Theoretical interpretation of anomalous tritium and neutron productions during Pd/D co-deposition experiments

Y.E. Kim

Department of Physics, Purdue University, Physics Building, West Lafayette, IN 47907, USA

1 Introduction

Recently, a series of experimental results have been reported on observation of triple tracks in solid-state nuclear track detectors, CR-39, during Pd/D co-deposition experiments [1–4]. Most recent results show that triple tracks in CR-39 detectors observed in Pd/D co-deposition experiments are indistinguishable from those generated upon exposure to a DT neutron source [4]. This experimental observation of “nuclear ashes” is important and significant in establishing the fact that these phenomena observed in the Pd/D co-deposition experiments are due to nuclear reactions.

There have been many reports of anomalous tritium and neutron productions in deuterated metal from electrolysis experiments [5–9] and gas/plasma loading experiments [10–16]. The reported branching ratio of neutron productions during Pd/D co-deposition experiments are due to nuclear reactions.

The reported branching ratio of [10–16]. The reported branching ratio of trolysis experiments [5–9] and gas/plasma loading experi-

ments [1–4]. Most recent results show that triple tracks in solid-state nuclear track detectors, CR-39, during Pd/D co-deposition experiments indicate that the triple tracks are due to ~14 MeV neutrons, which appear to originate from “hot” fusion reaction D(t,n)^4He. Nuclear theory interpretation of the origin of ~14 MeV neutrons is presented in terms of a sub-threshold resonance reaction involving (T + p) resonance state of ^4He(J^π = 0^+) at 20.21 MeV, which produces 1.01 MeV T. An upper limit of the branching ratio, R(n)/R(T), between neutron production rate and tritium production rate is calculated to be R(n)/R(T) < 10^{-4}. Experimental tests of the proposed theoretical interpretation are proposed.

2 Present status of anomalous experimental results

The conventions deuterium fusion in free space proceeds via the following nuclear reactions:

1} D + D \rightarrow p (3.02 MeV) + T (1.01 MeV);
2} D + D \rightarrow n (2.45 MeV) + ^3He (0.82 MeV); and
3} D + D \rightarrow ^4He + \gamma (23.8 MeV).

The cross-sections (or reaction rates) for reactions \{1\} and \{2\} have been measured by beam experiments at intermediate energies (≥ 10 keV). The cross-sections for reaction \{1\}–\{3\} are expected to be extremely small at low energies (≤ 10 eV) due to the Gamow factor arising from Coulomb barrier between two deuterons. The measured cross-sections have branching ratios: (σ\{1\}, σ\{2\}, σ\{3\}) ≈ (0.5, 0.5, 10^{-3}).

Experimental values of the conventional hot-fusion cross section σ(E) for reaction \{1\} or \{2\} have been conventionally parameterized as [17]:

σ(E) = \frac{S(E)}{E} \exp(-2\pi\eta) = \frac{S(E)}{E} \exp\left(-\frac{(E_G/E)^{1/2}}{2}\right)

(1)

with η = Z_1 Z_2 e^2/\hbar c. Exp (−2πη) is known as the “Gamow factor”, and E_G is the “Gamow energy” given by E_G = (2\pi\alpha Z_D Z_D)^2 M c^2/2 or E_G^{1/2} ≈ 31.39 (keV)^{1/2} for the reduced mass M ≈ M_D/2 for reactions \{1\} or \{2\}. The value E is measured in keV in the center-of-mass (CM) reference frame. The S-factor, S(E), is extracted from experimentally measured values [18] of the cross-section σ(E) for E ≥ 4 keV and is nearly constant [19]; S(E) ≈ 52.9 keV-barn, for reactions \{1\} or \{2\} in the energy range of interest here, E ≤ 100 keV. The S-factor is known as “astrophysical S-factor” [17].
From many experimental measurements by Fleischmann and Pons [20], and many others [5–16,20–23] over 20 years since then, the following experimental results have emerged. At ambient temperatures or low energies (≤ 10 eV), deuterium fusion in metal proceeds via the following reactions:

{4}\begin{equation}D(m) + D(m) \rightarrow p(m) + T(m) + 4.03 \text{ MeV} (m);\end{equation}

{5}\begin{equation}D(m) + D(m) \rightarrow n(m) + ^{3}\text{He}(m) + 3.27 \text{ MeV} (m);\end{equation}

and

{6}\begin{equation}D(m) + D(m) \rightarrow ^{4}\text{He}(m) + 23.8 \text{ MeV} (m),\end{equation}

where \(m\) represents a host metal lattice or metal particle. Reaction rate \(R\) for {6} is dominant over reaction rates for {4} and {5}, i.e., \(R(6) \gg R(4)\) and \(R(6) \gg R(5)\).

Experimental observations reported from electrolysis and gas-loading experiments are summarized below (not complete):

(1) The Coulomb barrier between two deuterons are suppressed.

(2) Excess heat production (the amount of excess heat indicates its nuclear origin).

(3) \(^{4}\text{He}\) production commensurate with excess heat production, no 23.8 MeV \(\gamma\)-ray.

