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The Equations of Radioactive Transformation in a Neutron Flux

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The standard equations of radioactive transformation are generalized to include the case where radioactive nuclei are subject to transformation by neutron absorption in addition to spontaneous decay. Opportunity is taken to derive the equations by a simple method which avoids recourse to the formal methods of solving differential equations. A method is pointed out whereby one may write simply by inspection the equation for the activity of a species which is descended from an ancestral species through a chain of any combination of decays and neutron absorptions. Some problems are solved as illustration.

WHEN a radioactive source is immersed in a neutron flux, the radioactive nuclei in the source are subject to transformation by neutron capture in addition to their transformation by spontaneous radioactive decay. Even with the largest fluxes we can foresee at present, the ratio of transformations by neutron capture to transformations by decay is very small for most radioactive nuclei. However, the general equations of radioactive transformation in a neutron flux will be useful for nuclei of long half-life and large cross-section, and especially for particular problems such as that illustrated by Katcoff's experiment discussed in Section IV below.

For convenience, the discussion is limited to the case of a neutron flux; the equations and methods are, of course, applicable to a flux of any kind of particle.

I. THE ACTIVITIES OF THE DIFFERENT MEMBERS OF A DECAY CHAIN DESCENDED FROM A PRIMAL ANCESTRAL ACTIVITY A_1^0

We consider a series of genetically related activities:

$$A_1 \xrightarrow{\lambda_1} A_2 \xrightarrow{\lambda_2} \cdots \xrightarrow{\lambda_{n-1}} A_n \xrightarrow{\lambda_n}.$$

Nuclei of the ith species transform spontaneously at rate

$$A_i(t) = \lambda_i N_i(t), \tag{I.1}$$

where $A_i(t)$ and $N_i(t)$ are, respectively, the activity (disintegrations per second) and number of nuclei of the ith species at time t, and λ_i is the corresponding decay constant.

Consideration of the fact that $A_i(t)$ is also the rate of creation of nuclei of the $(i+1)$th species leads, in the case of zero flux, to the well-known set of simultaneous linear differential equations with constant coefficients that governs the time dependence of the number of nuclei of the different species:

$$\begin{cases} dN_1/dt = -\lambda_1 N_1, \\ dN_2/dt = \lambda_1 N_1 - \lambda_2 N_2, \\ \cdot \\ \cdot \\ \cdot \\ dN_n/dt = \lambda_{n-1} N_{n-1} - \lambda_n N_n. \end{cases} \tag{I.2}$$

If the nuclei are immersed in a constant neutron flux of magnitude ϕ neutrons cm^{-2} sec.$^{-1}$, and the ith species has cross-section σ_i cm^2 for all neutron reactions that permanently alter the nuclei,[1] then the nuclei will transform by two processes: (1) spontaneous radioactive decay at rate $\lambda_i N_i$ and (2) neutron reaction at rate $\phi\sigma_i N_i$. Therefore, the total transformation rate of the ith species at time t is $(\lambda_i+\phi\sigma_i)\mathbf{N}_i(t)$, where bold face type is used to distinguish a symbol pertaining to the case of non-zero flux from the corresponding symbol in the case of zero flux. We denote

$$\Lambda_i \equiv \lambda_i + \phi\sigma_i. \tag{I.3}$$

Now, independently of the flux, nuclei of the $(i+1)$th species arise only by radioactive decay of nuclei of the ith species, so that their rate of creation is $\lambda_i \mathbf{N}_i$. Consequently, for a non-zero neutron flux we have, in place of Eq. (I.2),

$$\begin{cases} d\mathbf{N}_1/dt = -\Lambda_1 \mathbf{N}_1, \\ d\mathbf{N}_2/dt = \lambda_1 \mathbf{N}_1 - \Lambda_2 \mathbf{N}_2, \\ \cdot \\ \cdot \\ \cdot \\ d\mathbf{N}_n/dt = \lambda_{n-1} \mathbf{N}_{n-1} - \Lambda_n \mathbf{N}_n. \end{cases} \tag{I.4}$$

The solution of these equations can be written directly by appropriate substitution of Λ's for λ's in the solution of Eqs. (I.2).

