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Nuclear Data Evaluations and Recommendations

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NUCLEAR DATA EVALUATIONS AND RECOMMENDATIONS

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INTRODUCTION

The published scientific literature is scanned and periodically evaluated for neutron and non-neutron nuclear data and the resulting recommendations are published [1,2]. After the literature has been scanned and appropriate data collected, there are often problems with regard to the treatment of the various types of data during this evaluation process and with regard to the method by which the recommendations are drawn from the assessment of the collection of individual measurements. Some problems with uncertainties are presented.

PERIODIC TABLE OF THE CHEMICAL ELEMENTS

There have been recent measurements reported in the literature that claimed discovery of two new chemical elements [3] with proton number, Z = 113 and Z = 115. There is a process in place by which that type of information is reviewed and conclusions drawn as to which set of reported data on a new element has a priority claim on the discovery and identification of a new chemical element and whether or not there has been verification of that claim. The resulting conclusion of the review process would allow one group to claim the discovery and be given the privilege of supplying the name of this new element.

In earlier centuries, the basis for the claim of discovery of a chemical element used the properties of the new substances, the colors of their compounds, the shapes of their crystals and their reactivity to determine the existence of new elements. The honor of discovery has often been accorded not to the person who first isolated the element but to the person who discovered the original mineral, even when the ore was impure and that ore actually contained many elements. The atomic weight of an element and a spectral analysis of an element were not available in the early days, but both of these elemental properties would be required before discovery of the element would be accepted by the latter part of the nineteenth century.

The requirements for discovery claims have tightened through the years and claims that were previously accepted would no longer meet the minimum constraints imposed today. There are also cases where the honor of discovery is not given to the first person to actually discover the element but to the first person to claim the discovery in print. If a publication was delayed, the discoverer has often been "scooped" by another scientist.

There were claims and counter-claims between USA and USSR scientists thirty years ago and there are similar problems between German and Russian scientists today. Not all

problems are of recent vintage. Over the past three centuries, there has been a long list of false claims for elements. The following is a partial list of names proposed in some of these false claims: austrium, berzelium, carolinium, celtium, columbium, damarium, davyum, decipium, demonium, denebium, didymium, donarium, dubhium, eurosamarium, euxenium, glaucodymium, ilmenium, incognitium, ionium, junonium, kosmium, lucium, masurium, metacerium, monium, mosandium, moseleyum, neokosmium, neptunium, nipponium, pelopium, philippium, polonium, rogerium, russium, sirium, thorine, vestium, victorium, wasmium, welsium and ytterbium.

The above names of ytterbium, columbium, ionium and neptunium are not the presently accepted names of existing elements, nor even the controversial columbium/niobium element with proton number, Z = 41, or the older name assigned to ^{230}Th.

At the turn of the twentieth century, the International Committee on Atomic Weights (ICAW) was formed. Although ICAW did not set internationally approved names, a name with an atomic weight value in their table lent support for the adoption of that name by the chemical community. Twenty years later, the ICAW became part of IUPAC, the International Union of Pure and Applied Chemistry, when it was formed. In 1949, the responsibility for acceptance of the name of a chemical element was given by IUPAC to its Commission on Nomenclature of Inorganic Chemistry (CNIC). The CNIC did not deny the right of a discoverer to propose a name for a new chemical element. However, the general criteria were that the approved name should differ as little as possible in different languages; the name should be based on practicality and prevailing usage; and finally the choice of the name carries no implication at all about the priority of discovery. The names of the chemical elements are determined by the acceptance of that name by the chemical community, the priority rights of the discoverer not withstanding. In the latest reorganization of IUPAC, the CNIC no longer exists. The IUPAC Inorganic Chemistry Division Committee now has responsibility for acknowledging new elements.

Previously, chemical elements were discovered in nature. For more than half a century now, new chemical elements are not being discovered in nature but are being synthesized using nuclear reactions. As a result, a joint committee has been formed by IUPAC and by IUPAP (the International Union of Pure and Applied Physics). This joint IUPAC/IUPAP committee has the responsibility to review the claims of discovery of a new element and to investigate whether or not there has been a verification of each discovery. As a result, the element with proton number Z = 110, Darmstadtium, has been added to the Periodic Table [4]. A second element with proton number Z = 111, Roentgenium, has also been reviewed [5] and approved.

RADIOACTIVE DECAY CONSTANTS

The radioactive half-life of a nuclide (the reciprocal of the radioactive decay constant) is of interest to reactor dosimetry because the half-life of the product nucleus in a nuclear reaction has a direct impact on the determination of the reaction rates. For a given count rate, the reaction rate is inversely proportional to the half-life of the measured product

nucleus. Problems often arise in connection with the estimation of the systematic component of the error in the half-life or decay constant measurement. Because of their use in dating samples, there are some long-lived nuclides whose half-lives are of interest to the geological community. In many cases, these nuclides have reported half-lives, which completely disagree with each other.