(4) More tritium is produced than neutron \(R(4) \gg R(5)\).

(5) Production of nuclear ashes with anomalous rates:

\begin{itemize}
  \item D/metal \(> 0.9\), electric current, pressure gradient, etc.,
  \item Requirement of deuteron mobility \((D/Pd \gg 1)\).
\end{itemize}

All of the above experimental observations are explained either quantitatively or qualitatively in terms of theory of Bose-Einstein condensation nuclear fusion (BECNF) in previous publications [24,25]. In this paper, additional theoretical interpretation is described more in details for observation (4), which involve anomalous production of tritium and neutrons.

### 3 Deuteron mobility and Bose-Einstein condensation of deuterons in metals

Deuteron fusion in metal is based upon a single hypothesis that deuterons in metal are mobile and hence are capable of forming Bose-Einstein condensates.

#### 3.1 Deuteron mobility and high-density deuteron-electron plasma in metal

Experimental proof of proton (deuteron) mobility in metals was first demonstrated by Coehn in his hydrogen electromigration experiment [26,27]. A theoretical explanation of Coehn’s results [26] is given by Isenberg [28]. The Coehn’s experimental fact is not well known in review articles and textbooks.

There are other experimental evidences [29–33] that heating and/or applying an electric field in a metal causes hydrogens and deuterons in a metal to become mobile, thus leading to a higher density for quasi-free mobile deuterons in a metal. It is expected that the number of mobile deuterons will increase, as the loading ratio \(D/metal\) of deuterium atoms increases and becomes larger than one, \(D/metal\) ≥ 1.

Mobility of deuterons in a metal is a complex phenomenon and may involve a number of different processes [33]: coherent tunneling, incoherent hopping, phonon-assisted processes, thermally activated tunneling, and over-barrier jump/fluid like motion at higher temperatures. Furthermore, applied electric fields as in electrolysis experiments can enhance the mobility of absorbed deuterons.

The physical significance of Coehn’s results [26] is that a deuterium atom is ionized to become deuteron and an electron, thus creating a high-density deuteron-electron plasma in metal. For the case of PdD, deuteron-electron plasma density of ~1022 cm−3 is achieved in metal.

#### 3.2 BEC fraction of deuterons in metal

Fraction of deuterons in a metal satisfying BEC condition can be estimated as a function of the temperature. The BEC condensate fraction \(F(T) = N_{BE}/N\) can be calculated from integrals:

\[N_{BE} = \int_{0}^{E_{C}} n(E)N(E)dE\]

where \(n(E)\) is either Bose-Einstein or Maxwell-Boltzmann distribution function, \(N(E)\) is the density of (quantum) states, and \(E_{C}\) is the critical kinetic energy of deuteron satisfying the BEC condition \(\lambda_{c} = d\), where \(\lambda_{c}\) is the de Broglie wavelength of deuteron corresponding to \(E_{C}\) and \(d\) is the average distance between two deuterons. For \(d = 2.5\ \text{Å}\), we obtain \(F(T = 300 \text{ K}) \approx 0.084 (8.4\%), F(T = 77.3 \text{ K}) \approx 0.44 (44\%),\) and \(F(T = 20.3 \text{ K}) \approx 0.94 (94\%).\) At \(T = 300 \text{ K}, F = 0.084 (8.4\%)\) is not large enough to form BEC since motions of deuterons are limited to several lattice sites and the probability of their encounters are very small. On the other hand, at liquid nitrogen (77.3 K) and liquid hydrogen (20.3 K) temperatures, probability of forming BEC of deuterons is expected to be \(\Omega \approx 1\). This suggests that experiments at these low temperatures can provide tests for enhancement of the reaction rate \(R_t\), equation (4) below, as predicted by BECNF theory.

### 4 Nuclear theory and theoretical interpretation of experimental data

#### 4.1 Bose-Einstein condensation theory of deuteron fusion in metal

In developing the BEC theory of deuteron fusion in metal, we make one basic assumption that mobile deuterons in
a micro/nano-scale metal particle form a BEC state. The validity of this assumption is to be verified by independent experimental tests suggested in this paper. Because of the above assumption, the theory cannot be applied to deuterons in bulk metals, which do not provide well-defined localized trapping potentials for deuterons.

For applying the concept of the BEC mechanism to deutron fusion in a nano-scale metal particle, we consider \( N \) identical charged Bose nuclei (deuterons) confined in an ion trap (or a metal grain or particle). Some fraction of trapped deuterons are assumed to be mobile as discussed above. The trapping potential is 3-dimensional (nearly-sphere) for nano-scale metal particle, or quasi 2-dimensional (nearly hemi-sphere) for micro-scale metal grains, both having surrounding boundary barriers. The barrier heights or potential depths are expected to be an order of energy (\( \leq 1 \) eV) required for removing a deutron from a metal grain or particle. For simplicity, we assume an isotropic harmonic potential for the ion trap to obtain order of magnitude estimates of fusion reaction rates.