The solutions of the latter in a symmetrical form have been known for a long time. They may be ob-

[1] σ_i is the sum of the (n, γ), (n, p), $(n, 2n)$, etc. cross sections, but does not include the (n, n) cross section unless the latter reaction results in an excited state whose lifetime for return to the lowest state is not insignificant compared to the lifetime of the nucleus for decay.

tained in a straightforward manner by methods treated in all the texts on differential equations, or by the method of Bateman.[2] However, it is possible to obtain the solutions of these equations without appeal to the formal methods used in differential equations, by a very simple method which emphasizes the physical content of the situation and is at least as compact as the other methods.

We note first that a single nuclear species of decay constant λ and cross-section σ in a constant neutron flux ϕ transforms at a rate given by the first of Eqs. (I.4), so that

$$\mathbf{A}(t)=\lambda\mathbf{N}(t)=A^0e^{-\Lambda t}, \tag{I.5}$$

where $A^0\equiv\mathbf{A}(0)=\lambda N^0$.

Now, suppose that nuclei of this species are created at rate $R(t')$ during time interval $0\leqslant t'\leqslant t$. Of the $R(t')dt'$ nuclei created during time interval dt' at time t', the number still untransformed at time t (which is an amount of time $t-t'$ later), is, by Eq. (I.5),

$$d\mathbf{N}(t', t)=R(t')\exp[-\Lambda(t-t')]dt'.$$

Then the activity at time t is, assuming zero activity at $t'=0$,

$$\mathbf{A}(t)=\lambda\mathbf{N}(t)=\lambda e^{-\Lambda t}\int_0^t R(t')e^{\Lambda t'}dt'. \tag{I.6}$$

We can now derive any of the transformation equations by appropriate specialization of $R(t')$.[3]

We shall assume in the following that $\mathbf{A}_1(0)=A_1{}^0$, and $A_2{}^0=\cdots=A_n{}^0=0$.

The rate of creation $R_2(t')$ of daughter nuclei is equal to the rate of radioactive decay of parent nuclei:

$$R_2(t')=\mathbf{A}_1(t')=A_1{}^0e^{-\Lambda_1 t'}.$$

Putting this into Eq. (I.6) and integrating, we get a result that can be put into the symmetrical form,

$$\mathbf{A}_2(t)=\lambda_2\mathbf{N}_2(t)=\lambda_2\lambda_1N_1{}^0\left(\frac{e^{-\Lambda_1 t}}{\Lambda_2-\Lambda_1}+\frac{e^{-\Lambda_2 t}}{\Lambda_1-\Lambda_2}\right). \tag{I.7}$$

Similarly, the rate of creation $R_3(t')$ of granddaughter nuclei is equal to the rate $\mathbf{A}_2(t')$ of radioactive decay of daughter nuclei. Using Eq. (I.6) again, we find, after a little algebraic manipulation to get the symmetrical form,

$$\mathbf{A}_3(t)=\lambda_3\mathbf{N}_3(t)=\lambda_3\lambda_2\lambda_1N_1{}^0\left[\frac{e^{-\Lambda_1 t}}{(\Lambda_2-\Lambda_1)(\Lambda_3-\Lambda_1)}+\frac{e^{-\Lambda_2 t}}{(\Lambda_1-\Lambda_2)(\Lambda_3-\Lambda_2)}+\frac{e^{-\Lambda_3 t}}{(\Lambda_1-\Lambda_3)(\Lambda_2-\Lambda_3)}\right]. \tag{I.8}$$

The symmetrical form of Eqs. (I.7) and (I.8) permits us to write directly the expression for the activity of the nth generation species:

$$\mathbf{A}_n(t)=\lambda_n\mathbf{N}_n(t)=\left(\prod_{i=1}^{n}\lambda_i\right)N_1{}^0\sum_{i=1}^{n}\mathbf{a}_ie^{-\Lambda_i t}\quad(n>1), \tag{I.9}$$

where

$$\mathbf{a}_i=\prod_{j\neq i}(\Lambda_j-\Lambda_i)^{-1}\quad(j=1, 2, \cdots, n).$$

This equation can be verified formally by mathematical induction.

