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OPTIMIZATION OF NEUTRON ACTIVATION ANALYSIS OF RARE-EARTH ELEMENTS

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What is Neutron Activation Analysis (NAA)?

- NAA is a method used to determine the element composition of a material
- Process:
  - The material is irradiated, essentially bombarded by neutrons, typically via a reactor
  - The sample then emits a beta particle followed by a sequence of gamma rays, which can then be recorded and used to identify the specific isotope and elemental composition
  - The amount of that element is then determined by comparing the sample mass values to known values of mass for the respective element (comparative method or the k_0 method)

NAA and the REE Elements

Challenges in NAA

a. Activation product has too short of a half-life

b. A half-life that is too long can be tedious
   i. Must irradiate the sample longer or use a bigger use a bigger detector

c. Thermal Cross Section (TCS)
   i. TCS tells us the likelihood a neutron will react with the nucleus of the material undergoing NAA
   ii. The larger the TCS value, the greater the chance of neutrons irradiating the nucleus and causing it to emits its respective gamma rays
   iii. Similarly for the resonance integral (RI) of an element

Rare Earth Element Database

The goal of this research is to develop a methodology to maximize the analytical efficiency of NAA of REEs

Basic Breakdown:

- Predetermined which REE isotopes were relevant to include based on their abundance/natural occurrence
- Gathered basic information relevant to NAA
  - Abundance, thermal cross section, and resonance integral of the isotope, the half-life of its activation product
  - Most common strong gamma rays and their branching ratio, I_{gamma}(%), and I_{gamma}(rel.) values
    - This information was found using the decay schemes of the different elements

Element:	Abundance:	Common Rxn:	Half-Life:	Thermal Cross Section (Barns):	T.C.S Error:	Resonance Integral (Barns):	R.I. Error:	Gamma Ray:
139La	99.91	139La (n, γ) 140La	1.68 d	9.04	0.04	12.1	0.6	815 (44%; 24.2%; 23.28%)
139La	99.91	139La (n, γ) 140La	1.68 d	9.04	0.04	12.1	0.6	328 (44%; 19.6%; 20.32%)
139La	99.91	139La (n, γ) 140La	1.68 d	9.04	0.04	12.1	0.6	487 (19.2%; 44.7%; 45.5%)
139La	99.91	139La (n, γ) 140La	1.68 d	9.04	0.04	12.1	0.6	1596 (4.8%, 100%, 95.4%)

Weight Factor Parameters

WF Value	Abundance	Half-life	TCS	RI	BR	I_{gamma}(rel.)	Energy*
0.0	<10%	0-1.99 hrs	0	0	0	0	>1600 keV
0.0175	10-24.99%	n/a	0.01-0.99 barns	0.01 -0.99 barns	0.01-0.99%	0.01-0.99%	1000-1600 keV
0.035	25-49.99%	≥30 days	1-9.99 barns	1-9.99 bars	1-9.99%	1-9.99%	500-999.99 keV
0.07 0.075*	50-74.99%	2-11.99 hrs; 7-29.99 days	10-99.99 barns	10-99.99 bars	10-49.99%	10-49.99%	150-499.99 keV
0.14	75-100%	12 hrs-6.99 days	≥100 barns	≥100 barns	50-100%	50-100%	80-149.99 keV

There are 7 factors, each with a maximum possible weight factor (WF) value of 0.14.

How the Parameters Were Determined (1)

- Abundance:
  - As abundance of stable isotope increases, the likelihood of neutron activation increases
  - For this reason, 0.14 is assigned for 75-100%, and then for each 25% decrease we halved the WF value
- Half-life:
  - WF for half-life of the activation product is more complicated
  - 0-1.99 hours may be too short and would lead to poor counting statistics
  - 2-11.99 hours is better and assigned a value of 0.07
  - 12 hours – 6.99 days is the ideal time frame and assigned a perfect value of 0.14
  - 7 days – 29.99 days is a bit lengthy and may require a longer irradiation time and is assigned a value of 0.07
  - >30 days may require a longer counting time and assigned a value of 0.035

How the Parameters Were Determined (2)

- Thermal Cross Section (TCS):
  - For TCS the greater the value, the more likely a neutron will interact with the stable isotope during activation
  - As TCS increases, the WF thus increases, with a TCS of 100 barns or greater resulting in a perfect score of 0.14

- Resonance Integral (RI):
  - It is similar to TCS., in that a high RI value corresponds to an increased likelihood it will undergo NAA
  - WF follows the same parameters as TCS

How the Parameters Were Determined (3)