\( N \)-body Schroedinger equation for the system is given by:

\[
H \Psi = E \Psi \tag{2}
\]

with the Hamiltonian \( H \) for the system given by:

\[
H = \frac{\hbar^2}{2m} \sum_{i=1}^{N} \Delta_i + \frac{1}{2} m \omega^2 \sum_{i=1}^{N} r_i^2 + \sum_{i< j} e^2 |r_i - r_j| \tag{3}
\]

where \( m \) is the rest mass of the nucleus. Only two-body interactions (Coulomb and nuclear forces) are considered since we expect that three-body interactions are expected to be much weaker than the two-body interactions.

Electron degrees of freedom are not explicitly included, assuming that electrons and host metal atoms provide a host trapping potential. In presence of electrons, the Coulomb interaction between two deuterons can be replaced by a screened Coulomb potential in equation (3). Hence, equation (3) without the electron screening effect represents the strongest case of the reaction rate suppression due to the coulomb repulsion.

The approximate ground-state solution of equation (2) with \( H \) given by equation (3) is obtained using the equivalent linear two-body method [34,35]. The use of an alternative method based on the mean-field theory for bosons yields the same result (see Appendix in [36]). Based on the optical theorem formulation of low energy nuclear reactions [37], the ground-state solution is used to derive the approximate theoretical formula for the deuteron-deuteron fusion rate in an ion trap (micro/nano-scale metal grain or particle). The detailed derivations are given elsewhere including a short-range nuclear strong interaction used [36,38].

Our final theoretical formula for the nuclear fusion rate \( R_{\text{trap}} \) for a single trap containing \( N \) deuterons is given by [24]:

\[
R_{\text{trap}} = 4 \left( \frac{3}{4\pi} \right)^{3/2} \Omega S B \frac{N^2}{D_{\text{trap}}} \times \frac{\Omega}{D_{\text{trap}}^2} \tag{4}
\]

where \( N \) is the average number of Bose nuclei in a trap/cluster, \( D_{\text{trap}} \) is the average diameter of the trap, \( B = 2r_B/(\pi \hbar) \), \( r_B = \hbar^2/(2m \epsilon^2) \), and \( S \) is the \( S \)-factor for the nuclear fusion reaction between two deuterons, as defined by equation (1). For \( D(d,p)T \) and \( D(d,n)^{10} \)He reactions, we have \( S \approx 55 \) keV-barn. We expect also \( S \approx 55 \) keV-barn or larger for reaction [6], \( B = 1.4 \times 10^{-18} \) cm\(^3\)/s with \( S \) in units of keV-barn in equation (4).

The total fusion rate \( R_t \) is given by:

\[
R_t = N_{\text{trap}} R_{\text{trap}} = \frac{N_D}{N} R_{\text{trap}} \propto \frac{\Omega N^2}{D_{\text{trap}}^2} \tag{5}
\]

where \( N_D \) is the total number of deuterons and \( N_{\text{trap}} = N_D/N \) is the total number of traps.

Equation (5) shows that the total fusion rates, \( R_t \), are very large if \( \Omega \approx 1 \).

The total reaction rate \( R_t \) for each exit reaction channel can be calculated for given values of \( \Omega \) and \( S \), using equations (4) and (5). The \( S \)-factor can be either inferred from experimental data or can be calculated theoretically using equation (9) (see Sect. 4.3 and Appendix). The branching ratio between two different exit reaction channels can be obtained as the ratio between two \( S \)-factors (see Tab. 2 in Sect. 5.1).

Equations (4) and (5) provide an important result that nuclear fusion rates \( R_{\text{trap}} \) and \( R_t \) do not depend on the Gamow factor in contrast to the conventional theory for nuclear fusion in free space. This could provide explanations for overcoming the Coulomb barrier and for the claimed anomalous effects for low-energy nuclear reactions in metals. This is consistent with the conjecture noted by Dirac [39] and used by Bogoliubov [40] that boson creation and annihilation operators can be treated simply as numbers when the ground state occupation number is large. This implies that for large \( N \) each charged boson behaves as an independent particle in a common average background potential and the Coulomb interaction between two charged bosons is suppressed. This provides an explanation for the observation (1). There is a simple classical analogy of the Coulomb field suppression. For an uniform charge distribution in a sphere, the electric field is a maximum at the surface of the sphere and decreases to zero at the center of the sphere.

4.2 Sub-threshold resonance reactions

In this section, we present a theoretical explanation of this anomalous tritium production based on the BECNF theory, utilizing a sub-threshold resonance \(^{4}\)He\(^{+} \) (0\(^{+}\)) state at 20.21 MeV with a resonance width of \( \Gamma(T + p) = 0.5 \) MeV as shown in Figure 1.