If $\phi=0$, Eq. (I.9) becomes the solution of the last of Eqs. (I.2):

$$A_n(t)=\lambda_nN_n(t)=\left(\prod_{i=1}^{n}\lambda_i\right)N_1{}^0\sum_{i=1}^{n}a_ie^{-\lambda_i t}\quad(i>1), \tag{I.10}$$

where

$$a_i=\prod_{j\neq i}(\lambda_j-\lambda_i)^{-1}\quad(j=1, 2, \cdots, n).$$

Note that Eq. (I.9) breaks down if any two Λ's are equal. The equation for this case is most easily obtained by setting the Λ's equal before integration. For instance, if $\Lambda_2=\Lambda_1$, the integrand of the integral leading to Eq. (I.7) becomes equal to 1, and

$$\mathbf{A}_2(t)=\lambda_2A_1{}^0te^{-\Lambda_2 t}.$$

It remains to notice that if $A_2{}^0, A_3{}^0\cdots A_n{}^0$ are not all zero, then $\mathbf{A}_n(t)$ is obtained by summing the contributions made to it by the different initial activities,

$$\mathbf{A}_n(t)=\lambda_n\mathbf{N}_n(t)=\sum_{i=1}^{n}\left(\prod_{k=i}^{n}\lambda_k\right)N_i{}^0\sum_{k=i}^{n}\mathbf{a}_ke^{-\Lambda_k t}, \tag{I.11}$$

where $\mathbf{a}_k=1$ when $i=n$, and when $i<n$,

$$\mathbf{a}_k=\prod_{p\neq k}(\Lambda_p-\Lambda_k)^{-1}\quad(p=i, i+1, \cdots, n).$$

If the flux is suddenly shut off at time t, the different activities which are present at that time in amounts given by Eq. (I.5) and the general Eq. (I.9), transform subsequently by radioactive decay alone according to the zero flux equations. With the elapse of T time units after flux shut-off, the activity $\mathbf{A}_1(t, T)$ of the primal ancestor becomes

$$\mathbf{A}_1(t, T)=\mathbf{A}_1(t)e^{-\lambda_1 T}=A_1{}^0\exp[-(\Lambda_1 t+\lambda_1 T)];$$

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[2] H. Bateman, Proc. Camb. Phil. Soc. 15, 423 (1910).

[3] This fact is noted by Meyer and Schweidler, *Radioaktivität* (B. G. Teubner, Leipzig, 1927), p. 57. They write (translating to the notation of this paper)

$$N(t)=e^{-\lambda t}\left(N^0+\int_0^t R(t')e^{\lambda t'}dt'\right)$$

as the solution of the differential equation $dN(t')/dt'=R(t')-\lambda N(t')$.

the activity of the daughter will be

$$\mathbf{A}_2(t, T)=\lambda_2\mathbf{A}_1(t)\left(\frac{e^{-\lambda_1 T}}{\lambda_2-\lambda_1}+\frac{e^{-\lambda_2 T}}{\lambda_1-\lambda_2}\right)+\mathbf{A}_2(t)e^{-\lambda_2 T}$$

with $\mathbf{A}_1(t)$ given by Eq. (I.5), and $\mathbf{A}_2(t)$ by Eq. (I.7); and in general

$$\mathbf{A}_n(t, T)=\left(\prod_{i=1}^{n}\lambda_i\right)N_1^0 \times\sum_{i=1}^{n}\left[\sum_{j=1}^{i}\mathbf{a}_j e^{-\Lambda_j t}\sum_{k=i}^{n}a_k e^{-\lambda_k T}\right], \quad (I.12)$$

where

$\mathbf{a}_j=1$ when $i=1$, and when $i>1$,

$$\mathbf{a}_j=\prod_{m\neq j}(\Lambda_m-\Lambda_j)^{-1} \quad (m=1, 2, \cdots, i),$$

$a_k=1$ when $i=n$, and when $i<n$,

$$a_k=\prod_{p\neq k}(\lambda_p-\lambda_k)^{-1} \quad (p=i, i+1, \cdots, n).$$

II. THE ACTIVITIES OF THE DIFFERENT MEMBERS OF A DECAY CHAIN DESCENDED FROM A PRIMAL ANCESTOR WHICH IS CREATED AT A CONSTANT RATE

The problem we treat in this section will be discussed in terms of a fission product chain of which the primal ancestor is created at constant rate r by fission in a constant neutron flux ϕ.

