

## REDUCTION OF RADIOACTIVITY IN MOLYBDENUM OXIDE (MoO<sub>3</sub>) POWDER WITH DEEP PURIFICATION METHOD

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### Abstract

Advanced Molybdenum based Rare process Experiment (AMoRE) is going to use molybdenum based crystals to search for neutrinoless double beta ( $0\nu\beta\beta$ ) decay of <sup>100</sup>Mo. Ultimate sensitivity of molybdenum based crystals is limited by internal background impurities in the form of natural radioactive isotopes, such as <sup>208</sup>Tl from the <sup>232</sup>Th chain and <sup>214</sup>Bi among the <sup>226</sup>Ra decay products in the <sup>238</sup>U chain. Therefore, effective removal of those impurities from molybdenum trioxide (MoO<sub>3</sub>) powder, prior to grow the molybdenum based crystals, is very crucial. In the present study, deep purification of MoO<sub>3</sub> powder was conducted with methods, co-precipitation (CaCl<sub>2</sub>), precipitation and combining these two methods together. Contents of Ba, Pb, Sr, Th and U in MoO<sub>3</sub> powder before and after deep purification were measured using ICP-MS analysis. Co-precipitation method was more effective to remove radioactive impurities than precipitation alone. The results of preliminary experiments of combining these two methods together were found to be more effective in removing the impurities than either of the two methods alone. However, further experiments for combined method are ongoing.

Keywords – AMoRE, neutrinoless double beta decay, molybdenum trioxide, co-precipitation, precipitation

## I. INTRODUCTION

The AMoRE is conducting several experiments to determine neutrino mass value and nature and particle type (Dirac or Majorana). Detection of  $0\nu\beta\beta$  decay would confirm effective mass of neutrino and its type (Majorana). Experimental search for  $0\nu\beta\beta$  decay detection requires high energy resolution scintillator crystals and low background detector. Scintillator crystal containing enriched <sup>100</sup>Mo would be one of the best candidates because of the highest value of  $Q_{\beta\beta} = 3034$  keV [1]. The sensitivity of  $0\nu\beta\beta$  decay experiments basically depends on the internal background of a detector which is governed by the purity of material and possibility for selection of background events [2]. To optimize the requirements for the experiment, the raw materials to be used in growing scintillator crystal must be of radiopure; specifically, those radioactive isotopes which contribute around 3034 keV (<sup>208</sup>Tl from the <sup>232</sup>Th chain and <sup>214</sup>Bi among the <sup>226</sup>Ra decay products in the <sup>238</sup>U chain) [3].

Wet chemistry technique like co-precipitation and precipitation are very useful to separate these radioactive impurities from raw powder. The radioactive impurities can be removed from the raw MoO<sub>3</sub> powder, used for scintillator crystal, prior to crystal growing. The “LUMINEU program” also applied the co-precipitation technique to purify MoO<sub>3</sub> powder after sublimation and found that the co-precipitation was effective to remove the impurities, however, the pH and stay time needed to be considered remarkably [4].

Preliminary results of the experiments of combining co-precipitation and precipitation methods together in the present study were found to be more effective in removing the impurities than either of the two methods alone. Therefore, further experiments to validate the results of the combined method in removing the impurities are ongoing.

## II. EXPERIMENTAL

### II.A. Co-precipitation

One hundred grams of MoO<sub>3</sub> (99.95% purity grade) was dissolved in ammonium hydroxide solution and a carrier (CaCl<sub>2</sub>) was added, based on the stoichiometry, to precipitate calcium molybdate (3%) from the analytic solution. After several hours the precipitates were separated using filter paper. The filtrate solution was completely evaporated to get

purified ammonium hepta-molybdates and/or poly-molybdates. The concentration of the impurities of initial raw powder and the final products were investigated using an inductively coupled plasma mass spectrometer (ICP-MS) equipped with a collision cell (Agilent 7900, Agilent Technologies, USA) operating with high-purity helium. Decontamination factors (DF) for barium (Ba), lead (Pb), strontium (Sr), thorium (Th) and uranium (U) were calculated and yield efficiency of molybdenum was determined in order to find out the loss of molybdenum during experiment.

## II.B. Precipitation

MoO<sub>3</sub> powder of same purity grade was dissolved in ammonium hydroxide solution as did in co-precipitation. pH of the solution was adjusted to 2 with hydrochloric acid. Poly-ammonium molybdate (PAM) precipitates were collected by filtration. The concentration of impurities, decontamination factors, and yield efficiency were determined as in co-precipitation method.

## II.C. Combined co-precipitation and precipitation

In combined method 100 g of MoO<sub>3</sub> (99.95% purity grade) was dissolved in ammonium hydroxide solution and a carrier (CaCl<sub>2</sub>) was added, based on the stoichiometry, to precipitate calcium molybdate (3%) from the analytic solution. After several hours the precipitates were separated using filter paper. pH of the filtrate was adjusted to 2 with hydrochloric acid. Poly-ammonium molybdate (PAM) precipitates were collected by filtration. The concentration of impurities, decontamination factors, and yield efficiency were determined as in described in above two methods.

## III. RESULTS AND DISCUSSION

The comparative study of these experiments indicated that co-precipitation method was more effective to remove the impurities than precipitation (Fig.1). The combined method was expected to be the most effective among the three methods applied; however, the result was not as per expectation. The reason behind less effectiveness of combined method might be due to contamination in HCl that was added to the filtrate obtained after co-precipitation because nothing else was added to the filtrate before subjecting it for precipitation. The DF for Sr, Ba, Pb, Th, and U after mixing HCl to the filtrate were 1.4, 146, 160, 0.7, and 15, respectively but when precipitation was conducted for the filtrate obtained after co-precipitation, the DF was reduced for Sr, Ba, Pb, and U to 1, 2, 82, and 3, respectively but increased for Th (Fig .1.). To confirm this contamination in HCl further experiments on combined method were already been repeated in which the ultrapure HCl was used. The ICP-MS analysis is ongoing and the result will confirm the contamination and expected efficiency of combined method for separation of radioactive impurities from raw powder.

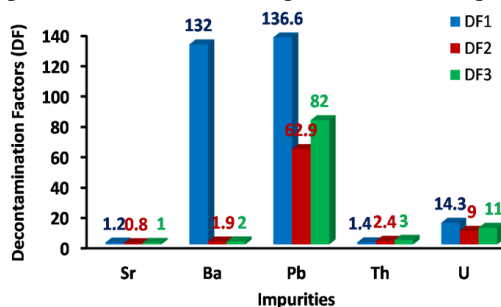


Fig.1. Decontamination factors for co-precipitation (DF1), precipitation (DF2) and combined method (DF3).

## IV. CONCLUSION

Three different wet chemistry technique were applied to separate radioactive impurities from the raw MoO<sub>3</sub> powder. Among them co-precipitation method was found to be more effective. However, the combined method was expected to be most effective but some contamination had immersed during experiment. To confirm the contamination, combined method experiment has already been repeated. The yield efficiency of each experiment was more than 90%. Combined method is expected to be optimum to separate impurities to get the required level of radiopure MoO<sub>3</sub> for AMoRE.

## REFERENCES

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