In Figure 1, reaction channels \([4], [5], \) and \([6]\) described in Section 2 are shown. Entrance channel \([7]\), and exit channels \([7a]\) and \([7b]\) (described below) are also
The exit reaction channels for D + D fusion in Bose-Einstein condensate state. Parallel bars indicate break in energy scale. Due to a selection rule derived in reference [24], both \{4\} and \{5\} channels are suppressed, and we have \( R(4) \ll R(5) \ll R(6) \). In free space, \{6\} would be forbidden due to the momentum conservation, while \{7\} would satisfy the momentum conservation for \( Q = 0 \). For this section (Eqs. (8)–(11)), we use a new energy level scale which sets \( E = 0 \) for \((\text{D} + \text{D})\) state, and \( E = -23.85 \text{ MeV} \) for the \(^4\text{He}\) ground state. For a single trap (or metal particle) containing \( N \) deuterons, the deuteron-deuteron fusion can proceed with the following two reaction channels:

\[
\{6\} \psi_{\text{BEC}} \{ (N - 2) \text{D}'s + (\text{D} + \text{D}) \} \rightarrow \psi^* \{ ^4\text{He} (0^+, 0) + (N - 2) \text{D}'s \} \quad (6)
\]

and

\[
\{7\} \psi_{\text{BEC}} \{ (N - 2) \text{D}'s + (\text{D} + \text{D}) \} \rightarrow \psi^* \{ ^4\text{He}^* (0^+, 0) + (N - 2) \text{D}'s \} \quad (7)
\]

where \( \psi_{\text{BEC}} \) is the Bose-Einstein condensate ground state (a coherent quantum state) with \( N \) deuterons and \( \psi^* \) are final excited continuum states. \(^4\text{He}\) in equation (6) represents the ground state with spin-parity, \( 0^+ \), while \(^4\text{He}^*\) in equation (7) represents the \( 0^+ \) excited state at \( 20.21 \text{ MeV} \) with the resonance width of \( \Gamma(T + p) = 0.5 \text{ MeV} \). It is assumed that excess energy \((Q \text{ value})\) is absorbed by the BEC state and shared by \((N - 2)\) deuterons and reaction products in the final state. It is important to note that reaction \{6\}, described by equation (6), cannot occur in free space due to the momentum conservation, while reaction \{7\} described by equation (7) can occur with \( Q = 0 \) in free space without violating the momentum conservation, due to the resonance width of \( \Gamma(T + p) = 0.5 \text{ MeV} \) for the \(^4\text{He}\) ground state [41]. A detailed description of reaction \{6\} is given in previous publications [24,25]. In the following, detailed description and discussion of reaction \{7\} are given.

The reaction \{7\}, described by equation (7), can proceed via a sub-threshold resonance reaction [42–47]. The cross section for the sub-threshold resonance reaction is given by Breit-Wigner expression:

\[
\sigma(E) = \pi \tilde{\lambda}^2 w \frac{\Gamma_f(E)\Gamma_f}{(E - E_R)^2 + (\Gamma_f/2)^2} \quad (8)
\]

where \( \tilde{\lambda} = \lambda/2\pi \), \( \lambda = \hbar/mv \) (de Broglie wavelength), \( w \) is a statistical factor, \( E_R \) is the sub-threshold resonance energy, \( \Gamma_f \) is a partial decay width and \( E \) is the total decay width to the final states. If \( E \) is measured from the threshold energy \( E = 0 \) of \((\text{D} + \text{D})\) state, \( E_R = (20.21 \text{ MeV} - 23.85 \text{ MeV}) = -3.64 \text{ MeV} \).
4.3 Determination of S-factor and reaction rates

After combining equation (1) with equation (8), the $S(E)$ factor can be written as [42,47]:

$$S(E) = E \exp \left( \frac{2\pi n}{\pi^2} \right) \frac{\Gamma_f(E) \Gamma_i}{(E - E_R)^2 + (\Gamma/2)^2}$$  \hspace{1cm} (9)

The $S(E)$ factor near zero energy for the $\ell = 0$ state can then be written as (see Appendix for detailed derivation):

$$S(E) = \frac{\pi h^4}{\mu Z^2} \frac{1}{K_1^2(x)} w_{\theta}^2 F_{\text{BW}}(E),$$ \hspace{1cm} (10)

with

$$F_{\text{BW}}(E) = \frac{\Gamma_f}{(E - E_R)^2 + (\Gamma/2)^2}$$ \hspace{1cm} (11)

where $\mu$ is the reduced mass in units of atomic mass unit (AMU) (931.494 MeV), $R_a$ is the nuclear radius, and $K_1(x)$ is the modified Bessel function of order unity with argument $x = (8Z_1 Z_2 e^2 R_a \mu / \hbar^2)^{1/2}$. $K_1(x)$ is related to irregular Coulomb wave function $G_0(E, R_a)$ [48,49] (see Appendix). We note that $F_{\text{BW}}(E_R)$ is a maximum at $E = E_R = -3.64$ MeV. At $E = 0$, $F_{\text{BW}}(0)$ is reduced to $F_{\text{BW}}(0) = 0.47 \times 10^{-2} F_{\text{BW}}(E_R)$. Equation (10) shows that the $S(E)$ factor has a finite value at $E = 0$ and drops off rapidly with increasing energy. $\theta_f^2$ is the reduced width of a nuclear state [44], representing the probability of finding the excited state in the configuration $i$, and the sum of $\theta_f^2$ over $i$ is normalized to 1. $\theta_i$ is the overlap integral between the initial and final nuclear state components, $(\psi_{\text{final}} | \psi_{\text{initial}})$. The dimensionless number $\theta_f^2$ is generally determined experimentally and contains the nuclear structure information.