If the flux is so small that $\phi\sigma_i\ll\lambda_i$ for all i, then the variation in number of nuclei of the different chain members is governed by the differential equations

$$\begin{cases}\dfrac{dN_1}{dt}=r-\lambda_1 N_1,\\ \cdot\\ \cdot\\ \cdot\\ \dfrac{dN_n}{dt}=\lambda_{n-1}N_{n-1}-\lambda_n N_n.\end{cases} \quad (II.1)$$

The corresponding set of equations for the high flux case is (in the notation of the preceding section)

$$\begin{cases}\dfrac{d\mathbf{N}_1}{dt}=r-\Lambda_1\mathbf{N}_1,\\ \cdot\\ \cdot\\ \cdot\\ \dfrac{d\mathbf{N}_n}{dt}=\lambda_{n-1}\mathbf{N}_{n-1}-\Lambda_n\mathbf{N}_n.\end{cases} \quad (II.2)$$

The solutions of these equations, like those of the preceding section, may be obtained without recourse to the formal methods used in solving differential equations.

We consider first the case of a high flux. Nuclei of the primal ancestor are produced by fission at constant rate r during a time interval $0\leqslant t'\leqslant\tau$. The activity $\mathbf{A}_1(\tau)$ of the primal ancestor at time τ is easily obtained from Eq. (I.6) with $R(t')=r$.

$$\mathbf{A}_1(\tau)=\lambda_1\mathbf{N}_1(\tau)=\frac{\lambda_1}{\Lambda_1}r(1-e^{-\Lambda_1\tau}). \quad (II.3)$$

The corresponding expression for $\mathbf{A}_n(\tau)$ can be most simply obtained by modifying the method of Eq. (I.6) in the following way.[4] The $R(t')dt'$ primal ancestor nuclei created during the time interval dt' at time t' $(0\leqslant t'\leqslant\tau)$ will give rise to a certain amount $d\mathbf{A}_n(t', \tau)$ of nth generation activity at time τ, i.e., at an amount of time $\tau-t'$ after the creation of the ancestral nuclei. If $R(t')=r=$constant, we have, in view of Eq. (I.9),

$$d\mathbf{A}_n(t', \tau)=\left(\prod_{i=1}^{n}\lambda_i\right)r dt'\sum_{i=1}^{n}\mathbf{a}_i\exp[-\Lambda_i(\tau-t')].$$

Integrating between $t'=0$ and $t'=\tau$, and multiplying and dividing through by

$$\prod_{i=1}^{n}\Lambda_i,$$

we get

$$\mathbf{A}_n(\tau)=\lambda_n\mathbf{N}_n(\tau)=\left(\prod_{i=1}^{n}\frac{\lambda_i}{\Lambda_i}\right)r \times\sum_{i=1}^{n}\mathbf{b}_i(1-e^{-\Lambda_i\tau}) \quad (n>1), \quad (II.4)$$

where

$$\mathbf{b}_i=\prod_{j\neq i}\frac{\Lambda_j}{\Lambda_j-\Lambda_i} \quad (j=1, 2, \cdots, n).$$

If the flux is shut off at time τ, fission stops, and the different activities $\mathbf{A}_i(\tau)$, present at that time in amounts given by Eq. (II.3) and the general Eq. (II.4), change subsequently by radioactive decay alone. With the lapse of t time units after flux shut off, the activity of the primal ancestor becomes

$$\mathbf{A}_1(\tau, t)=\mathbf{A}_1(\tau)e^{-\lambda_1 t}=\frac{\lambda_1}{\Lambda_1}r(1-e^{-\Lambda_1\tau})e^{-\lambda_1 t}; \quad (II.5)$$

the activity of the daughter becomes

$$\mathbf{A}_2(\tau, t)=\lambda_2\mathbf{A}_1(\tau)\left(\frac{e^{-\lambda_1 t}}{\lambda_2-\lambda_1}+\frac{e^{-\lambda_2 t}}{\lambda_1-\lambda_2}\right)+\mathbf{A}_2(\tau)e^{-\lambda_2 t},$$

where $\mathbf{A}_1(\tau)$ and $\mathbf{A}_2(\tau)$ are given by Eqs. (II.3) and (II.4), respectively, and in general, the activity of

