AMoRE experiment: a search for neutrinoless double beta decay of 100Mo with $^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic scintillation detector

V.N.Kornoukhov
ITEP (Moscow)
on behalf of AMoRE Collaboration

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AMoRE Collaboration: since October 2009
(Advanced Mo based Rare process Experiment)

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  Kyungpook national University: H.J.Kim, J.So, Gul Rooh, Y.S.Hwang (4)
  

- Russia (16)
  
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- Ukraine (11)
  

- China (3)
  
  Tsinghua University: J.Li, Y.Li, Q.Yue (3)

4 countries
8 institutes
69 participants
Goals of AMoRE Collaboration

• Investigation of neutrinoless double beta decay of Mo-100 isotope:
  \[ Q_{\beta\beta} = 3034 \text{ keV}: \text{the biggest of the DBD isotopes can be produced at tenths – hundred kg scale} \] (by centrifuges, in Russia);
  \[ T_{1/2}(2\nu) = 7,1\times10^{18} \text{ years} \rightarrow \text{need for high energy resolution}. \]

• Dark Matter Search
AMoRE: cryogenic scintillation detector based on CaMoO$_4$ single crystal

- CaMoO$_4$ sheelite-type self-activated scintillator
- $T_{\text{melt}} = 1445 \, ^\circ\text{C}$ (Pt or Ir crucible)
- Technology: Chochralsky method
- Light yield (RT): up to 9300 photon/MeV
- Emission peak at 9 K: 540 nm
- Kinetics of scintillation light (main component):
  - under RT = 16 µsec
  - at 6 K = 345 µsec
- Debaye temperature: 438 K (Ge: 360 K, Si: 625 K)
$^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic scintillation detector as a tool for Mo-100 DBD search

“Detector = Source”: $\varepsilon \sim 80$ - 90% efficiency;

- High content of working isotope (Mo) in compound: 50% (stoichiometry ratio);
- **Good energy resolution** and high light yield for scintillation mode;
- **High energy resolution** for phonon mode (comparable with the resolution for HPGe detectors): no $2\nu\beta\beta$ of Mo–100 background;
- $\alpha/\beta$- pulse shape discrimination ($\alpha/\beta$-ratio = 0.20)
- Scalability of the experiment (increasing of mass by break-in “crystal by crystal”);
- Chochralsky technology of the production $\rightarrow$ High purity, including radioactive isotopes
- Mo-100 production at **hundred kg scale**: centrifuges
AMoRE:

$^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic scintillation detector

1. Absorber:
   - Isotope enrichment
   - Purification
   - Crystals growing
   - Isotope recovery from waste

2. Bolometer: R&D on
   MMC & SQUID
   TES & SQUID

Light detector →

Si or Ge  TES/MMC/NTD

CaMoO$_4$

Phonon sensor (MMC)
- Dimensions of CaMoO$_4$ crystals up to 15 cm$^3$;
- $\text{LY} \sim 400$ photons/MeV;
- Low transparency (absorption band at 395 nm was observed).
- High content Bi-214(U-238) = 286 mBq/kg
- Th-232(Tl-208) $\leq 25$ mBq/kg
The results: 2007

The scintillation natural CaMoO$_4$ crystal of cylindrical geometry with dimensions of D40x40 mm
Scintillator element $^{40}\text{Ca}^{100}\text{MoO}_4$

D(42 x 40) x 42 mm, m = 260 g

$M = 815 \text{ gr}$

Annealing →
JSC FOMOS Materials (Moscow): Equipment for crystal growth, heat and mechanical treatment

Puller KRYSIAL 2M
Furnace
Saw
Flow chart of the crystal growth process.” One-by-one” option.

Pellets

Crystallized initial charge manufacturing

Crystal mass 550 g

Melt mass 1100 g

End crystal growing

Melt mass 1100 g

Crystal mass 550 g

Pellet mass 550 g

Melt mass 1100 g

Total initial charge – 1650 g
End crystal mass – 550 g
Waste for recuperation – 1060 g
Losses – 40 g (possible lower)

End crystal mass – 550 g
Waste 530 g

Waste 530 g
**Bkg measurements at Baksan Neutrino Observatory:** HPGe detectors (3 x ~1 kg) at low background underground lab (H = 660 m.w.e.)