Equations (10) and (11) were used extensively in analysis of $(p,\gamma)$ reactions involved in nucleosynthesis processes in astrophysics [17,43,45-47].

Once $S(E)$ is calculated using equations (10) and (11), the reaction rates can be calculated from equations (4) and (5), using the calculated values of $S(E)$.

4.4 Reaction rates for anomalous production of tritium

For the entrance channel $\{7\}$, $D + D \rightarrow ^4\text{He}^*(0^+, 0, 23.85 \text{ MeV}, Q = 0)$, there are two possible decay channels as shown in Figure 1:

- $\{7\a\} ^4\text{He}^*(0^+, 0) \rightarrow T(1.01 \text{ MeV}) + p(3.02 \text{ MeV})$
- $\{7\b\} ^4\text{He}^*(0^+, 0) \rightarrow ^4\text{He}(0^+, 0, 0.0 \text{ MeV})$ (ground state).

$S(E)$ factors are calculated from equation (10) using $E = 0$ at a tail of the $^4\text{He}^*(0^+, 0)$ resonance at 20.21 MeV. $E = 0$ corresponds to 23.85 MeV above $^4\text{He}(0^+, 0)$ ground state. The calculated $S(E)$ can be used in equations (3) and (4) to obtain the total fusion reaction rate. We will estimate $S(E)$ factors for the decay channels, $\{7\a\}$ and $\{7\b\}$, using equation (10) in the following.

For the decay channel $\{7\a\}$, $\Gamma_f = \Gamma(\{7\a\}) = 0.5 \text{ MeV}$ [41]. When this value of $\Gamma_f$ is combined with other appropriate inputs in equation (10), the extracted $S$-factor for the decay channel $\{7\a\}$ is $S(\{7\a\}) \approx 1.4 \times 10^2 \theta(7)^2 \text{ keV-barn}$ for $E \approx 0$. In reference [24], it was shown that the neutron production rate $R(5)$ is suppressed, i.e. $R(5) \ll R(6)$ due to a selection rule. Since $^4(\text{He} + n)$ state has a resonance width of $\Gamma_f(\text{He} + n) = 0$ [41], this value of $S(\{7\a\})$ may provide an explanation of the reported branching ratio of $R(T)/R(n) \approx 10^7-10^9$ [10-16] or $R(n)/R(T) \approx 10^{-7}-10^{-9}$, as shown below.

For the decay channel $\{7\b\}$ ($0^+ \rightarrow 0^+$ transition), $\gamma$-ray transition is forbidden. However, the transition can proceed via the internal $e^+e^-$ pair conversion. The transition rate for the internal electron pair conversion is given by:

$$\omega = \frac{1}{135\pi} \left( \frac{e^2}{\hbar c} \right)^2 \frac{\gamma^5}{\hbar^5 c^2} R_\phi^2,$$

where $\gamma$ is the transition energy, $R_\phi$ is the nuclear radius, and $\phi = (\psi_{\text{exc}} | \psi_{g.s.})$, which is the overlap integral between the initial and final nuclear state components. Equation (12) was derived by Oppenheimer and Schwinger [50] in 1939 for their theoretical investigation of $0^+ \rightarrow 0^+$ transition in $^{16}\text{O}$. The rate for the internal electron conversion is much smaller by many order of magnitude.

For our case of $0^+ \rightarrow 0^+$ transition $\{7\b\}$, we obtain $\omega \approx 1.75 \times 10^{13} \text{s}^{-1}$, and $\Gamma_f = \hbar \omega \approx 1.15 \times 10^{-2} \text{ eV using appropriate inputs in equation (12). Using } \Gamma_f = \Gamma(\{7\b\}) = 1.15 \times 10^{-2} \text{ eV in equation (11), the extracted S-factor for decay channel } \{7\b\} \text{ is } S(\{7\b\}) \approx 3.3 \times 10^{-6} \theta(7)^2 [\phi(7b)]^2 \text{ keV-barn for } E \approx 0, \text{ yielding a branching ratio, } R(\{7\b\})/R(\{7a\}) \approx S(\{7\b\})/S(\{7\a\}) \approx 2.4 \times 10^{-8} \theta(7)^2 \text{. Experiments are needed for testing this predicted branching ratio.}$

5 Theoretical interpretation of reaction rates for neutron production

Experimental observation of $R(n)/R(T) \approx 10^{-7}-10^{-9}$ [10-16] is anomalous since we expect $R(n)/R(T) \approx 1$ from "hot" fusion reactions, $\{1\}$ and $\{2\}$. In this section, we explore nuclear reactions producing neutrons at anomalously low rates.

