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# A TWO STEP MODEL OF CLUSTER FORMATION AND BARRIER PENETRATION IN RADIOACTIVE NUCLEI

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Abstract: A new model is proposed for the mechanism of first the cluster formation and then penetration of the confining nuclear interaction barrier in radioactive nuclei. The clustering formation probability is given by the quantum mechanical fragmentation probability at the touching configuration, which de-excites itself to the ground state during its penetration through the interaction barrier. The WKB penetrability is solved analytically. Applications of the model are made to C-14 decay of Ra-222-224 and Ne-24 decay of U-232.

(radioactive decay, heavy clusters, theory, clustering formation and decay, analytical solution)

## Introduction

A new phenomenon of cluster emission in spontaneous decay of radioactive nuclei has been observed very recently /1/. Clusters like C-14 and Ne-24 have been observed to be emitted from Ra-222-226 and U-232,233, Th-230 and Pa-231 /2-10/. These new decay modes, intermediate between alpha decay and nuclear fission, were predicted much before they were actually seen in laboratory /11/. Various theoretical attempts have been made to understand this exotic new decay /11-16/. Mainly two different approaches have been used : (i) using Gamow's theory of $\alpha$-decay, where recently both the formation and decay processes have been studied /13/, (ii) the normal nuclear fission models. Also, a new mechanism of alpha nucleus C-12 transforming to C-14 by picking up two neutrons on way to tunneling, has been studied /14/. However, the formation amplitude of alpha-particle is so small that heavier clusters can not be considered as simple aggregates of alpha-partices moving outside the nuclear core.

In this paper, we propose a new mechanism for the clustering formation in nuclei, as the quantum mechanical fragmentation process, and give an analytical method for calculating the WKB penetrability of the confining nuclear interaction barrier. In our model, the coupled motions of mass- asymmetry and relative separation coordinates, in the decoupled approximation, give the two steps of clustering formation and tunneling of the interaction barrier.

## The Model

We consider step (i) the formation of two fragments (the cluster and the daughter nuclei) in their ground states with probability Po, described by the dynamical collective coordinates of mass and charge asymmetries of the two fragments /17-21/

$$\eta=\frac{A_1-A_2}{A} \quad and \quad \eta_Z=\frac{Z_1-Z_2}{Z} \tag{1}$$

and step (ii) the tunneling of the confining nuclear interaction barrier V(R) with probability P by impinging on it with frequency $\nu$ . In principle these two steps, describing the $\eta$ and R motions, are coupled. However, it has been shown by Gupta and collaborators /22,23/ that the coupling effects of relative motion to asymmetry coordinates in the potential are very small. Also, it is known /17-19/ that the cranking coupling masses $B_{R\eta}$ and $B_{R\eta_Z}$ are very small such that $B_{R\eta} \ll (B_{RR}\ B_{\eta\eta})^{1/2}$ and $B_{R\eta_Z} \ll (B_{RR}\ B_{\eta_Z\eta_Z})^{1/2}$. In view of these results, we treat the two motions in a decoupled approximation and define the decay constant or half-life time for a metastable system, as

$$\lambda = P_0 \nu P \quad or \quad T = \ln 2/\lambda \tag{2}$$

### The Clustering Formation Probability Po

We define the pre-formation probability as a quantum mechanical probability of finding the fragments $A_1$ and $A_2$ (with fixed charges $Z_1$ and $Z_2$, respectively) at a point R of the relative motion. For this purpose we solve the stationary Schrödinger equation in $\eta$ at fixed $\eta_Z$ and R :

$$\left[-\frac{\hbar^2}{2\sqrt{B_{\eta\eta}}}\frac{\partial}{\partial\eta}\frac{1}{\sqrt{B_{\eta\eta}}}\frac{\partial}{\partial\eta}+V(\eta)\right]\psi^{(\nu)}_{R\eta_Z}(\eta)=E^{(\nu)}_R\psi^{(\nu)}_{R\eta_Z}(\eta) \tag{3}$$