<table>
<thead>
<tr>
<th>Sample, material</th>
<th>Isotope</th>
<th>Specific activity, Bq/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mo oxide, $^{100}$MoO$_3$</td>
<td>$^{40}$K</td>
<td>$(5.3\pm0.8)\times10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>$^{228}$Ac = $^{232}$Th</td>
<td>$\leq 3.8\times10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>$^{208}$Tl [(232Th)]*</td>
<td>$\leq 1.0\times10^{-3}$ $[\leq 2.8\times10^{-3}]$</td>
</tr>
<tr>
<td></td>
<td>$^{214}$Bi = $^{238}$U</td>
<td>$\leq 2.3\times10^{-3}$</td>
</tr>
<tr>
<td>Calcium carbonate, $^{40}$CaCO$_3$</td>
<td>$^{40}$K</td>
<td>$(7.3\pm3.1)\times10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>$^{228}$Ac = $^{232}$Th</td>
<td>$(1.6\pm0.2)\times10^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$^{208}$Tl [(232Th)]*</td>
<td>$(4.4\pm3.6)\times10^{-3}$ $[(1.2\pm1.0)\times10^{-2}]$</td>
</tr>
<tr>
<td></td>
<td>$^{214}$Bi = $^{238}$U</td>
<td>$(2.6\pm0.2)\times10^{-1}$</td>
</tr>
<tr>
<td>Single crystal SB-29 $^{40}$Ca$^{100}$MoO$_4$</td>
<td></td>
<td>$\leq 1.2\times10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>$^{228}$Ac = $^{232}$Th</td>
<td>$\leq 3.1\times10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>$^{208}$Tl [(232Th)]*</td>
<td>$\leq 8.3\times10^{-4}$ $[\leq 2.4\times10^{-3}]$</td>
</tr>
<tr>
<td>Calcium formate, Ca(HCOO)$_2$</td>
<td></td>
<td>$\leq 7.0\times10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>$^{228}$Ac = $^{232}$Th</td>
<td>$\leq 3.0\times10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>$^{208}$Tl [(232Th)]*</td>
<td>$\leq 8.9\times10^{-4}$ $[\leq 2.5\times10^{-3}]$</td>
</tr>
</tbody>
</table>

Ac228 $\leq 3.1$ mBq/kg  
TI208 $\leq 0.83$ mBq/kg  
Bi214 (Ra226) $\leq 6.4$ mBq/kg

**Double crystallization procedure:** Purification $\geq 35$ times!
Background measurement at YangYang of SB28 $^{40}\text{Ca}^{100}\text{MoO}_4$ crystal

$\beta-\alpha$ decay in 238U
$^{214}\text{Bi}$ (Q-value : 3.27-MeV) $\rightarrow$ $^{214}\text{Po}$ (Q-value : 7.83-MeV) $\rightarrow$ $^{210}\text{Pb}$

$\alpha-\alpha$ decay in 232Th
$^{220}\text{Rn}$ (Q-value : 6.41-MeV) $\rightarrow$ $^{216}\text{Po}$ (Q-value : 6.91-MeV) $\rightarrow$ $^{212}\text{Pb}$

$\pm 1$ sigma of energy cut was used to analyze the internal background.

Preliminary

$^{216}\text{Po}$ : 1.61-MeV, 21 events $\approx 0.026\text{mBq/kg}$
$^{214}\text{Po}$ : 1.93-MeV, 34 events $\approx 0.043\text{mBq/kg}$

$^{220}\text{Rn}$ : 1.45-MeV
$^{214}\text{Bi}$ (Q-value : 3.27-MeV)
Measurements of the energy resolution at YangYang laboratory
(Example: S35 element at 661 keV (Cs-137).

