{205}Tl and ^{205}Pb have the same magnetic rigidity and therefore identical energy before they pass through the absorber, we can extract from our data (Fig. 6a) a nuclear charge resolution for $Z = 82$ of $\Delta Z/Z = 10^{-2}$. Making use of the additional information contained in the time-of-flight, energy-loss and total energy signals from the focal plane

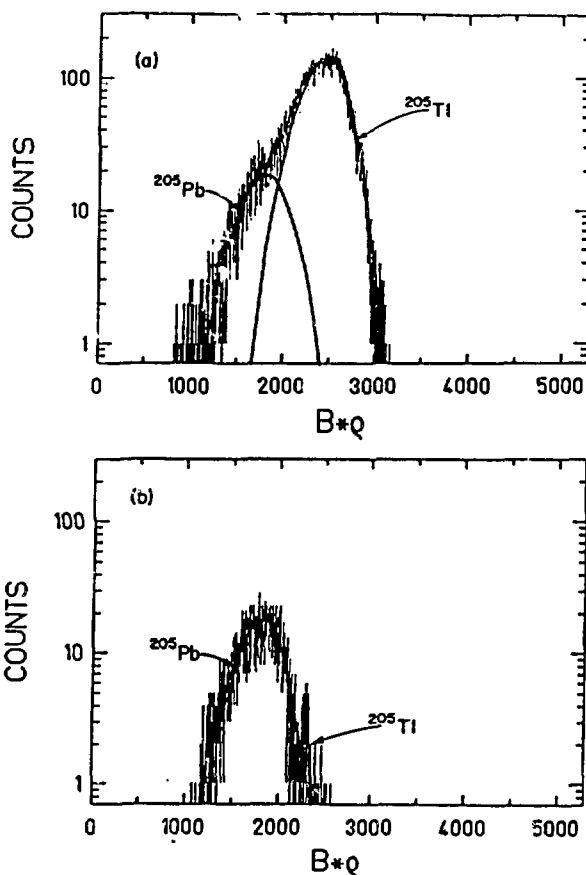


Fig. 6. Magnetic rigidity spectra for ^{205}Pb and ^{205}Tl ions as measured in the spectrograph focal plane detector: a) Total ungated spectrum; b) Spectrum gated with conditions on the various other detector signals for the selection of ^{205}Pb ions. The isobutane gas pressure in the absorber cell was 350 mbar.

detector we are able to achieve a suppression of ^{205}Tl with respect to ^{205}Pb of approximately 1 in 10^3 (Fig. 6b). The fraction of ^{205}Pb ions lost in the gating procedures amounts to less than 8%.

SUMMARY AND CONCLUSION

The suppression of neighbouring isotopes is quite adequate if compared to the requirements defined at the beginning of this paper. On the other hand better charge separation is desirable to further reduce the ^{205}Tl background and to increase the accuracy of the ^{205}Pb identification. This can presumably be achieved by optimizing the absorber thickness with respect to energy straggling versus separation of neighbouring nuclear charges, and by increasing the beam energy. Improving the ΔE signals from the ionization chamber seems also possible by optimizing the gas pressure in the counter.

The major problem identified in our measurements is the very low efficiency of producing Pb^{9+} ions in the present ion source (10^{-6} to 10^{-5}). The transmission of the accelerator and beam-line system and the detection efficiency contribute another factor of $\sim 10^{-3}$ yielding a total efficiency of 10^{-9} to 10^{-8} . Since only a limited number of ^{205}Pb atoms will be available ($\sim 10^7$), the efficiency of the ion source at present is unacceptable. A solution of this problem may be the preacceleration of a low charge state ion (1^+) in conjunction with a stripping process before entering the main accelerator. The production of this ion beam should be attainable with an efficiency of up to 20%. Preacceleration to an energy of 6 MeV can be achieved for example with a Van de Graff accelerator but other preacceleration systems may be used. A stripping efficiency of $\sim 30\%$ for the 8^+ charge state yields a total efficiency of 1.5%, taking into account that only 25% of the beam is accepted by the UNILAC due to its duty cycle.

Another possibility is the use of the recently developed electron cyclotron resonance (ECR) source which provides high charge states with good efficiency.¹⁰ The most attractive feature of these sources seems to be the fact that due to resputtering from the walls of the essentially closed plasma cavity, close to 100% of the original source sample will eventually be emitted as ions.¹¹ The time needed for this "clean-out" seems to be quite tolerable, typically a few hours to a day. After such a modification of the injector the overall efficiency may allow the detection of the expected 100 events in the spectrometer from about 10^7 ^{205}Pb ions in the ion source sample. Assuming a neutrino capture rate of 430 SNU,¹ this corresponds to measuring neutrino-produced ^{205}Pb atoms from 100 kg of lorandite (TlAsS_2) or 20 tons of ore.

From our measurements and the possible improvements, we conclude that accelerator mass spectrometry with a large heavy-ion accelerator seems capable of providing sufficient resolution and sensitivity for

identification of neutrino-produced ^{205}Pb . Since, however, both the hitherto unknown neutrino capture cross section and a possible lower solar neutrino flux could result in a considerable lower SNU value, an overall efficiency greater than the estimated 10^{-5} would be desirable. It may not be unreasonable to assume that future developments in ion source, accelerator and detector technology will eventually bring the efficiency up to the 10^{-3} range, already routinely achieved for accelerator mass spectrometry of lighter ions such as ^{14}C .

Operation of a large accelerator like the UNILAC for an experiment in accelerator mass spectrometry is a very difficult task. We would like to express our appreciation to Dr. N. Angert and the operating staff of the UNILAC accelerator for their continuous support and their patient endurance of a multitude of requests from sometimes not so patient experimenters.

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