Radiological Evaluation associated to the Mining of Monazite in Central Spain

R. García-Tenorio\textsuperscript{a}, E. Sanz\textsuperscript{b}, E. Burkhalter\textsuperscript{c}, G. Manjón\textsuperscript{a}, I. Vioque\textsuperscript{a} and I. Diaz\textsuperscript{a}

\textsuperscript{a}Department of Applied Physics II, University of Sevilla, Spain
\textsuperscript{b}Geomnia Natural Resources, Madrid, Spain
\textsuperscript{c}Quantum Mineria, Madrid, Spain
A detailed radiological evaluation has been performed associated with the mining of the “Mulas” monazite deposit. This evaluation has been based in the following studies:

a) determination of the activity concentrations of several radionuclides from the Uranium and Thorium series in representative samples of the material mined,

b) construction of an external gamma dose-rate map of the mining area,

c) study of the distribution of the natural radioactivity in the material extracted as a function of the grain size,

d) radon determinations in the area, and

e) laboratory leaching experiments.

Although the rare earth extraction mining is one of the activities recognized in the positive list of NORM activities, all the results allow concluding that the rare earth mining activity performed in Central Spain can be considered as exempted, being not needed the adoption of radiological countermeasures.
“DIMENSION” OF MATAMULAS ORE

Graph showing the relationship between REO grade and ore tonnage for various locations, including worldwide resources, European resources, and current producers.
CRITICAL RAW MATERIALS
MONAZITE CHARACTERISTICS

100% REO

<table>
<thead>
<tr>
<th>Compound</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeO₂</td>
<td>42.93</td>
</tr>
<tr>
<td>Nd₂O₃</td>
<td>27.26</td>
</tr>
<tr>
<td>La₂O₃</td>
<td>15.95</td>
</tr>
<tr>
<td>Pr₆O₁₁</td>
<td>5.77</td>
</tr>
<tr>
<td>Sm₂O₃</td>
<td>4.18</td>
</tr>
<tr>
<td>Gd₂O₃</td>
<td>1.78</td>
</tr>
<tr>
<td>Y₂O₃</td>
<td>0.60</td>
</tr>
<tr>
<td>Dy₂O₃</td>
<td>0.40</td>
</tr>
<tr>
<td>Eu₂O₃</td>
<td>0.40</td>
</tr>
<tr>
<td>Ho₂O₃</td>
<td>0.38</td>
</tr>
<tr>
<td>Tb₄O₇</td>
<td>0.21</td>
</tr>
<tr>
<td>Yb₂O₃</td>
<td>0.06</td>
</tr>
<tr>
<td>Tm₂O₃</td>
<td>0.05</td>
</tr>
<tr>
<td>Er₂O₃</td>
<td>0.03</td>
</tr>
<tr>
<td>Lu₂O₃</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Hardness: 5 – 5.5 Mohs scale

Real Density: 4.65 g/cm³
MONAZITE CHARACTERISTICS

Quantitative results

Weight%

O Ne Al Si P K Fe La Ce Nd

Ca

Full Scale 1291 cts  Cursor: 7.517 keV (54 cts) keV

Spectrum 1
MONAZITE CHARACTERISTICS

Quantitative results:

- Weight%:
  - 0
  - 5
  - 10
  - 15
  - 20
  - 25
  - 30

Elements:
- O
- Al
- Si
- P
- K
- Fe
- La
- Ce
- Nd

Full Scale 824 cts  Cursor: 7.464 keV (28 cts) keV

Spectrum 1
SAMPLING

STEP A

GRADDED APPROACH

STEP B
EXPERIMENTAL TECHNIQUES

GAMMA SPECTROMETRY

SEM - EDX

ALPHA SPECTROMETRY

GAMMA DOSE RATE
**Activity Concentrations (Bq/kg)**

<table>
<thead>
<tr>
<th></th>
<th>Muestra 1</th>
<th>Muestra 2</th>
<th>Muestra 3</th>
<th>Muestra 4</th>
<th>Muestra 5</th>
<th>Muestra 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th</td>
<td>94 ± 12</td>
<td>82 ± 10</td>
<td>91 ± 4</td>
<td>77 ± 5</td>
<td>105 ± 6</td>
<td>66 ± 4</td>
</tr>
<tr>
<td>$^{230}$Th</td>
<td>60 ± 3</td>
<td>55 ± 3</td>
<td>47 ± 4</td>
<td>43 ± 2</td>
<td>50 ± 3</td>
<td>46 ± 3</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>52 ± 12</td>
<td>51 ± 12</td>
<td>46 ± 10</td>
<td>51 ± 10</td>
<td>57 ± 12</td>
<td>42 ± 10</td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>47 ± 3</td>
<td>45 ± 3</td>
<td>49 ± 3</td>
<td>44 ± 4</td>
<td>54 ± 3</td>
<td>49 ± 2</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>740 ± 41</td>
<td>594 ± 37</td>
<td>851 ± 39</td>
<td>824 ± 43</td>
<td>688 ± 32</td>
<td>845 ± 40</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>2.3 ± 0.3</td>
<td>&lt; 1.0</td>
<td>&lt; 1.0</td>
<td>3.1 ± 0.4</td>
<td>3.0 ± 1.0</td>
<td>&lt; 1.0</td>
</tr>
</tbody>
</table>