There are four possible processes which can produce neutrons. The first process is the secondary "hot" fusion reaction $\{2\}$ producing 2.45 MeV neutrons. The rate for this secondary reaction is extremely small, $R(\{2\})/R(\{6\}) = R(n)/R(4\text{He}) < 10^{-11}$, as shown previously [25].

The second process is 3D BECNF reaction. In reference [24], it was shown that both reaction $\{4\}$ and $\{5\}$ are suppressed due to a selection rule [24]. It was also suggested that the following 3D BECNF is possible:

$$\{8\} D + D + D \text{ (in BEC state)} \rightarrow n + p + ^4\text{He} + 21.6 \text{ MeV}.$$

This reaction is a secondary effect since the probability for $\{8\}$ is expected to be much smaller than 2D BECNF.
reaction \([6]\). Furthermore, it is suppressed further due to the selection rule described in \([24]\).

The third process is a "hot" fusion reaction \(D(t, n)^3\)He:

\[
\{9a\} \quad T + D \rightarrow n + ^4\text{He}, \quad Q = 17.59 \text{ MeV}
\]

induced by 1.01 MeV T produced from reaction \(\{7a\}\). Since the cross-section for reaction \(\{9a\}\) is large and a maximum (several barns) at \(E_D \approx 100 \text{ keV}\) \([51]\), neutrons from this process may contribute substantially to the branching ratio \(R(n)/R(T) = 10^{-7} - 10^{-9}\), as discussed in the next section.

Since Mosier-Boss et al. \([4]\) used 0.3 M PdCl\(_2\) and 0.3 M LiCl in D\(_2\)O in their experiment \([4]\), there is a possibility that a fourth process \(\{9b\}\), \(^7\)Li\((t, n)^9\)Be, may be involved in generating energetic neutrons:

\[
\{9b\} \quad T + ^7\text{Li} \rightarrow n + ^9\text{Be}, \quad Q = 10.439 \text{ MeV}.
\]

The neutron production rate for \(\{9b\}\) is expected to be much smaller than that for \(\{9a\}\). Energetic neutrons from the third process \(\{9a\}\) and the fourth process \(\{9b\}\) described above could induce the following reactions:

\[
\{10\} \quad ^{12}\text{C}(n, n')^4\text{He}, \quad ^{12}\text{C}(n, n')^8\text{Be} (^8\text{Be} \rightarrow ^2\text{He}),
\]

as reported recently by Mosier-Boss et al. \([1–4]\).

5.1 Branching ratios between neutron and tritium productions

The probability \(P(E_i)\) for a triton to undergo the conventional hot-fusion reaction \(\{9a\}\) while slowing down in the deuterated palladium metal can be written as \([52]\):

\[
P(E_i) = 1 - \exp \left( \int_0^{E_i} dE \sigma(E) \sigma(E_{TD}) \right) \approx \int_0^{E_i} dE \sigma(E) \sigma(E_{TD})
\]

\[
= n_D \int_0^{E_i} dE \frac{1}{E_{TD}} \sigma(E_{TD}). \quad (13)
\]

Quantities \(E_T\) and \(E_{TD}\) are the triton kinetic energies in the LAB and CM frames respectively. The stopping power for triton in PdD for \(E_T \leq 3 \text{ MeV}\) can be obtained from the following formula \([53]\).

The stopping power for a proton by the target atom \(j\) with the density \(n_j^\prime\) is taken from reference \([53]\). For a proton laboratory kinetic energy of \(E \leq 10 \text{ keV}\), it is given by:

\[
\frac{dE}{dx} = n_j^\prime A_1 E^{1/2} \times 10^{-18} \text{ keV cm}^2. \quad (14)
\]

For \(10 \text{ keV} \leq E \leq 1 \text{ MeV}\), it is given by

\[
\left[ \frac{dE}{dx} \right]^{-1} = \left[ \frac{dE}{dx} \right]_{\text{slow}}^{-1} + \left[ \frac{dE}{dx} \right]_{\text{high}}^{-1}, \quad (15)
\]

where

\[
\left[ \frac{dE}{dx} \right]_{\text{slow}} = n_j^\prime A_2 E^{0.45} \times 10^{-18} \text{ keV cm}^2, \quad (16)
\]

\[
\left[ \frac{dE}{dx} \right]_{\text{high}} = n_j^\prime A_3 E^{1.50} \times 10^{-18} \text{ keV cm}^2.
\]

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{Element (z)} & \text{H (1)} & \text{O (8)} & \text{Ti (22)} & \text{Pd (46)} \\
\hline
A_1 & 1.262 & 2.652 & 4.862 & 5.238 \\
A_2 & 1.44 & 3 & 5.496 & 5.9 \\
A_3 & 242.6 & 1920 & 5165 & 1.038 \times 10^4 \\
A_4 & 1.2 \times 10^4 & 2000 & 568.5 & 630 \\
A_5 & 0.1159 & 0.0223 & 0.009474 & 0.004758 \\
\hline
\end{array}
\]

and

\[
\left[ \frac{dE}{dx} \right]_{\text{high}} = n_j^\prime (A_3/E) \ln [(A_4/E) + (A_5/E)] \times 10^{-18} \text{ keV cm}^2. \quad (17)
\]

The coefficients \(A_1\) through \(A_5\) are given in Table 1 above \([53]\).