[4] This is essentially the method used in Rutherford, Chadwick, and Ellis, *Radiations from Radioactive Substances* (Cambridge University Press, Teddington, 1930), p. 15.

the nth generation species becomes

$$\mathbf{A}_n(\tau, t) = r(\prod_{i=1}^{n}\lambda_i)\sum_{i=1}^{n}[(\prod_{j=1}^{i}\Lambda_j^{-1}) \times \sum_{j=1}^{i}\mathbf{b}_j(1-e^{-\Lambda_j\tau})\sum_{k=i}^{n}a_k e^{-\lambda_k t}], \quad (II.6)$$

where

$\mathbf{b}_j = 1$ when $i=1$, and when $i>1$,

$$\mathbf{b}_j = \prod_{m\neq j}\frac{\Lambda_m}{\Lambda_m-\Lambda_j} \quad (m=1, 2, \cdots, i),$$

$a_k = 1$ when $i=n$, and when $i<n$,

$$a_k = \prod_{p\neq k}(\lambda_p-\lambda_k)^{-1} \quad (k=i, i+1, \cdots, n).$$

The corresponding expression for the low flux case ($\phi\sigma_i \ll \lambda_i$) can be obtained in a much simpler form. The rdt' primal ancestor nuclei created during time interval dt' at time $t'(0\leqslant t'\leqslant\tau)$ will, at a time $\tau+t-t'$ later, have an activity

$$rdt' \exp[-\lambda_1(\tau+t-t')],$$

and the activity of the nth generation descendant at that later time will be, by Eq. (I.10),

$$(\prod_{i=1}^{n}\lambda_i)rdt'\sum_{i=1}^{n}a_i \exp[-\lambda_i(\tau+t-t')].$$

Integrating these expressions between $t'=0$ and $t'=\tau$, we get

$$A_1(\tau, t) = \lambda_1 N_1(\tau, t) = r(1-e^{-\lambda_1\tau})e^{-\lambda_1 t}, \quad (II.7)$$

$$A_n(\tau, t) = \lambda_n N_n(\tau, t) = r\sum_{i=1}^{n}b_i(1-e^{-\lambda_i\tau})e^{-\lambda_i t}, \quad (II.8)$$

where

$$b_i = \prod_{j\neq i}\frac{\lambda_j}{\lambda_j-\lambda_i} \quad (j=1, 2, \cdots, n).$$

III. THE ACTIVITY OF A RADIOACTIVE SPECIES WHICH IS CREATED AS A RESULT OF NEUTRON ABSORPTION BY ANOTHER RADIOACTIVE SPECIES

Suppose that nuclei of species u can absorb neutrons to generate nuclei of species v. Then, if nuclei of species u are created at constant rate r in a neutron flux of magnitude ϕ,

$$\frac{d\mathbf{N}_u}{dt} = r-\Lambda_u\mathbf{N}_u, \quad (III.1)$$

$$\frac{d\mathbf{N}_v}{dt} = \phi\sigma_u\mathbf{N}_u-\Lambda_v\mathbf{N}_v, \quad (III.2)$$

where σ_u is the cross section of species u for the creation of species v.[5]

[5] This is not necessarily the same as the σ_u in $\Lambda_u \equiv \lambda_u+\phi\sigma_u$ which may be the sum of several different cross sections in addition to the one in question.

