Determination of Th, U and K in ultrapure Li2CO3 by Inductively Coupled Plasma Mass Spectrometry for neutrino-less double beta decay experiment

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Date: 2025-03-14T00:00:00+00:00

Abstract

Li2CO3, a raw material of Li2MoO4 crystal used for searching the neutrino-less double beta decay (0 $\beta\beta$), must be of high purity and low content of Th, U and K. In this study, quantitative methods were developed to determinate the content of Th, U and K in ultrapure Li2CO3 samples by inductively coupled plasma mass spectrometry (ICP-MS). The research showed the signals of Th, U and K were significantly inhibited with the increasing of lithium concentration. When the lithium concentration reached 8000 ug g-1, the recovery of Th, U and K decreased to 10 ~ 30 %. For the determination of Th and U, both the recovery correction method and the TEVA - UTEVA combined resin method can eliminate the influence of the lithium matrix. The recovery correction method was simpler to operate, while the TEVA - UTEVA combined resin method was lower in detection limit. For measurement of K, the plasma gas (Ar) in the ICP - MS also induced significant interference. Compared with the standard measurement mode of ICP - MS, the cold flame mode with reduced radio frequency (RF) power can effectively reduce the interference from Ar, greatly improving the detection sensitivity of K. Meanwhile, the detection limit of K in Li2CO3 samples can be as low as 7.53 ng g-1, correcting the inhibitory effect of the lithium matrix by the recovery correction method. A neutron activation analysis (NAA) measurement was also performed for the comparison, yielding results consistent with ICP-MS.

Full Text

Preamble

Determination of Th, U and K in Ultrapure Li_2CO_3 by Inductively Coupled Plasma Mass Spectrometry for Neutrino-less Double Beta Decay Experiments

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Abstract

Li_2CO_3, a raw material for Li_2MoO_4 crystals used in neutrino-less double beta decay ($0\ \beta\beta$) searches, must be of exceptionally high purity with minimal Th, U, and K content. This study developed quantitative methods for determining Th, U, and K concentrations in ultrapure Li_2CO_3 samples using inductively coupled plasma mass spectrometry (ICP-MS). The research demonstrated that signals for Th, U, and K were significantly suppressed as lithium concentration increased. When lithium concentration reached 8000 g g^{-1}, recoveries for Th, U, and K decreased to 10–30%. For Th and U determination, both the recovery correction method and the TEVA-UTEVA combined resin method effectively eliminated lithium matrix effects. The recovery correction method offered simpler operation, while the TEVA-UTEVA combined resin method provided lower detection limits. For K measurement, plasma gas (Ar) in ICP-MS caused significant interference. Compared with standard ICP-MS measurement mode, the cool flame mode with reduced radio frequency (RF) power effectively reduced Ar interference, substantially improving K detection sensitivity. Using the recovery correction method to compensate for lithium matrix suppression, the detection limit for K in Li_2CO_3 samples reached as low as 7.53 ng g^{-1}. Neutron activation analysis (NAA) measurements performed for comparison yielded results consistent with ICP-MS.

Keywords: Th, U and K; Li_2CO_3; Li_2MoO_4 crystal detector; Quantitative method; Neutrino-less double beta decay

Introduction

The international search for neutrino-less double beta decay (0 $\beta\beta$) is now regarded as the most promising approach to explore the mysterious nature of neutrinos following the discovery of neutrino masses in oscillation experiments. After several decades of development, large-scale 0 $\beta\beta$ experiments have adopted several technical approaches, including GERDA [1], MAJORANA [2], and LEGEND [3] using high-purity germanium detectors; KamLAND-Zen [4] using liquid scintillator; EXO-200 [5] and nEXO [6] using liquid xenon time projection chambers (TPC); NEXT [7], PandaX-III [8], and NvDEx [9] using high-pressure gas TPC; and CUORE [10] and CUPID [11] using cryogenic crystal bolometers. CUORE (Cryogenic Underground Observatory for Rare Events) is a ton-scale cryogenic detector located at Laboratori Nazionali del Gran Sasso (LNGS) in Italy searching for 0 $\beta\beta$ decay in ^{130}Te, achieving a background level of (1.38$±$0.07) $\times$ 10^{-2} counts/(keV $\cdot$ kg $\cdot$ year) in the 0 $\beta\beta$ decay region of interest [12]. CUPID, the CUORE Upgrade with Particle Identification, will search for 0 $\beta\beta$ decay of ^{100}Mo and expects to reduce background noise by two orders of magnitude through optical and thermal dual-channel readout [13,14]. Li_2MoO_4 crystal is considered an excellent candidate because it has a high Q-value, a high molybdenum concentration (55 wt%), and is relatively easy to grow [15]. To avoid interference from natural cosmic rays, the detector must be placed deep underground [16]. Additionally, all materials selected for the detector must be of high purity, particularly regarding Th, U, and K content [17,18].