Quite uniform the radionuclide distribution
- Comparable with the obtained ones in big extensions over the country

The “grey” monazite is enriched in natural radionuclides?
RADIONUCLIDE DETERMINATIONS (II)

\[ ^{232}\text{Th} \quad 30 - 100 \text{ Bq/kg} \]
\[ ^{238}\text{U} \quad 10 - 50 \text{ Bq/kg} \]
EXTERNAL GAMMA DOSE RATE MAP
Monazite separation and concentration

Only physical processes applied
Intermediate fraction less than 5% in weight
Radionuclide Granulometric Distribution

Fracción fina
< 0.5 mm

- $^{232}$Th 100 ± 8
- $^{238}$U 44 ± 12
- $^{234}$U 51 ± 2
- $^{40}$K 590 ± 30

Fracción intermedia
0.5 – 1.5 mm

- $^{232}$Th 630 ± 30
- $^{238}$U 240 ± 20
- $^{234}$U 250 ± 10
- $^{40}$K 750 ± 50

Fracción Gruesa
1.5 mm – 1 cm

- $^{232}$Th 66 ± 4
- $^{238}$U 42 ± 10
- $^{234}$U 38 ± 3
- $^{40}$K 850 ± 40

Esteriles
0.5 – 1.5 mm

- $^{232}$Th 450 ± 20
- $^{238}$U 120 ± 15
- $^{234}$U 150 ± 9
- $^{40}$K 720 ± 40

Concentrado
0.5 – 1.5 mm

- $^{232}$Th 2830 ± 120
- $^{238}$U 890 ± 35
- $^{40}$K 660 ± 40
Negligible radiological impact in the mining process due to external radiation.

Negligible radiological impact in the mining and concentration process due to inhalation. The fine material susceptible to be resuspended/inhaled in the mining and concentration processes “depleted” in natural radionuclides.

Radiological impact due to $^{222}$Rn should be discarded, because the mining and concentration processes are carried out at open-air.

Occupational doses susceptible to be received by the workers in charge of the different concentration processes and of the handling and storage of the concentrates in the plants evaluated as 0.15 – 0.20 mSv/a due to the external radiation. This estimation was performed adopting very conservative assumptions.
Radiometric determinations in
- 5 underground waters collected from wells located in the mining area
- vegetables (lettuces, Spinachs, chards, tomatoes)
- fruits, olives, cereals ......

**DO NOT SHOW ENHANCEMENTS IN NATURAL RADIONUCLIDES IN THE MINING AREA**

Leaching experiments submitting monazite fractions during 24 hours to the action of
- rainwater
- ground water
- 0-16 M HCl

show not detectable activities in the leachates (by alpha and gamma spectrometry)

**MONAZITE QUITE REFRACTORY MATERIAL**
Nd is enriched in the central parts of the particles, whereas the Ce content increases in the opposite direction.
Th internal distribution

Th = 1.0 Wt%  

Th = 0.75 Wt%
The “positive list”

Industrial sectors most likely to require some form of regulatory consideration

<table>
<thead>
<tr>
<th>Uranium mining and processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rare earths extraction</td>
</tr>
<tr>
<td>Thorium extraction and use</td>
</tr>
<tr>
<td>Niobium extraction</td>
</tr>
<tr>
<td>Non-U mining- including radon</td>
</tr>
<tr>
<td>Oil and gas</td>
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<tr>
<td>Production and use of TiO2</td>
</tr>
<tr>
<td>Phosphate industry</td>
</tr>
<tr>
<td>Zircon and zirconia</td>
</tr>
<tr>
<td>Metals production (Sn, Cu, Al, Fe, Zn, Pb)</td>
</tr>
<tr>
<td>Burning of coal</td>
</tr>
<tr>
<td>Water treatment-including radon</td>
</tr>
</tbody>
</table>
Thanks
Gracias