These equations are identical in form with Eqs. (II.2) for $n=2$. The "parent" has a "decay constant" $\phi\sigma_u$ for the formation of the "daughter," and therefore we can write the expression for the activity of species v at time τ directly, by reference to Eq. (2.4) with $n=2$:

$$\mathbf{A}_v(\tau) = \lambda_v\mathbf{N}_v(\tau) = r_u\frac{\lambda_v(\phi\sigma_u)}{\Lambda_v\Lambda_u} \times\left[\frac{\Lambda_v}{\Lambda_v-\Lambda_u}(1-e^{-\Lambda_u\tau}) + \frac{\Lambda_u}{\Lambda_u-\Lambda_v}(1-e^{-\Lambda_v\tau})\right]. \quad (III.3)$$

The modes of reproduction differ, but the laws of population growth are the same.

This observation is of general validity. It permits us to write directly the equation for the activity of a species whose line of descent consists of any combination of decays and neutron absorptions. We have only to make an appropriate substitution of $\phi\sigma$'s for λ's in the appropiate equation derived in the preceding sections.

A few examples illustrating this procedure are given in the next section.

IV. SOME ILLUSTRATIVE PROBLEMS

As the first illustration, we will solve a fairly complicated case. Consider two two-membered fission product chains:

$$A_{11}\xrightarrow{\lambda_{11}}A_{12}\xrightarrow{\lambda_{12}}$$

$$A_{21}\xrightarrow{\lambda_{21}}A_{22}\xrightarrow{\lambda_{22}}$$

such that the members of the 1 chain can absorb neutrons to become corresponding members of the 2 chain. We suppose that each of the species has its own cross-section σ_{ij} and its own rate of direct formation in fission r_{ij}, and ask after the activity $\mathbf{A}_{22}(\tau)$ of the second member of the second chain at time τ (assuming $A_{ij}{}^0=0$).

Nuclei of species 22 are formed in all of the following ways:

(i) The A_{11} formed directly in fission (at rate r_{11}) decays to A_{12} which absorbs a neutron to give A_{22}:

$$A_{11}\xrightarrow{\lambda_{11}}A\xrightarrow{\lambda_{12}}{}_{12}$$

$$\downarrow\phi\sigma_{11}\quad\downarrow\phi\sigma_{12}$$

$$A_{22}\xrightarrow{\lambda_{22}}$$

$$\downarrow\phi\sigma_{22}.$$

Here we use the granddaughter equation with

daughter "decay constant" $\phi\sigma_{12}$:

$$\mathbf{A}_{22}{}^{(i)}(\tau)=\frac{\lambda_{22}(\phi\sigma_{12})\lambda_{11}}{\Lambda_{22}\Lambda_{12}\Lambda_{11}}r_{11}f_3(\tau),$$

where $f_3(\tau)$ is the expression under the summation sign in Eq. (II.4) with $n=3$.

(ii) The A_{11} formed directly in fission (at rate r_{11}) absorbs a neutron to give A_{21} which decays to A_{22}:

$$\begin{array}{l} A_{11}\overset{\lambda_{11}}{\longrightarrow} \\ \downarrow\phi\sigma_{11} \\ A_{21}\overset{\lambda_{21}}{\longrightarrow}A_{22}\overset{\lambda_{22}}{\longrightarrow} \\ \downarrow\phi\sigma_{21}\ \downarrow\phi\sigma_{22}. \end{array}$$

Here we again use the granddaughter equation, this time with parent "decay constant" $\phi\sigma_{11}$:

$$\mathbf{A}_{22}{}^{(ii)}=\frac{\lambda_{22}\lambda_{21}(\phi\sigma_{11})}{\Lambda_{22}\Lambda_{21}\Lambda_{11}}r_{11}f_3(\tau).$$

(iii) The A_{12} formed directly in fission (at rate r_{12}) absorbs a neutron to give A_{22}:

$$\begin{array}{l} A_{12}\overset{\lambda_{12}}{\longrightarrow} \\ \downarrow\phi\sigma_{12} \\ A_{22}\overset{\lambda_{22}}{\longrightarrow} \\ \downarrow\phi\sigma_{22}. \end{array}$$

Here we use the daughter equation with "parent decay constant" $\phi\sigma_{12}$:

$$\mathbf{A}_{22}{}^{(iii)}=\frac{\lambda_{22}(\phi\sigma_{12})}{\Lambda_{22}\Lambda_{12}}r_{12}f_2(\tau),$$

where $f_2(\tau)$ is the expression under the summation sign in Eq. (II.4) with $n=2$.