The decay of Th, U, and their daughter products can generate gamma rays similar to those in neutrino-less double beta decay, increasing the detector background [19-21]. As the primary raw material for Li_2MoO_4 crystal preparation, the Th, U, and K contents in Li_2CO_3 must be strictly controlled.

Several quantitative analytical techniques exist for material purity assay, including gamma spectroscopy [22-24], neutron activation analysis (NAA) [24,25], and inductively coupled plasma mass spectrometry (ICP-MS) [19-21,24,26]. Gamma spectroscopy and NAA are not ideally suited for rapid or accurate determinations. Gamma spectroscopy requires large quantities of material (frequently many kilograms) and prolonged counting times (often months) [20,24]. NAA does not require preliminary sample manipulation, but it is expensive and may create complications through the formation of interfering radionuclides. ICP-MS is considered one of the most powerful analytical methods for trace and ultratrace analysis, offering sub-picogram per milliliter detection limits for U and Th with minimal analysis time.

However, matrix constituents are among the main factors affecting detection limits. High levels of matrix components can cause deposition on the sampler and skimmer cones of ICP-MS and reduce the response signal [19]. Thus, dissolved samples may need dilution to lower total dissolved solids content, which degrades achievable detection limits. In practice, the most frequently used methods in-

volve separating analytes from the matrix through various techniques such as ion chromatography, coprecipitation, extraction, and distillation [27-30]. However, due to process complexity, additional problems arise, including increased contamination risk and extended analysis time.

In the CUPID project, our laboratory is involved in screening Li_2MoO_4 crystals and raw material purity. For this purpose, an ICP-MS approach was selected, and we investigated detection capability, matrix effects, and matrix separation methods. This represents the first comprehensive assessment of ultratrace Th, U, and K content in high-purity Li_2CO_3 using ICP-MS. Meanwhile, to further confirm result reliability, NAA measurements were used as an independent reference.

Instrumentation and Materials

An ICP-MS (Thermo Fisher X series) equipped with a cyclonic glass spray chamber and pneumatic nebulizer was used in this work. Sample weighing was performed using an analytical balance (0.01 mg, METTLER TOLEDO AB265-S). TEVA and UTEVA resin (100–150 m, TRISKEM International) were used for extraction chromatography. Empty column tubes (2 mL, Beijing UDLER Technologies) were used to contain the resins. Polypropylene (PP) tubes (50 mL, Environmental Express) were used for sample dissolution. All dissolution, separation, and dilution procedures were conducted in a class-100 fume hood and a class-1000 clean room.

Chemicals and Reagents

All acids used for dissolution and separation procedures were of super analytical grade (HNO_3 and H_2O_2: TAMA Pure AA-10, Tama Chemical Inc.; HCl: G6, Shanghai Aoban Technology). High-purity water (18.2 $M\Omega \cdot cm^{-1}$) was obtained from a Milli-Q Element water purification system (Millipore). Single-element standard solutions (Th, U, K, and Li) were purchased from SPEX CertiPrep.

Investigation of Lithium Matrix Effects

High-concentration lithium standard solution (10,000 g g^{-1}) and Li_2CO_3 samples were diluted stepwise to obtain lithium matrix solutions with different lithium concentrations (1–8000 g g^{-1}). Th, U, and K standard solutions were added simultaneously to achieve final concentrations of approximately 1 ng g^{-1} each. The influence of lithium and Li_2CO_3 matrix on Th, U, and K was investigated through recovery measurements.

Recovery Correction Method

Approximately 1 g of Li_2CO_3 sample was dissolved and diluted stepwise, with final lithium concentrations ranging from 1 to 8000 g g^{-1}. The specific concentration range was determined based on the actual Th, U, and K contents

in the samples. Each sample at every concentration level was prepared in two aliquots: one serving as the sample solution and the other spiked with Th, U, and K standard solutions to evaluate recovery.

The Th, U, and K contents in samples were calculated using the following formula:

$$C = \frac{\sum_{i=1}^{n} \frac{C_i \cdot D_i}{R_i}}{n}$$

where C (pg g^{-1}) is the Th, U, or K content in the sample; C_i (pg g^{-1}) is the measured concentration of Th, U, or K in a particular diluted Li_2CO_3 sample; D_i is the dilution factor; R_i (%) is the recovery of Th, U, or K in that diluted sample; and n is the number of diluted Li_2CO_3 samples.