For the case of triton with laboratory kinetic energy of \(E \leq 3 \text{ MeV}\), \(E\) in equations \((14)–(17)\) is to be replaced by \(E/3\).

For the case of PdD, the target densities are assumed to be \(n_D = 6.8 \times 10^{22} \text{ cm}^{-3}\). For the incident triton with a kinetic energy \((\text{lab})\) of \(E_i = 1.01 \text{ MeV}\), equation \((13)\) yields \(P(E_i) = 0.31 \times 10^{-4}\), and hence the branching ratio of \(R(n)/R(T) = 0.31 \times 10^{-4}\) in PdD.

For the case of D\(_2\)O, with \(n_O = 3.3 \times 10^{22} \text{ cm}^{-3}\) and \(n_D = 6.6 \times 10^{22} \text{ cm}^{-3}\), we obtained the calculated value of \(P(E_i) = 0.94 \times 10^{-4}\) and the branching ratio \(R(n)/R(T) = 0.94 \times 10^{-4}\).

From the above calculated results with PdD, D\(_2\)O, and TiD, we have \(R(n)/R(T) < 10^{-4}\). The calculated results of the S-factors for different exit reaction channels are summarized in Table 2. The calculated branching ratio, \(R(n)/R(T)\), is also shown in Table 2.

5.2 Range of tritons

The range \(\Delta x(E_i)\) of triton in PdD can be calculated as

\[
\Delta x(E_i) = \int dx = \int_0^{E_i} \left( \frac{dE}{dx} \right)^{-1} dE. \quad (18)
\]

For the case of PdD with \(E_i = 1.01 \text{ MeV}\), the calculated value of the range using equation \((18)\) is \(\Delta x(E_i) = 0.42 \times 10^{-3} \text{ cm}\), while \(\Delta x(E_i) = 1.4 \times 10^{-3} \text{ cm}\) for the case of D\(_2\)O. For the case of TiD, \(\Delta x(E_i) = 0.64 \times 10^{-3} \text{ cm}\).

Because of different ranges of 1.01 MeV triton in different surrounding media (PdD, D\(_2\)O, and TiD) with different extents of media, the branching ratios of \(R(n)/R(T) = 0.31 \times 10^{-4}, 0.94 \times 10^{-4}\), and \(0.37 \times 10^{-4}\) in PdD, D\(_2\)O, and TiD as calculated in Section 5.1 are upper limits since the branching ratio \(R(n)/R(T)\) depends on deuteron density and extent of deuterons surrounding 1.01 MeV triton produced from reaction \(\{7a\}\). Thus reported values of
To test the above theoretical interpretation, based on predictions, proposed experimental tests of theoretical reductions from reaction $^1$H$(n)/^2$H, as discussed in Section 5.1.

6 Proposed experimental tests of theoretical predictions

To test the above theoretical interpretation, based on the third process [9a], we need to measure/detect (i) $1.01$ MeV tritium production and $3.02$ MeV proton production from reaction [9a], (ii) Bremsstrahlung radiations from energetic electrons going through metal, (iii) $0.51$ MeV $\gamma$-rays from $e^+e^-$ annihilation, (iv) energetic electrons from $e^+e^-$ pair production, (v) $\gamma$-rays from secondary reactions (see below).

For (i) detection of $1.01$ MeV tritons, it may be necessary to design new experimental set-ups capable of counting $1.01$ MeV tritons directly. Alternative method would be to use Ti/D systems as done in [10].

Tritium with a half life of $\sim 12.3$ years emits a low energy $\beta$ particle ($e^-$) with an end-point energy of $18.6$ KeV and an average energy of $5.7$ KeV. The range of $\beta$ with the maximum energy is as small as $9$ mg/cm$^2$ (in medium $Z$ materials). Tritium located only within this range can be detected by direct counting $\beta$ particle, which may be very inefficient. However, in the case of titanium samples, the energetic upper half of the $\beta$ spectrum is able to excite the characteristic Ko (4.5 KeV) and K$\beta$ (4.9 KeV) X-rays of titanium. This does not happen in Pd, since Pd has characteristic K X-rays with energies greater than $21$ KeV. Therefore, the use of Ti/D instead of Pd/D is proposed for future experimental tests.

For (iii), it could be accomplished by performing a coincidence measurement on the two $0.51$ gamma-rays that are emitted in positron annihilation. The use of two high purity germanium (HPGe) detectors in close to the electrodes might be efficient enough to detect the annihilation gamma-rays.

For (v) detection of $\gamma$-rays from secondary reactions, we may consider the following reactions:

Table 2. S-factors for Reactions in BECNF $\theta^2 (or \phi^2) = |(\psi_f|\psi_i)|^2$.