(iv) The A_{21} formed directly in fission (at rate r_{21}) decays to A_{22}:

$$\begin{array}{l} A_{21}\overset{\lambda_{21}}{\longrightarrow}A_{22}\overset{\lambda_{22}}{\longrightarrow} \\ \downarrow\phi\sigma_{21}\ \downarrow\phi\sigma_{22}. \end{array}$$

Here we use the regular daughter Eq. ((II.4), with $n=2$):

$$\mathbf{A}_{22}{}^{(iv)}=\frac{\lambda_{22}\lambda_{21}}{\Lambda_{22}\Lambda_{21}}r_{21}f_2(\tau).$$

(v) A_{22} is formed directly in fission at rate r_{22}:

$$\begin{array}{l} A_{22}\overset{\lambda_{22}}{\longrightarrow} \\ \downarrow\phi\sigma_{22}. \end{array}$$

Here we use Eq. (II.3),

$$\mathbf{A}_{22}{}^{(v)}(\tau)=r_{22}\frac{\lambda_{22}}{\Lambda_{22}}f_1(\tau).$$

The answer sought is the sum of the equations for the five paths of formation.

As a second illustration, we ask after the activity $A_\nu(\tau)$ of a radioactive species that results from ν successive neutron absorptions by a species created at constant rate r. The required equation is obtained directly from Eq. (II.4) on substitution of $\phi\sigma$'s for all the progenitor λ's:

$$\mathbf{A}_\nu(\tau)=\lambda_\nu\mathbf{N}_\nu(\tau)=r\frac{\lambda_\nu}{\Lambda_\nu}\left(\prod_{k=1}^{\nu-1}\frac{\phi\sigma_k}{\Lambda_k}\right)\sum_{i=1}^{\nu}\mathbf{b}_i(1-e^{-\Lambda_i\tau}),$$

$$\mathbf{b}_i=\prod_{j\neq i}\frac{\Lambda_j}{\Lambda_j-\Lambda_i}\quad(j=1,2,\cdots,\nu).$$

If all the members of the chain of neutron absorptions are stable against radioactive decay, we have

$$\mathbf{N}_\nu(\tau)=\frac{r}{\phi\sigma_\nu}\sum_{i=1}^{\nu}\mathbf{B}_i(1-e^{-\phi\sigma_i\tau}),$$

where

$$\mathbf{B}_i=\prod_{j\neq i}\frac{\sigma_j}{\sigma_j-\sigma_i}\quad(j=1,2,\cdots,\nu).$$

If, in addition, $\phi\sigma_i\tau\ll1$ for all i, the corresponding expression for $N_\nu(\tau)$ can be obtained (laboriously) by expanding the exponentials in the above equation to the νth order. It is easier to solve the problem directly. Assume all $N_i{}^0=0$. Then, for $r\gg\phi\sigma_1N_1$, and $\sigma_iN_i(t)\gg\sigma_{i+1}N_{i+1}(t)$,

$$\frac{dN_1}{dt}=r;\quad N_1(\tau)=r\tau$$

$$\frac{dN_2}{dt}=\phi\sigma_1N_1;\quad N_2(\tau)=r\phi\sigma_1\frac{\tau^2}{2!}$$

$$\vdots$$

$$\frac{dN_\nu}{dt}=\phi\sigma_{\nu-1}N_{\nu-1};\quad N_\nu(\tau)=r\phi^{\nu-1}\sigma_1\sigma_2\cdots\sigma_{\nu-1}\frac{\tau^\nu}{\nu!}.$$

As a final illustration, we shall solve a practical problem that has risen in the measurement of the neutron cross section of radioactive isotopes. Katcoff mentions parenthetically in a paper[6] that he has measured the (n,γ) cross-section of 85-m Ba^{139} by neutron bombardment of Ba^{138} in the pile. Let us

[6] S. Katcoff, Phys. Rev. 72, 1160 (1947). The work was completely reported in Manhattan Project Report CC-2908 (April 7, 1945).

derive the necessary equation by our procedure. The sequence of transformations was

$$\begin{array}{c}\text{stable } Ba^{138}\\ \downarrow\phi\sigma_s\\ \text{85-m } Ba^{139}\xrightarrow{\lambda_1}\\ \downarrow\phi\sigma_1\\ \text{12.5-d } Ba^{140}\xrightarrow{\lambda_2}\end{array}$$

where the notation is self-explanatory.