Then, on proper scaling and normalizing the solution of (3), we get for the formation probability in the ground state,

$$P_0=|\psi^{(0)}_{R\eta_Z}(\eta)|^2\sqrt{B_{\eta\eta}}\ \frac{4}{A} \tag{4}$$

For the fragmentation potential V( $\eta$ ) in (3), we also use the two spheres approximation. This approximation simplifies the calculations and is justified in view of our earlier calculations of V($\eta$) at different R- values, using two-centre shell model (TCSM ) in Strutinsky method for the overlap regions and the experimental binding energies for asymptotic R (see e.g. Fig.5 in ref. 24; Fig. 1 in ref. 25 or Fig. 1 in ref. 26). It is shown clearly that the general shape of the potential V( $\eta$ ), including the positions of all the potential energy minima, is independent of the the choice of R- value. Then, for two touching (or overlapping ) spheres, we define V( $\eta$ ) as sum of the experimental binding energies, Coulomb interaction and the proximity potential /27/,

$$V(\eta,R)=-\sum_{i=1}^{2}B_i(A_i,Z_i)+\frac{Z_1Z_2e^2}{R}+V_P \tag{5}$$

Here the charges $Z_i$ are fixed by minimizing the sum of the two binding energies in charge asymmetry coordinate $\eta_Z$ .

For mass parameters $B_{\eta\eta}(\eta)$ in (3), we use the classical model of Kröger and Scheid /28/.

## The Assault Frequency

For the assault or escape frequency $\nu$ , we use the simlpe relation

$$\nu = \frac{v}{R_o} = (2E_2/\mu)^{1/2}/R_o \quad with \quad E_2 = \frac{A_1}{A} Q \tag{6}$$

Here Ro is the radius of parent nucleus, $E_2$ the kinetic energy of emitted cluster, $Q=E_1+E_2$.

## The Tunneling Probability P

We use the WKB approximation and calculate the tunneling probability analytically. The nuclear interaction potential V(R), calculated from Eq. (5), is illustrated in Fig. 1 (solid lines) for $^{222}Ra \to {}^{14}C + {}^{208}Pb$. For $R < R_t$ , we simply join smoothly the potential calculated at $R=R_t$ to the Q-value at the parent nucleus radius, R=Ro. For the region $Ro < R < R_t$ , we are not interested in the actual shape of the potential since our calculations below suggest to use $R=R_t$ for the evaluation of the pre-formation factor. In other words, the first (inner) turning point in the WKB penetrability integral is chosen at $R=R_t$ . The outer (second) turning point, $R=R_b$, is taken to give the Q-value of the reaction i.e. $V(R_b)=0$.

We notice in Fig. 1 that for the inner and outer turning points, respectively, at $R=R_t$ and $R_b$ the transmission probability P consists of three conributions: the penetrability $P_i$ from $R_t$ to $R_i$, the de-excitation probabilty $W_i$ at $R_i$ and then the penetrabilty $P_b$ from $R_i$ to $R_b$ . Thus,

$$P = P_i W_i P_b \tag{7}$$

Following M. Greiner and W. Scheid /15/ (see also Ref. 14,29) the choice of starting the tunneling process at an energy $V(R_t)$ i.e. above the Q value, is a decay into the excited states of the daughter nucleus (or the cluster or both). These authors suggest to scale the de- excitation probability $W_i$ , exponentially with the excitation energy $E_i$,

$$W_i = exp(-bE_i) \tag{8}$$

This apparently means that the de-excitation process is restricted to only first order transitions. The ansatz (8) is applied to $\alpha$-decay of Ra-222,224 and U-232 and it is shown /15/ that for $R_i$ -values of interest, the parameter b must be very small. Therefore, for heavy cluster emission, the authors assumed b=0, which means $W_i = 1$. Then, Eq.(7) reduces to

$$P = P_i P_b \tag{9}$$

where, in WKB theory, the penetrabilities $P_i$ , $P_b$ are defined as

$$P_i = exp\left(-\frac{2}{\hbar}\int_{R_t}^{R_i}\{2\mu[V(R)-V(R_i)]\}^{1/2} dR\right) \tag{10}$$

$$P_b = exp\left(-\frac{2}{\hbar}\int_{R_i}^{R_b}\{2\mu[V(R)-Q]\}^{1/2} dR\right) \tag{11}$$