^{176}Lu results from counting experiments report half-life values of 3.69 (.02) x 10^{10} years [6] and also 4.08 (0.03) x 10^{10} years [7], where the value in parentheses represents the uncertainty. If one accepts the reported uncertainty as one standard deviation, these two measurements differ between 13 and 19 standard deviations and could not be considered to be estimating the same physical quantity. In a similar manner, the data reported for ^{187}Re (a liquid scintillation counting measurement) of 3.51 (0.37) x 10^{10} years [8] and (a cryogenic micro-calorimeter) of 6.2 (0.6) x 10^{10} years [9], are significantly different estimates of this half-life. These problems will be discussed in a later section of this paper.

There has been a significant interest in the possible decay modes of the stable isotopes of the chemical elements. For the very long-lived nuclides (many of which have always been considered stable isotopes) there have been attempts to determine the decay rate for exotic reactions such as proton decay, long lived alpha decay, long lived beta decay and double beta decay. Previously [10], attempts to discover violations to the principle of charge conservation were discussed. There have been no new measurements since the study of the ^{73}Ge beta decay to ^{73}As, which is energetically forbidden. The measurement [11] led to a lower half-life limit > 2.6 x 10^{23} years.

A few years ago, half-lives and decay modes of long-lived nuclides were presented [10]. A latest revision of these data is presented in Table 1. There have been changes, particularly in the case of the double beta decay ($\beta\beta$) mode nuclides. Only decay modes that involve the emission of 2 neutrinos are listed in the table, since ($\beta\beta$) decay modes, without the emission of neutrinos (0ν) have not been detected. Their lower limits are one or more orders of magnitude larger than ($2\nu\ \beta\beta$) decay modes.

Table 1. Radioactive Half-lives and Decay Modes of long-lived (quasi-stable) nuclides

Nuclide	Decay	Years	Nuclide	Decay	Years	Nuclide	Decay	Years
^{1}H	proton	$>35\cdot10^{27}$	^{40}Ca	ec-ec	$59\cdot10^{20}$	^{48}Ca	$\beta^-\beta^-$	$43\cdot10^{18}$
^{50}Cr	β^+-ec	$13\cdot10^{17}$	^{58}Ni	β^+-ec	$>4\cdot10^{19}$	^{64}Zn	ec-ec	$>23\cdot10^{17}$
^{71}Ga	β^-	$35\cdot10^{25}$	^{76}Ge	$\beta^-\beta^-$	$18\cdot10^{20}$	^{82}Se	$\beta^-\beta^-$	$12\cdot10^{19}$
^{78}Kr	$\beta^+\beta^+$	$>20\cdot10^{20}$	^{94}Zr	$\beta^-\beta^-$	$>1\cdot10^{17}$	^{96}Zr	$\beta^-\beta^-$	$21\cdot10^{18}$
^{92}Mo	β^+-ec	$>19\cdot10^{18}$	^{100}Mo	$\beta^-\beta^-$	$82\cdot10^{17}$	^{96}Ru	$\beta^+\beta^+$	$>31\cdot10^{15}$
^{106}Cd	$\beta^+\beta^+$	$>24\cdot10^{19}$	^{108}Cd	ec-ec	$10\cdot10^{17}$	^{114}Cd	$\beta^-\beta^-$	$>6\cdot10^{17}$
^{116}Cd	$\beta^-\beta^-$	$38\cdot10^{18}$	^{124}Sn	$\beta^-\beta^-$	$>22\cdot10^{17}$	^{128}Te	$\beta^-\beta^-$	$2\cdot10^{24}$
^{130}Te	$\beta^-\beta^-$	$8\cdot10^{20}$	^{124}Xe	ec-ec	$>1\cdot10^{17}$	^{134}Xe	$\beta^-\beta^-$	$>11\cdot10^{15}$
^{136}Xe	$\beta^-\beta^-$	$> 8\cdot10^{20}$	^{130}Ba	ec-ec	$2\cdot10^{20}$	^{132}Ba	ec-ec	$13\cdot10^{20}$
^{136}Ce	$\beta^-\beta^-$	$>18\cdot10^{15}$	^{138}Ce	ec-ec	$>9\cdot10^{13}$	^{150}Nd	$\beta^-\beta^-$	$7\cdot10^{18}$
^{160}Gd	$\beta^-\beta^-$	$>19\cdot10^{18}$	^{176}Yb	$\beta^-\beta^-$	$>16\cdot10^{16}$	^{180}W	ec-ec	$7\cdot10^{16}$
^{186}W	$\beta^-\beta^-$	$37\cdot10^{17}$	^{209}Bi	α	$19\cdot10^{18}$	^{238}U	$\beta^-\beta^-$	$20\cdot10^{20}$

The claim [12] by Klapdor-Kleingrothaus to have measured a ($0\nu\ \beta\beta$) decay in ^{76}Ge, which would imply lepton number non-conservation has not yet been substantiated, in spite of the very number of publications on the subject, both by the original authors and many others.