The activity of Ba^{140} after bombardment of Ba^{138} in a flux ϕ for a time τ can be calculated as follows. Let M be the number of Ba^{138} nuclei bombarded. Then the rate of formation of Ba^{139} is

$$r=\phi\sigma_s M.$$

Using Eq. (II.4) with $n=2$, and a "parent decay constant" $\phi\sigma_1$, we have for the Ba^{140} activity $\mathbf{A}_2(\tau)$,

$$\mathbf{A}_2(\tau)=\phi\sigma_s M\frac{\lambda_2(\phi\sigma_1)}{\Lambda_2\Lambda_1(\Lambda_2-\Lambda_1)}\times[\Lambda_2(1-e^{-\Lambda_1\tau})-\Lambda_1(1-e^{-\Lambda_2\tau})].$$

Now, $\Lambda_1\gg\Lambda_2$, and the circumstances of the experiment were such that τ was large, and $\lambda_2\gg\phi\sigma_2$, $\lambda_1\gg\phi\sigma_1$. Under these circumstances, the above equation becomes

$$\mathbf{A}_2(\tau)=\frac{\phi^2\sigma_s\sigma_1 M}{\lambda_1}(1-e^{-\lambda_2\tau}).$$

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Calculation of the Energy of Vaporization of Perfluorocyclopentane from Intermolecular Forces

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By assuming a probable representative orientation of perfluorocyclopentane molecules in the liquid state, the energy of vaporization has been calculated, using both the London-Margenau equation of dispersion forces and the Slater-Kirkwood formula. Using only the data from density and dispersion measurements, fair agreement was found to the value experimentally determined. It is seen on the basis of this calculation that the contribution of the dipole-quadrupole interaction to the London formula is important.

THE extremely low dispersion of the perfluoropentanes as described in a previous paper[1] is one of the types of evidence indicating very small intermolecular forces in fluorocarbons. Of these compounds, perfluorocyclopentane, because of its symmetrical structure, lends itself more easily to theoretical treatment than either the linear or branched chain molecules. On this account this substance has been chosen as a material whose experimental energy of vaporization could be compared with that calculated using existing theories of intermolecular attraction and the physical constants determined in these laboratories.

The molar energy of vaporization U of a liquid represents the energy required to separate all the molecules of a gram mole of that liquid. If we let ΣV_i be the total attractive energy between a given molecule and all of its neighbors, then the molar energy of vaporization would be given by the relation

$$U=-\tfrac{1}{2}N\sum_{i=1}^{N}V_i, \qquad (1)$$

where N is Avogadro's number and $\frac{1}{2}$ is included to avoid counting each bond twice. Equation (1) assumes, of course, that the force acting on every molecule is the same, which cannot be the case for the surface molecules. However, for any large number of molecules being considered, this factor may be neglected. This equation further neglects the small configurational energy of the vapor and also assumes that there is no difference between the kinetic energies in the liquid and vapor phases.

Attraction between the non-polar molecules of perfluorocyclopentane must be due entirely to van der Waals forces of the dispersion type. As a first approximation these forces are known to be proportional to the inverse 6th power of the distance R, such a relation for the attraction between two spherical atoms being first derived by London.[2] It is generally recognized at present that there is attractive contribution from still higher inverse terms, and a better representation for the attractive potential is given by[3]

$$V=-C_1/R^6-C_2/R^8-C_3/R^{10}, \qquad (2)$$

[1] G. H. Rohrback and G. H. Cady, J. Am. Chem. Soc., to be published.

[2] F. London, Zeits. f. physik. Chemie **B11**, 222 (1930).

[3] H. Margenau, Rev. Mod. Phys. **11**, 1 (1939). This paper is an excellent review of the theories of intermolecular forces