We solve the integrals in Eqs. (10) and (11) analytically. For this purpose we parameterize the potential V(R), calculated from Eq. (5) and illustated in Fig. 1, as follows :

$$V(R) = \begin{cases} V(R_t) + s(R-R_t) & R_t \le R \le R_s \\ V_B - \frac{1}{2}k(R-R_B)^2 & R_s \le R \le R_h \\ V(R_h) - C_1(R-R_h)/R & R_h \le R \le R_i \\ V(R_i) - C_2(R-R_i)/R & R_i \le R \le R_b \end{cases} \tag{12}$$

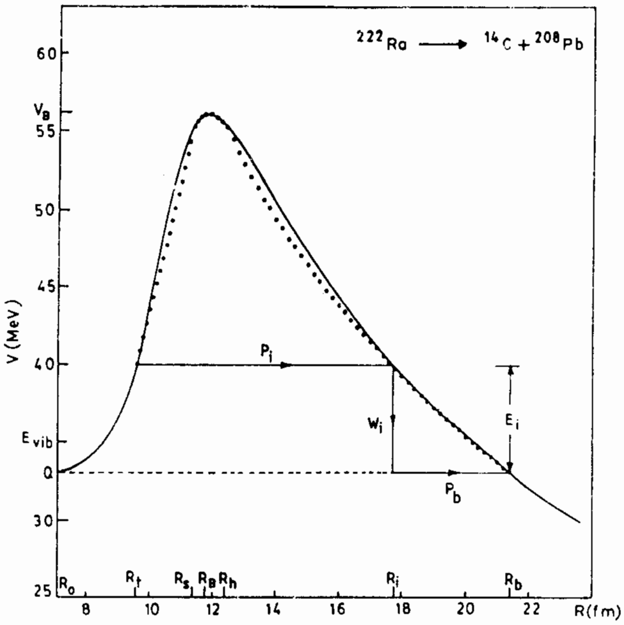


Fig.1 The calculated and fitted nuclear interaction potential for $^{222}Ra \to {}^{14}C + {}^{208}Pb$ and the path of tunneling.

Then, using the fact that

$$\int_{R_t}^{R_i} f(R)\,dR = \int_{R_t}^{R_s} f(R)\,dR + \int_{R_s}^{R_h} f(R)\,dR + \int_{R_h}^{R_i} f(R)\,dR \tag{13}$$

we get $P_i$ and $P_b$ and hence the analytical expression for the tunneling probability P. This in turn, substituted in (2) along with (4) and (6), gives the decay constant $\lambda$ or the half-life time T.

## Calculations and Results

### The Relative Cluster Formation Probability

In this section, we have first calculated the fragmentation potentials $V(\eta)$ at various R-values, starting from touching configuration ($R=R_t$ ) to a large overlap of two spheres. This is illustrated in Fig. 2 for Ra-222. We notice that the positions and depths of all potential energy minima are almost independent of the R value. Similar results are obtained for other nuclei /30/. In each case, deep potential energy minima are found to occur at the usual fission fragments (e.g. Kr-88 and its complementary fragment Te-134 in Ra-222) and He-4, Be-10, C-14 and Ca-46-50 clusters. For U-232, a deep minimum also appears at Ne-24. It is also shown that inclusion of proximity potential does not bring any alteration in these minima, except that the minima at large mass asymmetry become slightly deeper. This evidently means that the potential energy minima, determining the possible clustering formation (and decay) channels are only due to shell effects /30/.