DATA ON THE VERY HEAVY CHEMICAL ELEMENTS

For the very heavy elements, there are other modes of decay (determining factors in stability), spontaneous fission (sf) decay and cluster decay. In sf decay, the nucleus breaks up into two approximately equal reaction products, both of which have very large masses (some 15 to 40 times larger than an alpha particle). Cluster decay is radioactive decay in which the emitted particle has a much smaller mass than a fission product but it is still larger than the alpha particle (some 3 to 8 times larger). For very heavy chemical elements, a path to the super-heavy elements is being explored. In the process of this investigation, new nuclides and elements are being discovered. The present list of the highest Z elements (Z > 109) and their nuclides is shown in Table 2.

Table 2. Nuclear Data on the Very Heavy Chemical Elements

Nuclide	Half-life	Mode Decay	Energy (MeV)	Nuclide	Half-life	Mode Decay	Energy (MeV)
^{267}Ds	3 μ-sec	α	11.6	^{269}Ds	0.17 ms	α	11.1
^{270m}Ds	~6 ms	α	11.0-12.2	^{270g}Ds	~10 μs	α	11.0
^{271m}Ds	~1.1 ms	α	10.7	^{271g}Ds	~56 ms	α	10.7
^{273m}Ds	0.76 ms	α	11.8	^{273g}Ds	118 ms	α	9.73
^{279}Ds	0.18 s	sf 90% α 10%	9.70	^{281}Ds	~9.6 s	sf	
^{272}Rg	1.5 ms	α	11.0	^{274}Rg	9.26 ms	α	11.15
^{279}Rg	~0.17 s	α	10.4	^{280}Rg	~3.6 s	α	9.8
^{277}112	0.48 ms	α	11.2-11.7	^{282}112	0.5 ms	sf	
^{283}112	~ 4. s	α 100% sf $\leq$10%	9.54	^{284}112	0.10 s	sf	
^{285}112	~ 34. s	α	9.16	^{278}113	0.34 ms	α	11.7
^{283}113	~0.1 s	α	10.1	^{284}113	~0.5 s	α	10.0
^{286}114	0.16 s	sf 60% α 40%	10.2	^{287}114	0.5 s	α	10.02
^{288}114	0.8 s	α	9.95	^{289}114	~2.7 s	α	9.8
^{287}115	0.03 s	α	10.6	^{288}115	~87 ms	α	10.5
^{290}116	~ 15 ms	α	10.9	^{291}116	~6 ms	α	10.74
^{292}116	~18 ms	α	10.66	^{293}116	~0.05 s	α	10.5
^{294}118	~1.8 ms	α	11.7				

THE PROBLEM OF ERROR ESTIMATION

Absolute certainty is a privilege of uneducated minds and fanatics. It is for scientific folk, an unattainable ideal [13]. Most scientists would agree with the above assessment. However, in practice, all too often we assume that with a little effort, we can easily reduce all of our experimental errors to the level of insignificance. There has been an increased emphasis on uncertainties in measurements with the development of computerized programs to handle sensitivity analysis using variance-covariance matrices to treat the correlation among data. However, if input uncertainties have been poorly estimated, computer programs merely enable you to determine the wrong answer much more quickly. There has been a tendency with the advent of hand-held calculators for users to believe that all of the digits listed on the display screen are significant.

In spite of the importance of consistency in working with uncertainties, there was no international consensus on these matters until the mid-1990s. The International Committee for Weights and Measures (Comité International des Poids et Mesures, CIPM) initiated the development of a guide that would establish and promote general rules for calculating and expressing uncertainty. This guide is entitled "Guide to the Expression of Uncertainty in Measurement [14]. It is usually referred to by its abbreviation, "GUM". GUM introduces a new method for categorizing uncertainties, depending upon how they are evaluated.

"Type A uncertainties" are those uncertainties evaluated by the application of standard statistical methods to a series of observations. The standard deviation of the mean is termed a "standard uncertainty obtained by a type A evaluation" according to GUM. "Type B" evaluations of uncertainty apply methods other than statistical analysis of a series of observations in order to determine a standard uncertainty. "Type B uncertainties" have previously been referred to as systematic uncertainties. The determination of a quantity may require use of other sources of information, such as a data of a calibration standard, data taken from a reference document, or data taken from a manufacturer's specification. Whatever the source, a type B evaluation requires that a standard uncertainty be extracted from the information by a careful consideration of the way in which the uncertainty has been expressed by the source. If the source indicates a stated level of confidence, such as 95% or 99%, convert that uncertainty to a standard uncertainty by treating the quoted uncertainty as if a normal probability distribution had been used to calculate it and dividing by the appropriate factor for such a distribution. This would imply factors of 1.96 and 2.576 in the level of confidence stated above.