First enriched 40Ca100MoO4 crystals tested at YangYang:
SB28: D(50x41 mm) x 26 mm, m =196 g
SB29: D(48x51 mm) x 50 mm, m =390 g
S35: D(44x40 mm) x 42 mm, m =259 g
Mo-100 isotope production:
ECP (Zelenogorsk, Russia)

• We bought $^{100}\text{MoO}_3$ oxide (2.5 kg of Mo-100)

Enrichment:
Mo-100 = 96.1%

Impurities (the results from ICP MS measurements):
- $U \leq 0.07$ ppb
- $Th \leq 0.1$ ppb

HPGe (Baksan):
- $^{226}\text{Ra} \leq 2.3$ mBq/kg,
- $^{228}\text{Ac} \leq 3.8$ mBq/kg

Regular productivity of Mo-100 is up to $\sim 30$ kg per year
Svetlana Department, ECP

Anatoly Shubin (1939 - 2008)
Ca-40 isotope production in Russia
Electrochimiprobor

$^{40}$CaCO$_3$ compound: 1.25 kg of Ca-40

**Enrichment:**

Ca-40 = 99.964 +/- 0.005%
Ca-48 < 0.001% (K depletion is 187 times)

**Impurities (ICP-MS):**

U \leq 0.2 ppb
Th \leq 0.8 ppb

**HPGe (Baksan):**

$^{226}$Ra = 260 mBq/kg,
$^{228}$Ac = 160 mBq/kg
The industrial separator SU20

30 kg of Ca-40 ($^{40}\text{CaCO}_3$) is available now
Ca-48 < 0.001%
Good for 150 kg of $^{40}\text{Ca}^{100}\text{MoO}_4$

Productivity: 4 – 5 kg/year
Ca-48 Enrichment/Depletion at KAERI
(Korean Atomic Energy Research Institute)

- **ALSIS (Advanced Laser Stable Isotope Separation)**
  - Features: Isotope-Selective Optical Pumping (ISOP) followed by Non-selective Resonant Photoionization (RPI)
  - ISOP gives good isotope-selectivity and non-selective RPI high yield.

Engineering Demonstration (2010~2012)
- **Production capability : 1kg/yr**

Production Demonstration for (2013 – 2014)
- **Production capability : 5 kg/yr**
AMoRE:

$^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic scintillation detector

1. **Absorber:**
   - Isotope enrichment
   - Purification
   - Crystals growing
   - Isotope recovery from waste

2. **Bolometer:** R&D on
   - MMC & SQUID
   - TES & SQUID

Light detector $\rightarrow$

Si or Ge  TES/MMC/NTD

CaMoO$_4$

Phonon sensor (MMC)
Magnetic material (Au:Er) in dc SQUID junctions

Au:Er (10~1000ppm)  
paramagnetic system  
metallic host: fast thermalization ( ~ 1ms)  
Can control heat capacity by magnetic field

$g = 6.8$  
$\Delta \varepsilon = 1.5 \, \mu\text{eV}$  
$1 \text{keV} \rightarrow 10^9 \text{spin flips}$

• Fast  
• Wide working temperature  
• Absorber friendly
**AMoRE cryogenic scintillation detector: R&D at KRISS (Korea)**

- **Emission line of Mo:** 18 keV
- **FWHM = 11.2 keV**
- **FWHM = 1.7 keV**
- **$E_{\alpha} = 5.5$ MeV**
- **$E_{\gamma} = 60$ keV**
- Crystal size: $\sim 1 \text{ cm} \times 0.7 \text{ cm} \times 0.6 \text{ cm}$
- $\sim 500 \mu\text{m}$ thick brass

For details: Astropart. Physi. 34, 732, 2011
New sensor for large heat capacity

Meander is made in U. of Heidelberg.

2.5 x 2.5 x 0.07 mm³ gold foil

C = 0.6 nJ/K at 20 mK

60 cm³ CaMoO₄
C = 0.17 nJ/K at 10 mK
1.4 nJ/K at 20 mK
KRISS (Korea): **July 2011:**

Experimental setup (crystal D40 x 40 mm)

- **241Am source**
- **teflon-coated copper springs**
- **meander MMC from Heidelberg**
- **thermalization pad (gold film)**
YangYang underground laboratory
YangYang underground laboratory

<table>
<thead>
<tr>
<th>Depth</th>
<th>700 m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature</td>
<td>20 ~ 25 °C</td>
</tr>
<tr>
<td>Humidity</td>
<td>35 ~ 60 %</td>
</tr>
<tr>
<td>Composition of the rock</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{238}$U $\leq 0.5$ ppm</td>
</tr>
<tr>
<td></td>
<td>$^{232}$Th $5.6 \pm 2.6$ ppm</td>
</tr>
<tr>
<td></td>
<td>$^{40}$K $270$ ppm</td>
</tr>
<tr>
<td>Muons flux</td>
<td>$2.7 \times 10^{-7}$ /cm$^2$/sec</td>
</tr>
<tr>
<td>Neutrons flux</td>
<td>$8 \times 10^{-7}$ /cm$^2$/sec</td>
</tr>
<tr>
<td>Content of $^{222}$Rn</td>
<td>$1\sim2$ pCi/L</td>
</tr>
</tbody>
</table>
AMORE: CaMoO$_4$ DBD projected sensitivity