Separation and Pre-concentration Using TEVA-UTEVA Combined Resin

Approximately 2 g of sample was mixed with 10 mL of 8M HNO_3 in a 50 mL PP tube. To prevent violent reaction during dissolution, samples were moistened with a small amount of water after weighing, then dissolved by dropwise addition of HNO_3 solution. After complete dissolution, the solution underwent separation and pre-concentration following the procedure described in Ref. [29].

TEVA and UTEVA resins were loaded into empty column tubes with a bed volume of 2 mL each. The resins were prewashed with 5M HCl and 0.1M HCl separately, then conditioned with 3M HNO_3. Sample solutions passed successively through TEVA and UTEVA columns. Th retained on TEVA resin was eluted with 5 mL of 5M HCl, and U retained on UTEVA resin was eluted with 5 mL of 0.1M HCl. The combined Th and U eluates were treated with 0.5 mL H_2O_2 and 0.5 mL 3M HNO_3, then heated at 100°C for resin digestion and HCl removal. The final solutions were weighed and used for Th and U measurement.

ICP-MS Measurement

Th and U measurements were performed in standard mode, while K measurements were conducted in both standard and cool flame modes. Operating parameters are summarized in Table 1 . No significant day-to-day instrument performance variations were encountered during this work. Th, U, and K were quantified using external standard calibration. Calibration solutions of 0–10 ng g^{-1} were used for Th, U, and K matrix investigations and for K measurement in actual samples. Calibration solutions of 1–10 pg g^{-1} were used for Th and U measurement in actual samples. Calibration and sample solutions were measured alternately.

NAA Measurement

For comparison, a Li_2CO_3 sample from the same production batch as those analyzed by ICP-MS was measured using NAA. A total of 4.98 g of Li_2CO_3 powder was placed in a clean polyethylene vial and irradiated with neutrons at the TRIGA Mark II research reactor in Pavia, Italy, for six consecutive hours. The irradiation was conducted alongside K, U, and Th standards for calibration. After irradiation, the sample and standards were analyzed using gamma spectroscopy at the Radioactivity Laboratory of the Physics Department of University of Milano Bicocca, Italy, to detect the activated isotopes ^{42}K, ^{239}Np, and ^{233}Pa. These measurements were used to determine the concentrations of their respective precursors, ^{41}K, ^{238}U, and ^{232}Th, in the powder. The concentration of ^{40}K was then determined from ^{41}K by rescaling according to their respective isotopic abundances. A detailed discussion of the irradiation procedure and methods is beyond the scope of this paper; the methodology is thoroughly described in Ref. [31]. The results are reported in the next section.

Results and Discussion

Detection Limits of ICP-MS

Since Li_2CO_3 serves as the raw material for Li_2MoO_4 crystals, extremely low Th, U, and K contents are required. Therefore, the detection limits of the ICP-MS instrument itself needed evaluation. In this work, 2% (V/V) HNO_3 was used as the evaluation solution, and three times the standard deviation of ten parallel measurements was taken as the ICP-MS detection limit. The detection limit results are shown in Table 2 . The ICP-MS detection limits for Th and U were 0.035 pg g^{-1} and 0.025 pg g^{-1}, respectively, proving that measurement of Th and U in ultrapure Li_2CO_3 by ICP-MS is feasible. In standard mode, the detection limit for K was 1.16 ng g^{-1}, while in cool flame mode it was reduced by more than two orders of magnitude to 0.008 ng g^{-1}. When measuring K by ICP-MS, the primary interference originates from plasma gas (Ar), specifically tailing from ^{40}Ar interfering with ^{39}K. In cool flame mode, reduced RF power decreases Ar ionization efficiency and background signal, while K remains essentially unaffected due to its low ionization energy, thereby improving detection capability. These results demonstrate that cool flame mode is more suitable than standard mode for K measurement in ultrapure Li_2CO_3, and it was adopted for subsequent work.