Eisenhart [15] noted that a reported value, whose accuracy is entirely unknown, is worthless. A measurement result is complete only when accompanied by a quantitative statement of its uncertainty. This uncertainty is required in order to decide if the result is adequate for its intended purpose and to ascertain if it consistent with other similar results.

A reported value whose accuracy is significantly overestimated is extremely misleading, particularly if a truly honest effort has not been made to identify and quantify the various sources of type B uncertainty. Most present day measurements rely on the measured values of various other parameters as so-called, auxiliary constants, and if all of these parameters

had unknown sources of type B uncertainty, which biased their results, we would have an enormous problem.

Usually, recommended values and uncertainties are based on the averages obtained by weighting each measurement by the reciprocal square of the quoted standard deviation, which is called variance weighted averaging. Occasionally, recommendations may be based on selecting the one good recent measurement. There are times when a discrepancy in values and uncertainties quoted by a given technique require that the techniques be averaged to produce a more reasonable recommendation.

There are measurements, where the authors quote uncertainties that are orders of magnitude smaller than all other measurements of that quantity. As a result, if variance weighting of the results is applied indiscriminately, these measurements would both disagree with and would exclude many other good recent measurements from consideration in determining the average value of the parameter. Undoubtedly, the "type B uncertainties" have not been carefully considered in these publications.

When experiments are performed at the level of five to ten percent accuracy, recording the number of counts is an important consideration. In Poisson statistics, increasing the number of counts can improve the overall accuracy, since the accuracy varies inversely with the total number of counts. However, when the overall accuracy reaches the level of one-half percent or better, the estimate of all "type B uncertainties" begin to become a controlling influence on the total accuracy. Continuing to improve the statistical precision, by collecting more raw data points does not significantly improve the total error, except superficially, in the absence of any effort to estimate the type B uncertainties. Using variance weighting indiscriminately in such cases, penalizes the authors who attempt the difficult task of estimating their "type B uncertainties", while benefiting the authors who make no such attempt to determine all of their sources of uncertainty.

If one has a choice in designing the ideal experiment to measure a physical quantity such as a half-life, one would choose to make measurements on many different samples, using a number of duplicate instruments and utilizing a variety of different methods or techniques. This procedure should help to provide the necessary information to correctly estimate the 'type B uncertainties" in the measurement.

PSYCHOLOGICAL FACTORS IN ESTIMATING ERROR

We now consider the psychological factor, which is involved with the treatment of type B uncertainties, as noted by both Birge [16] and Bridgman [17]. Individual authors search for all sources of error involved in their work, which would bring their result into agreement with all of the earlier measurements of their particular quantity. Whenever the authors' renormalized result agrees with all other measurements of the quantity, the author ceases to investigate their errors any further. In addition, if their measured value is too large, compared to other measurements, the authors do not look for any factors, which would

cause their result to increase, but merely for factors that result in the reduction of their measured value.

Consider the following two examples of psychological factors. For a large number of years, the value for the charge on the electron had been based on a set of measured values that were all consistent with one another. When Rutherford measured a value that was reported to be 60% larger than all of the previous results, all of the measurements made subsequent to Rutherford were suddenly found to be consistent with his new higher value.

In a similar manner, all of the early measurements of Planck's constant clustered about the same value until Birge, in 1941, raised that value by one and one-quarter percent in his analysis of the fundamental constants. After this breakthrough, all subsequent measurements of Planck's constant clustered around this "new" estimate.

Finally, consider the evaluation of the half-life for ^{239}Pu. In 1974, a best estimate [18] was reported of 24,395 (29) years, where the uncertainty was quoted at the confidence level corresponding to three standard deviations (3σ). Thus, the probability would be 99.7 % that the real value should fall within those uncertainty limits. This half-life result was obtained from experiments that used the alpha particle counting technique. All of these early alpha-counting experiments were consistent. However, there had been one calorimetric measurement [19], which reported a half-life value that was some 300 years lower. More recent measurements on the ^{239}Pu half-life (including alpha counting measurements) all now agree with the lower half-life estimate. The presently recommended value is now thirty standard deviations (30 σ) away from the 1974 recommendation.

CONCLUSIONS

We can conclude from this discussion that type B uncertainties are very difficult to estimate but they are extremely important to identify and to either eliminate them or to at least account for them in the quoted result.

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