Comments on Equilibrium, Transient Equilibrium, and Secular Equilibrium in Serial Radioactive Decay

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Equations describing serial radioactive decay are reviewed along with published descriptions of transient and secular equilibrium. It is shown that terms describing equilibrium are not used in the same way by various authors. Specific definitions are proposed; they suggest that secular equilibrium is a subset of transient equilibrium.

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Equilibrium is used to describe the condition where the derivative of a function is equal to zero. In radioactive decay it relates to the situation where the number of atoms of a particular kind does not change with time. Clearly, this is a contradiction, since radioactive decay implies change, whereas equilibrium does not. Whereas absolute equilibrium is not possible, equilibrium is adequately approximated in many important situations whenever the parent species is substantially longer-lived than the daughter nuclides.

The various approximations to equilibrium give rise to terms *transient equilibrium* and *secular equilibrium*. These terms, however, are not used in a consistent way in physics textbooks (*1–4*) and they appear to have been applied improperly in some reference works (*5–10*). As a result of this discordant information, considerable confusion exists in some technical personnel who do not deal with these concepts regularly, especially when these concepts are applied to available generator systems. As an example, it is not uncommon for examination review questions to be phrased something like the following. "A radionuclide generator system is described by the term _____ 1) transient equilibrium, 2) secular equilibrium, etc." (*11–12*). As this communication will show, the response to such a question will depend upon the reference books consulted and, hence, upon the individual's training. From this brief review it is hoped that whatever confusion exists will be dispelled and a more consistent usage of terms and concepts may emerge.

Consider serial radioactive decay where Species 1 decays only to Species 2, which in turn decays only to Species 3, and so on until finally a stable product is formed. The following definitions will be used:

N_1^0 = the number of atoms of the first species at time zero;

N_1 = the number of atoms of the first species at any time t;

λ_1 = the radioactive decay constant for atoms of the first species;

etc.

When there are only three nuclear species, which is of primary importance to nuclear medicine, the differential equations describing the decay and buildup of the various nuclides are (*1*)

$$\frac{dN_1}{dt} = -\lambda_1 N_1, \tag{1}$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2, \text{ and} \tag{2}$$

$$\frac{dN_3}{dt} = \lambda_2 N_2. \tag{3}$$

When only Species 1 is present initially, the standard solutions are:

$$N_1 = N_1^0 e^{-\lambda_1 t}, \tag{4}$$

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}), \text{ and} \tag{5}$$

$$N_3 = N_1^0 \left(1 + \frac{\lambda_1}{\lambda_2 - \lambda_1} e^{-\lambda_2 t} - \frac{\lambda_2}{\lambda_2 - \lambda_1} e^{-\lambda_1 t}\right). \tag{6}$$

Consider three special cases in the discussion of Eq. 5, which relates to the buildup and decay of the first daughter in the generator system.

Relatively short-lived parent—$\lambda_1 > \lambda_2$. Since $\lambda \approx \frac{0.7}{T_{1/2}}$, in the present case $\lambda_1 > \lambda_2$, and the amount of daughter element, N_2, will increase with time until it goes through a maximum. Eventually, when the parent element has decayed away, the daughter element will decay with its own characteristic half-life.

There are no commercially available radionuclide generators that possess the characteristic of $\lambda_1 > \lambda_2$. However, Brucer (*13*) has observed that such generator systems may be useful, pointing out an example, which he termed a reverse cow, in the commercial production of I-131. Quoting from Brucer, "In the early days of the large-scale production of I-131, reactor pro-

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duction time was purchased for tellurium irradiation in a Canadian reactor. During shipment to American pharmaceutical houses, the 1.3-day tellurium parent decayed to the desired 8-day I-131 daughter, which was readily separated." Brucer further comments these "reverse cows" may be useful in producing a label that originates at the site of deposition of another label. A similar suggestion was made by Spencer and Hosain in a discussion of generator systems with approximately equal decay constants for parent and daughter (*14*). No equilibrium is established under these conditions and no descriptive term has been suggested to denote it.

Relatively long-lived parent—$T_1 > T_2$ ($\lambda_1 < \lambda_2$). For sufficiently large values of t, $e^{-\lambda_2 t}$ will be negligibly small compared with $e^{-\lambda_1 t}$. Under these circumstances, Eq. 5 is approximated by

$$N_2 = \frac{\lambda_1 N_1{}^0 e^{-\lambda_1 t}}{\lambda_2 - \lambda_1}. \tag{7}$$

If the parent and daughter elements remain undisturbed then at sufficiently large values of t, Eq. 7 predicts that N_2 will appear to decay with the characteristic half-life of the parent. Combining Eqs. 4 and 7 we get:

$$\frac{N_2}{N_1} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \tag{8}$$

for large values of t.

Equation 8 predicts that eventually a constant ratio, independent of time, will be established between N_2 and N_1.

This condition, where N_2/N_1 is constant but both decrease with time, is referred to as transient equilibrium by Kaplan (*1*) and Hendee (*2*) but is called secular equilibrium by Lapp and Andrews (*3*). Acosta et al. (*4*) seem to take an intermediate approach: they do not define a transient equilibrium but do define a secular equilibrium (see Eqs. 13–16).