Table 1 gives our calculated pre-formation probabilities Po for C-14 and $\alpha$-particle in Ra-222 at different R-values (using the potentials of Fig. 2 with $V_P$ added and the classical mass parameters of Ref. 28). We notice that $P_0$ depends strongly on position R of the two fragments (cluster and daughter nuclei). At $R=R_1+R_2-2.2$ i.e. when the overlap of the cluster with the daughter nucleus is large, the formation yield for C-14 is zero and very small ($\sim 10^{-10}$) for $\alpha$-particle. However, it is interesting to observe

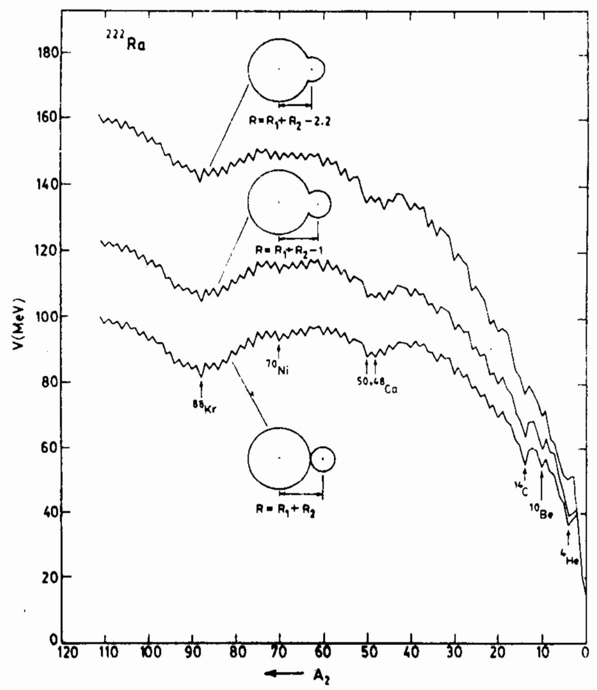


Fig.2 The fragmentation potential $V(\eta)$ for Ra-222, with $V_P=0$.

**Table 1. Cluster Preformation Probabilities relative to one.**

| R (fm) | $Po(^{14}C)$ | $Po(\alpha)$ | $\frac{Po(^{14}C)}{Po(\alpha)}$ |
|---|---|---|---|
| $R_1+R_2$ | $1.4\times10^{-14}$ | $9.9\times10^{-8}$ | $1.4\times10^{-7}$ |
| $R_1+R_2-0.5$ | $4.1\times10^{-15}$ | $1.1\times10^{-7}$ | $0.4\times10^{-7}$ |
| $R_1+R_2-1$ | $2.3\times10^{-6}$ | $9.3\times10^{-1}$ | $0.3\times10^{-7}$ |
| $R_1+R_2-2.2$ | 0 | $3.9\times10^{-10}$ | - |

that as the overlap of fragments decreases (the relative separation R increases), the relative cluster preformation yield with respect to $\alpha$-particle, i.e. the ratio Po (C-14)/Po ($\alpha$) remains almost constant. Interpreting this result in terms of zero point vibration energy Evib, introduced by Poenaru et al /31/ empirically to fit the cluster decay half-life times, we find that the configuration of larger overlap ($R=R_1+R_2-2.2$) refer to an excitation energy below Evib but the ones with smaller overlaps ($R=R_1+R_2-1$ to $R_1+R_2$) above it. This suggests that the clustering formation begins at relative position corresponding to $V(R)\approx$ Q+Evib and then the cluster formation probability relative to $\alpha$-particle remains almost the same upto the touching configuration. This result allows us to choose the touching configuration ($R=R_t$) as our starting point for the tunneling process. In addition to simplifying the calculations of relative cluster preformation yields, this choice of R has the added advantage of not including the un-determined part of the scattering potential from Ro to $R_t$ in our analytical calculations of the tunneling probabilities.

Table 2 gives our calculated relative cluster preformation yields with respect to $\alpha$-particle at $R=R_t$ for all the nuclei. The results of the calculations of other authors are also given here for comparisions. We notice that our calculations agree within an order of magnitude with all the authors, except with Blendowske et al /16/. Furthermore, in agreement with Iriondo et al /13/, our calculations also show, atleast qualitatively, a decrease in pre-formation probability with increase of cluster size.