100 kg $^{40}$Ca$^{100}$MoO$_4$ Cryogenic detector
Mo-100 ~ 50 kg
Efficiency ~ 0.8
After 5 years: $3 \times 10^{26}$ years ~ 50 meV

Werner Rodejohann, Int. J. M. Phys., 2011
Conclusion

- Large volume $^{40}\text{Ca}^{100}\text{MoO}_4$ crystals with enriched material have been developed (JSC FOMOS Materials, Moscow)
- Big quantity of Ca depleted on Ca-48 and availability of industrial scale production of Mo-100 permits to plan next generation experiment for search of $0\nu2\beta$- decay of Mo-100
- 100kg $^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic detector: $\sim 3 \times 10^{26}$ yrs ($\sim 0.05$ eV)
- Included in the National Facility Road Map (Republic of Korea) and Federal Aiming Program (Russia)
Back up slides
Sketch: Chochralsky method
1 – growing crystal; 2 – seedholder; 3,13 – water-cooling shaft; 4,11 – ceramic plates; 5,7,12 – ceramic tubes; 6 – induction coil; 8 – heat insulation ceramic; 9 – crucible; 10 – melt; 14 - seed

Just grown $^{40}\text{Ca}^{100}\text{MoO}_4$ crystal before annealing
$m = 0.55$ kg, D49 x 42 mm, $L_{\text{cylinder}} = 53$ mm
CMO crystal growth process at JSC Fomos-Materials Co.

Process stages:

1. Initial powder ICP analysis
2. Initial pellets preparation – pellets manufacturing 550g in mass each
3. Initial charge for crystal growing preparation including MoO$_3$ adds - 2 pellets + up to 3 mass % of MoO$_3$
4. Growth of the initial crystallized charge – crystals up to 550 g each
5. Initial crystallized charge for end-crystal growing preparation
6. Crystallizer assembling and end crystal growing
7. Aftergrowing heat treatment of the end crystal – heat treatment in oxidizing atmosphere
8. Mechanical treatment of the end crystal (cutting, lapping and polishing) – manufacturing of the CMO element according to the specification
Principle of gas-centrifuge isotope separation

Centrifuge acceleration: 500 000 g
Rotation ~ 1500 s⁻¹

Cascade: many hundred and even thousand units

Operation life of centrifuge: 30 years
Principle of electromagnetic separation of stable isotopes

Dispersion $d$ of the EM facility (a distance between ions beams):

$$d = 2\Delta R = R(\Delta M/M)$$

A value of $d$ for Ca isotopes (SU 20 facility) is about 25 mm

$$2R = 1800 \text{ mm}$$

The coefficient of the feed material utilization for EM method is in the range of 2 – 20% only

The coefficient of utilization for Ca isotopes is 4,5%
Electromagnet of the SU20 separator
(5-floors building & 3000 tons magnet!)

Fig. 1. Electromagnet of the separator. (1) Housing; (2) separation tank (chamber); (3) electromagnet coils; (4) ion source seats; (5) ion receiver seats; (6) diffusion pumps; (7) observation openings.
SU20: Separation tank \((2 \times 2 \times 5 = 20)\)

- \(R = 90\) cm
- \(U = 30\) kV
Detector composition

- Detector for Neutrinoless Double Beta Decay experiment at few milli-kelvin.
- Large CaMoO₄ single crystal is held by springs of holder. (216g, 4cm diameter, 4cm height, 430K Debye temperature)
- Gold film is evaporated on the crystal for make thermal connection between crystal and MMC. (200nm thickness)
- Metallic Magnetic Calorimeter is used to change thermal signals to magnetic signals.
- Gold wires are bonded on the gold film and MMC for thermal connection. (25μm diameter)
- SQUID is used to change magnetic signals to voltage signals.
- Meander-type heater is evaporated on the crystal surface. (7.355 Ohm at 4K.)

 MMC
 SQUID
 Gold wire
 Gold film