Since the activity $A = \lambda N$, it follows from Eq. 8 that

$$\frac{A_2}{A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1}. \tag{9}$$

Equation 9 predicts that, given Special Case *B* and a long time, t, the activity of Species 2 will always be greater than that of Species 1.

An example of the application of Eqs. 5, 6, and 9 is given by the Te-132 → I-132 generator (*2*). Interestingly, most of the reference books consulted (*5–10*), but not all (*3*), have apparently applied these equations to the Mo-99 → Tc-99m generator, since most of the books do show that the activity of Tc-99m exceeds that of Mo-99 at large times.

In order to make Eq. 5 general for any generator system, such as the Mo-99 → Tc-99m generator, the fraction, F, of Species 1 decaying directly to Species 2 must be taken into account. Then

$$N_2 = \frac{F\lambda_1 N_1{}^0 e^{-\lambda_1 t}}{\lambda_2 - \lambda_1} \tag{10}$$

and

$$\frac{A_2}{A_1} = \frac{F\lambda_2}{\lambda_2 - \lambda_1}. \tag{11}$$

Package inserts from the manufacturers of generators and several literature sources (e.g., *2,14,15*) are available, and provide the correct technical information for the Mo-99 → Tc-99m generator.

Extremely long-lived parent—$T_1 \gg T_2$. Here $\lambda_1 \ll \lambda_2$, and under this condition we can approximate $\lambda_2 - \lambda_1$ by λ_2 and $e^{-\lambda_1 t}$ by 1. For radionuclide generators this latter assumption is not true for long time spans (λ, $t \neq 0$), i.e., time spans that approximate the useful life of the generator. However, if we restrict our attention to time spans that are long compared with the half-life of the daughter element, then the Sn-113 → In-113m is a good example where $e^{-\lambda_1 t}$ can be approximated by 1. Equation 5 can then be written for any generator as

$$N_2 = \frac{F\lambda_1 N_1{}^0}{\lambda_2}(1 - e^{-\lambda_2 t}). \tag{12}$$

At longer times, when $e^{-\lambda_2 t} \to 0$,

$$N_2 = \frac{F\lambda_1 N_1{}^0}{\lambda_2}. \tag{13}$$

At still longer times, when $e^{-\lambda_2 t} \to 0$ but we can no longer approximate $e^{-\lambda_1 t}$ by 1, Eq. 13 becomes

$$N_2 = \frac{F\lambda_1 N_1{}^0 e^{-\lambda_1 t}}{\lambda_2} = \frac{F\lambda_1 N_1}{\lambda_2}, \tag{14}$$

or

$$\frac{A_2}{A_1} = F. \tag{15}$$

The condition where Eq. 13 is a valid approximation to Eq. 5 is generally referred to as secular equilibrium (e.g., *1,2,3*), although not by all authors (e.g., *3,17*). Most authors stop at Eq. 13 in their derivation of secular equilibrium, but Acosta et al. (*4*) have carried their derivation on to Eq. 15.

DISCUSSION

It is common to define secular equilibrium as that condition where the activities of the parent and the daughter are equal. Further, most authors either implicitly or explicitly define secular equilibrium as that condition when the parent and daughter activities are equal and there is no important decay of the parent during the period of interest implied in the definition. Notice that Eq. 11, which refers to transient equilibrium, shows that with a judicious choice of F, one could theoretically describe a parent-daughter pair where parent and daughter activities remain equal even though substantial decay of the parent may take place during the period of interest. This definition would then include secular equilibrium as a subset of transient equilibrium. No example of this theoretical possibility comes to mind.

If one defines transient equilibrium as that condition where a constant ratio of activities obtains but the parent decays appreciably during the period of interest, then one must also define what is meant by the term, "period of interest." Clearly, for long time spans, all parent nuclides in generator systems decay appreciably. However, if one defines the period of interest as several elution periods, then most generator systems have parent nuclides that do not decay appreciably. For example, everyone may agree that the Mo-99 → Tc-99m generator could be described by the term "transient equilibrium;" but not everyone may agree on the appropriate term used for describing the Sn-113 → In-113m generator.

Based upon the information in this communication, the following definitions are proposed, in the belief that they are compatible with the majority of applications.

> *Transient equilibrium* is that condition in serial radioactive decay where the ratio of activities of the parent and daughter radionuclides is constant. The mathematical definition is given by Eqs. 7–11.

> *Secular equilibrium* is that condition in serial radioactive decay where the ratio of activities of the parent and daughter

radionuclides is a constant and where there is no important decay of the parent nuclide during the time interval of interest. Secular equilibrium should be considered as a subset of transient equilibrium. The mathematical definition is given by Eqs. 12 and 13. (Note that in such a secular equilibrium the activity of the *daughter* nuclide is also essentially constant.)

In accordance with the above definitions, all radionuclide generators in current use in nuclear medicine can achieve transient equilibrium. Most of them, but not all, can be described by both the terms transient equilibrium and secular equilibrium if the time of interest is restricted to a few elution periods rather than the life of the generator or historical time.

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