## The Half-life Times

We have compared in Table 3, our calculated half-lives with other calculations and the experimental data. We notice that our calculations and that of Blendowske et al /16/, both based on two -step mechanism of clustering formation and barrier penetration, give better comparisions with experiments for C-14 decay of Ra nuclei and that the fission calculations of Poenaru et al /32/ compare more favourably for Ne-24 decay of U-232. In this connection, it may be relevant to note that for this decay the measured spontaneous fission branching ratio /33/ $\lambda$(Ne-24)/ $\lambda$ ($\alpha$)=$1.2\times10^{-12}$ is comparable with the recently measured /6/ cluster decay branching ratio $\lambda$(Ne-24)/ $\lambda$ ($\alpha$)=$(2.0\pm0.5)\times10^{-12}$. This points out that the predominant phenomenon in Ne-24 decay of U-232 may be fission rather than the cluster- decay. Considering the fission of U-232 as a dynamical mass fragmentation process /17/, we have estimated the yield for Ne-24 fragment relative to $\alpha$-particle at the touching configuation using classical mass parameters. This comes out to be $\sim10^{-11}$, which compares rather well with other fission calculations /12,32/ and lie within a factor of 10 of the experimental data. This point certainly needs a further study.

## Summary

We have shown that the cluster radioactive-decay can be very nicely considered as a two step, decoupled mechanism of clustering formation and tunneling of the confining nuclear interaction barrier, at least for the lighter

**Table 2. Relative Cluster Formation Probabilities Po(cluster)/Po($\alpha$).**

| Nucleus | Emitted cluster | Present work | Ref.2 | Ref.14 | Ref.13 | Ref.16 |
|---|---|---|---|---|---|---|
| $^{222}Ra$ | $^{14}C$ | $1.4\times10^{-7}$ | | $\sim10^{-7}$ | $1.1\times10^{-5}$ | $3.1\times10^{-10}$ |
| $^{223}Ra$ | $^{14}C$ | $5.5\times10^{-8}$ | $7\times10^{-5}-4\times10^{-7}$ | | $9.6\times10^{-6}$ | $2.8\times10^{-10}$ |
| $^{224}Ra$ | $^{14}C$ | $1.7\times10^{-8}$ | | | $7.3\times10^{-6}$ | $1.9\times10^{-10}$ |
| $^{232}U$ | $^{24}Ne$ | $1.9\times10^{-11}$ | | | $4.0\times10^{-10}$ | |

Table 3. Calculated and Experimental Cluster-decay Half-life Times.

| Decay | Experiments | | Calculated values of log(T) | | | |
|---|---|---|---|---|---|---|
| | Ref. | log(T) | Present work | Ref.16 | Ref.32 | Ref.29 |
| $^{222}Ra \rightarrow ^{14}C + ^{208}Pb$ | 5),8) | 10.9-11.1 | 11.2 | 11.0 | 12.6 | 12.4 |
| $^{223}Ra \rightarrow ^{14}C + ^{209}Pb$ | 2-5),7) | 14.9-15.5 | 14.1 | 15.2 | 14.8 | 14.5 |
| $^{224}Ra \rightarrow ^{14}C + ^{210}Pb$ | 5) | 15.8-16.0 | 15.0 | 15.9 | 17.4 | 17.1 |
| $^{232}U \rightarrow ^{24}Ne + ^{208}Pb$ | 6) | 21.3-21.5 | 16.5 | | 20.4 | |

cluster like C-14. For the heavier clusters, perhaps the fission process is more predominant.

The clustering formation in our model is treated as the quantum mechanical mass- and charge- fragmentation process. The static fragmentation potential $V(\eta, R)$ calculations clearly show that, independent of R-value, the possible cluster formation (and decay) channels are fixed by shell effects in it. The dynamical clustering formation is shown to begin at the relative separation corresponding to $V(\eta, R) \approx Q + E_{vib}$ and its probability with respect to $\alpha$-particle formation is found to remain nearly constant upto a separation distance corresponding to touching configuration.

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