- Branching Ratio (BR):
  - The branching ratio is the percentage of beta particles populate an energy level
  - The greater this value is, the higher the likelihood a gamma ray will be emitted
  - 50-100% results in a branching ratio weight factor of 0.14, and it decreases as the BR lowers
- I_{gamma}(rel.):
  - Follows the same parameters as the BR weighting factors
  - This value tells us the relative percentage a specific gamma ray makes up decays from a energy level
- Energy:
  - Efficiency of the detector decreases as the energy of the photon emitted increases
  - As energy decreases, the WF increases to a max. of 0.14 being 80-149.99 keV

Decay Schemes

- Decay schemes were used to determine what the most common, “strong” gamma rays were, and their branching ratios
  - Each arrow is a specific gamma ray, red indicates the photon is strong, blue denotes it is weaker
- The vertical number is the energy in keV
- The branching ratio is the value on the left hand attached to the corresponding horizontal green line that the gamma ray decays from
  - A branching ratio can be comprised of multiple γ-rays therefore the I_{gamma}(rel.) value is also important, as a gamma ray could have a large % for the BR, but only make up a small % of the BR

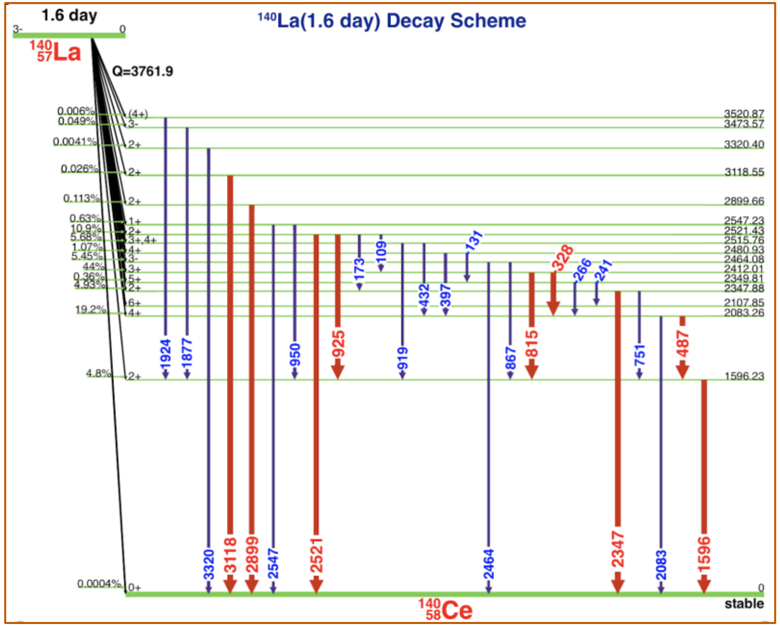

Figure 1. Decay Scheme of 140La (Reference _ with permission from Idaho National Laboratory)

Note: Each decay scheme has a respective gamma ray energies and intensities chart, which is how the I_{gamma}(relative) and I_{gamma}(%) values were obtained.

Weight Factor Example

139La		99.91	139La (n, γ) 140La	1.68 d	9.04	0.04	12.1	0.6	815 (44%; 24.2%; 23.28%)

- ^{139}La has an abundance of 99.91% -> abundance WF is 0.14
- Activation product, ^{140}La, has a half-life of 1.68 days -> HL WF is 0.14
- Thermal cross section of ^{139}La is 9.04 barns -> TCS WF is 0.035
- Resonance integral is 12.1 barns -> RI WF is 0.07
- For this example, we are specifically evaluating the 815 keV ray ^{140}La emits when it is irradiated and decays to ^{140}Ba Branching ratio of this gamma ray is 44% -> BR WF is 0.07 for this SPECIFIC photon
  - I_{gamma}(rel.) is 24.2% -> I_{gamma}(rel.) WF is 0.07
  - The energy of the ray is 815 keV -> Energy WF (also noted as “Strength WF” on the database) is 0.035

Ex: Total Weight Factor: (0.14 + 0.14 + 0.035 + 0.07 + 0.07 + 0.07 + 0.035)/0.98 = 0.5714286

Gamma Gamma Coincidence

- Each isotope in the database was also inspected to determine whether it was likely to undergo Gamma-Gamma Coincidence (GGC)
  - GGC assists in producing clearer NAA readings as it minimizes any background radiation "noise"
- An isotope is likely to undergo GGC if it has two or more gamma rays in coincidence with each other, especially if their branching ratios are high and if they are strong photons
  - To be in coincidence means one gamma ray "feeds" into the other one
  - Ex: Figure 1 shows the decay scheme for ^{140}La, which we can see that the 925 keV, 815 keV, and 487 keV rays all feed into the 1596 keV ray, thus they are in coincidence and ^{140}La is a good candidate for GGC
  - BR/I_{gamma} values matter because if these values are low, they won't give off a signal for a long period of time

Sources

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