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R-values

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I'll try to keep this short and simple. If something isn't clear, let me know.

$$R_{\text{LANL}} = \frac{\frac{(\text{beta cpm of } X_{\text{exp}} \text{ on system "A"})}{(\text{beta cpm of } {}^{99}\text{Mo}_{\text{exp}} \text{ on system "A"})}}{(\text{beta cpm of } X \text{ on system "A", from thermal on } {}^{235}\text{U})}}{(\text{beta cpm of } {}^{99}\text{Mo} \text{ on system "A", from thermal on } {}^{235}\text{U})}}$$

As I understand it, the above equation is the historical (as well as current) way of determining R-values using data from beta counting at LANL. The ratio in the denominator, a little "r," is the "baseline" or "calibration" value for a specific beta detector. Over time, if the detector "drifts" one would see a variation in this "r" during a thermal calibration measurement. This baseline is what LANL likes to track to monitor specific detector performance – this is not relevant to LLNL where gamma detection is used for determining R-values. LANL states that uncertainty is only dependent upon the count statistics for the isotopes measured. If one tries to convert this to an atom basis, the uncertainties will increase due to the incorporation of the uncertainties in the nuclear data used to convert the cpm to atoms.

LLNL switched to gamma detection methods in the 1970s thus replacing our beta counting effort. The equation below is how we have since determined R-values. The numerator ratios atom values of isotopes that are determined by measuring gamma cpm (usually? using several peaks per isotope) and then converting to particle decay in dpm using detector efficiency for each peak and the appropriate branch ratio for each gamma emission. Isotope decay is then converted to atoms using specific activity, mass or volume?, and Avogadro's number. The denominator is simply the ratio of published, cumulative fission product chain yields for isotopes produced in a thermal irradiation on ${}^{235}\text{U}$ – values of England & Ryder are used by LLNL for the NTNF program. Uncertainties in LLNL R-values are dependent upon gamma counting statistics *as well as* the nuclear data for each isotope.

$$R_{\text{LLNL}} = \frac{\frac{(\text{Atoms of } X_{\text{exp}})}{(\text{Atoms of } {}^{99}\text{Mo}_{\text{exp}})}}{\frac{(\text{Cumulative Fission Chain Yield of } X, \text{ from thermal on } {}^{235}\text{U})}{(\text{Cumulative Fission Chain Yield of } {}^{99}\text{Mo}, \text{ from thermal on } {}^{235}\text{U})}}$$

The next page tabulates fission chain yields and "atoms per gram" amounts measured in a recent NTNF Thermal Calibration. The R-values in the table are calculated using the LLNL method of determining R. The measure of success is demonstrated by how close to 1.00 the R-value is when determined during a Thermal Calibration. A value of 1.00 is the desired value. In the example below, only four isotopes lie outside of 1.00 by

more than 3 percent. These are the four isotopic measurements that obviously need to be improved.

Isotope	Cumulative Fission Chain Yield, per 100 fissions	Atoms per gram determined in a Thermal Calibration	Calculated R-values	More than 3 percent error in accuracy
89Sr	4.7331			
91Y	5.8282	7.559E+10	0.8992	Y
95Zr	6.5032	9.418E+10	1.0040	
97Zr	5.9840	8.824E+10	1.0223	
Mo99	6.1092	8.812E+10	1.0000	
103Ru	3.0311	4.406E+10	1.0077	
105Rh	0.9642	1.445E+10	1.0387	Y
111Ag	0.0174			
112Ag	0.0130			
115Cd	0.0116			
115mCd	0.0010			
136Cs	0.0055	8.117E+07	1.0161	
137Cs	6.1888	9.145E+10	1.0244	
140Ba	6.2149	9.034E+10	1.0077	
141Ce	5.8474	8.376E+10	0.9931	
143Ce	5.9562	8.416E+10	0.9796	
144Ce	5.4999	7.728E+10	0.9742	
147Nd	2.2469	3.284E+10	1.0132	
153Sm	0.1583	2.134E+09	0.9347	Y
156Eu	0.0149	2.740E+08	1.2790	Y
161Tb	0.0001			

Regardless of method, both R-LANL and R-LLNL (as well as others) should be able to provide values of unity during a Thermal Calibration within an agreed upon uncertainty budget, say 3 to 5 percent. If each lab obtains values that are unity, then their capability is healthy. If both methodologies provide values of unity, then the whole calibration concept across labs is healthy.

LANL's methodology is baselined solely to their physical beta counting equipment. LLNL's methodology is baselined to peer-reviewed, published nuclear data, namely fission chain yields of ²³⁵U during a thermal irradiation. If LANL loses a detector, it takes a thermal calibration to calibrate a new one; whereas, when LLNL loses a gamma detector, we just replace it with a new one and calibrate energy efficiencies with several standard NIST gamma sources.