Evaluation of Lithium Matrix Effects

During Li_2CO_3 sample dissolution, CO_2 escapes, leaving lithium ions as the main matrix component. Therefore, the lithium matrix effect was first investigated using a lithium single-element standard solution. To further confirm the Li_2CO_3 matrix influence, Li_2CO_3 samples were also dissolved and diluted for investigation. Figure 1 [Figure 1: see original paper] shows that the matrix effect from Li_2CO_3 sample solutions on Th, U, and K was very similar to that from

lithium standard solutions, demonstrating that Th, U, and K measurements in Li_2CO_3 samples are primarily affected by the lithium matrix. As lithium concentration increased, signals for Th, U, and K were significantly suppressed. The suppression trends for Th and U were essentially consistent with increasing lithium concentration, but K showed distinctly different behavior. When lithium concentration was below 250 g g^{-1}, Th and U recoveries exceeded 70%, then declined rapidly thereafter. At 8000 g g^{-1} lithium concentration, Th and U recoveries decreased to 10–30%. The lithium matrix interference on K signals was substantially greater than on Th and U signals; at 250 g g^{-1} lithium concentration, K recovery had already dropped to 10–20%. Moreover, within the 0–250 g g^{-1} lithium concentration range, K showed a continuous, steep decline. Since Li and K are both alkali metals with similar chemical properties and ionization energies, the lithium matrix caused severe K interference. While dilution could reduce lithium matrix suppression, it would severely compromise ICP-MS detection capability. Due to significant chemical property differences between Li and Th/U, lithium matrix separation was investigated for Th and U measurements. However, because Li and K have extremely similar chemical properties and K is highly prone to contamination, no separation methods were employed for K in this work.

Based on ICP-MS detection limits and recoveries at different lithium matrix concentrations, the detection limits of the recovery correction method were evaluated using Eq. 1. The specific results are also shown in Table 2: Th: 5.54 pg g^{-1}, U: 3.94 pg g^{-1}, and K: 7.53 ng g^{-1}. This K detection limit represents the lowest achievable in this work.

Influence of Th and U Spike Concentrations on Recovery

In recovery experiments, the spike amount should ideally approximate the analyte concentration. However, due to extremely low Th and U contents in ultrapure Li_2CO_3 samples, spiking at actual sample concentrations would increase uncertainties during sample preparation and measurement. Therefore, the influence of different Th and U spike concentrations on recovery was investigated.

Two different spike concentrations were evaluated: one close to the sample content (~5 pg g^{-1}) and another much higher than the sample content at a routine ICP-MS detection level (~1 ng g^{-1}). For the 5 pg g^{-1} spike, calibration curves of 0, 0.5, 1, 5, 10 pg g^{-1} and 0, 0.5, 1, 2, 5, 10 ng g^{-1} were used. For the 1 ng g^{-1} spike, only the higher concentration calibration curve was used. Figure 2 [Figure 2: see original paper] shows the recovery results for the two different spike concentrations. The obtained recoveries were essentially identical even when different calibration curves were used, indicating that Th and U recoveries were not affected by spike concentration. This may be because within the 0–10 ng g^{-1} range, Th and U signal intensities exhibit excellent linear relationships with concentration, as shown in Figure 3 [Figure 3: see original paper]. Therefore, within this concentration range, recovery is independent of concentration. This

result indicates that in the recovery correction method, ng g^{-1}-level spikes of Th and U can be directly used to measure recovery ( ). Consequently, there is no need to determine the actual Th and U concentrations in ultrapure Li_2CO_3 samples beforehand, which greatly reduces sample preparation and measurement complexity.

Evaluation of TEVA-UTEVA Combined Resin

UTEVA resin is widely used for separation and pre-concentration of Th and U in samples, particularly suitable for U [19,20,32-35]. When used for Th, oxalate reagents are typically required to elute Th adsorbed on the resin [19,32], increasing contamination risk and yielding poor Th recovery [19]. Yuan et al. [29] used both TEVA and UTEVA resins to separate Th and U in copper samples, achieving low detection limits. Considering that TEVA and UTEVA resins have good adsorption and elution characteristics for Th and U in copper samples, respectively, and only high-purity acids (HNO_3 and HCl) are used from sample loading to elution, this method was applied in this work.

Due to the complexity of the Th and U separation and pre-concentration process, procedural contamination and analyte loss are key factors affecting method accuracy. In this work, procedural contamination was evaluated through process blanks and method detection limits, while analyte loss was assessed through recovery measurements due to the lack of suitable isotope tracers in our laboratory.

In the separation and pre-concentration process, blanks containing all reagents were prepared simultaneously and run in parallel with samples. This allowed evaluation of background contamination from the entire separation procedure. The method detection limit was also determined as three times the standard deviation of ten parallel blank measurements. The specific results are shown in Table 2. After TEVA-UTEVA combined resin separation and pre-concentration, process blanks for Th and U were significantly higher than the instrument background, and detection limits were correspondingly increased. This indicated that Th and U contamination was introduced during the separation process, likely from the resin, containers, and pipettes used in the experiment. All materials were disposable and used without pretreatment. Since the process blank and separation method detection limit remained below current requirements, no further background reduction was attempted.

Blanks spiked with Th and U standard solutions were used to evaluate the separation and pre-concentration efficiency of TEVA-UTEVA combined resins. The specific results are shown in Figure 4 [Figure 4: see original paper]. In blank solutions, Th recovery ranged from 100% to 102%, and U recovery ranged from 90% to 104%, demonstrating that TEVA-UTEVA combined resins effectively separate and pre-concentrate Th and U. To confirm the effect of lithium on TEVA-UTEVA resins, lithium standard solutions (10,000 g g^{-1}) spiked with Th and U standard solutions were processed. The results, also shown in Figure

4, indicate that Th and U recoveries in lithium solution (95–99% for Th and 96–102% for U) were essentially identical to those in blank solution, demonstrating that lithium has negligible effect on TEVA-UTEVA resins.

Analysis of Li_2CO_3 Samples

One Li_2CO_3 sample, whose Th and U contents were below the ICP-MS detection limit when using direct dilution (lithium content: ~200 g g^{-1}) in preliminary measurements, was analyzed by both the recovery correction method and the TEVA-UTEVA method. In the recovery correction method, final lithium concentrations ranged from 500–8000 g g^{-1} for Th and U measurement and 10–8000 g g^{-1} for K measurement. The specific results are shown in Figure 5 [Figure 5: see original paper]. Without recovery correction, apparent Th, U, and K contents in Li_2CO_3 decreased as lithium matrix concentration increased, consistent with lithium matrix effect experiments. However, after recovery correction, Th, U, and K values fluctuated around mean values at different lithium matrix concentrations, proving that recovery correction effectively compensates for lithium matrix suppression.

The Th and U analysis results from different methods are summarized in Table 3 . Although NAA measurements could only assess upper limits, these are in good agreement with ICP-MS results. Additionally, the two ICP-MS-based methods yielded essentially consistent results. The U result was slightly higher with the recovery correction method, likely because after Li_2CO_3 dilution, the concentration approached the ICP-MS lower limit, resulting in larger measurement error.

Table 3. Analysis of Th and U in the Li_2CO_3 sample (Results in pg g^{-1}, mean±SD)

Sample	ICP-MS Recovery Correction Method	TEVA-UTEVA Method
Li_2CO_3	6.49$±1.14(Th), 9.51±1.09(U)|6.89±0.68(Th), 7.89±$0.41 (U)	

The K analysis results are summarized in Table 4 . The ^{40}K result was obtained by NAA, and the natural K value was calculated based on the ^{40}K isotopic abundance (^{40}K abundance: 0.0120%). The NAA and ICP-MS results were essentially consistent. The ICP-MS method showed relatively large standard deviations, likely due to various factors including environmental contamination, recovery errors, and argon interference.

Table 4. Analysis of K in the Li_2CO_3 sample (mean±SD)

Sample	ICP-MS (ng g^{-1})	ICP-MS (pg g^{-1})	NAA (natural K, ng g^{-1})
Li_2CO_3	518$±112|58.3±1.5|486±$13		

Conclusions

Two quantitative methods—the recovery correction method and the TEVA-UTEVA combined resin method—were developed and evaluated for determining U and Th at ppt levels in high-purity Li_2CO_3 samples by ICP-MS. The recovery correction method provides a relatively simple and fast analytical process, but with slightly higher detection limits (Th: 5.54 pg g^{-1}; U: 3.94 pg g^{-1}). It can be used for initial assessment of Th and U content in Li_2CO_3 samples. If Th and U contents are undetectable by this method, the TEVA-UTEVA combined resin method, though more complex, offers lower detection limits (Th: 0.178 pg g^{-1}; U: 0.155 pg g^{-1}) and can provide further confirmation. The TEVA-UTEVA combined resin method is prone to process contamination; theoretically, if procedural contamination can be strictly controlled, detection limits could be lower than those obtained in this work.

For K measurement by ICP-MS, the cool flame mode with reduced RF power is significantly more suitable than standard mode. Using the cool flame mode with recovery correction, the detection limit for K in Li_2CO_3 samples can be as low as 7.53 ng g^{-1}. Compared with NAA results, ICP-MS in cool flame mode with recovery correction is a relatively accurate and reliable method.

Acknowledgements

The authors are grateful to Monica Sisti (Istituto Nazionale di Fisica Nucleare, INFN) for providing the Li_2CO_3 samples and NAA data. The authors acknowledge support from the General Program (Grant No. 11875310) and Program (Grant No. 12141505) of the National Natural Science Foundation of China, and the Strategic Priority Research Program of the Chinese Academy of Sciences (No. XDB34030000).

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Note: Figure translations are in progress. See original paper for figures.