<table>
<thead>
<tr>
<th>Reactions</th>
<th>Reaction types/Products</th>
<th>S-factor (keV-barn)</th>
</tr>
</thead>
<tbody>
<tr>
<td>${7a}^4$He$^*(0^+, \text{STR}, 23.85 \text{ MeV}) \rightarrow T(1.01 \text{ MeV}) + p (3.02 \text{ MeV})$ $Q = 4.03 \text{ MeV}$</td>
<td>Sub-threshold $\gamma$-rays from secondary reactions,</td>
<td>$\approx 3.3 \times 10^{-7}$ $\theta^2(0^\circ)$</td>
</tr>
<tr>
<td>${7b}^4$He$^*(0^+, \text{STR}, 23.85 \text{ MeV}) \rightarrow ^4$He$(0^+, \text{g.s.}, 0.0 \text{ MeV}) + e^+e^- \text{pair}$ $Q = 23.85 \text{ MeV}$</td>
<td>Resonance reaction/</td>
<td>$\approx 3.3 \times 10^{-6}$ $\theta^2(0^\circ)$ $\phi(7a)$</td>
</tr>
<tr>
<td>${11} \text{ T} + \text{D} \rightarrow$ $\text{Neutron, } ^4\text{He}$, $\gamma$-heat $Q = 17.59 \text{ MeV}$</td>
<td>Neutron, $^4$He, heat</td>
<td>$S(11)/S(7a) &lt; 10^{-4}$</td>
</tr>
</tbody>
</table>

$R(n)/R(T) \approx 10^{-7}$ and $10^{-9}$ from the gas-loading experiments [10–16] are consistent with the calculated upper limit, $R(n)/R(T) < 10^{-3}$, as discussed in Section 5.1.

7 Summary and conclusions

Based on both the recently developed theory of Bose-Einstein condensation mechanism [24,25] for the entrance reaction channel, D + D reaction, in metal and the nuclear theory of the sub-threshold resonance reaction mechanism for the exit reaction channel, the reaction rate for $1.01$ MeV T from the exit reaction channel [7a] is calculated. The neutron production rate is also calculated as a secondary reaction, D(t, n)$^4$He, using the conventional nuclear theory. An upper limit of the branching ratio, $R(n)/R(T)$, between the neutron and triton reaction rates is estimated to be $R(n)/R(T) < 10^{-4}$. A set of experiments for testing theoretical predictions is proposed, in order to confirm the theoretical predictions and/or improve theoretical descriptions.

Appendix

In this Appendix, a detailed derivation is given for extraction of S-factor for the sub-threshold resonance reaction. The original derivation and results are given in references [42] and [47]. The cross section for reaction through compound nucleus resonance state is given by:

$$\sigma(E) = \frac{\pi \lambda^2 w}{E - E_0} \frac{I_i(E)I_f}{(E - E_0)^2 + (\Gamma/2)^2}$$ (A.1)

where $\lambda = \sqrt{2h\lambda}$ and $\lambda$ is de Broglie wave-length, $\lambda = h/mv$, and $w$ is the statistical factor given by:

$$w = \frac{2J + 1}{(2J_i + 1)(2J_f + 1)} (1 + \delta_{ij}).$$ (A.2)
To extract $S(E)$ from (A.1), we use $S(E)$ defined by the following definition of $S(E)$ [17]:

$$
\sigma(E) = \frac{S(E)}{E} e^{-2\pi \eta}
$$

(A.3)

with $\eta = Z_1 Z_2 e^2 / \hbar v$.

Equating (A.1) and (A.3), we obtain:

$$
S(E) = E e^{2\pi \eta} \pi^2 w \frac{\Gamma_i(E) \Gamma_f}{(E - E_R)^2 + (\Gamma/2)^2}
$$

(A.4)

with

$$
\Gamma_i,0(E) = \frac{2\hbar}{R_n} \left( \frac{2E}{\mu} \right)^{1/2} P_i(E, R_n) \theta_i^2
$$

(A.5)

where $\mu$ is the reduced mass.

The penetration factor $P_i(E, R_n)$ in equation (A.5) is given by:

$$
P_i(E, R_n) = \frac{1}{F_i^2(E, R_n) + G_i^2(E, R_n)}
$$

(A.6)

where $R_n$ is the nuclear radius, and $F_i$ and $G_i$ are regular and irregular Coulomb wave functions [48].

For the $s$-wave ($\ell = 0$) formation of the compound nucleus at energies $E$ near zero, we have $F_0(E, R_n) \approx 0$ and

$$
G_0(E, R_n) \approx 2 e^{\pi \eta} \left( \frac{\mu}{\rho} \right)^{1/2} K_1(x),
$$

(A.7)

where $x = 2\sqrt{2\pi \rho}$, $\rho = \sqrt{2\mu E R_n / \hbar}$, and $K_1(x)$ is the modified Bessel function of order one [49]. The argument $x$ is given by $x = (8Z_1 Z_2 e^2 R_n / \hbar^2)^{1/2} = 0.525 (\mu Z_1 Z_2 R_n)^{1/2}$, and $\mu$ is the reduced mass in units of atomic mass unit (931.494 MeV).

The penetration factor for $\ell = 0$, $P_0(E, R_n)$, is then given by:

$$
P_0(E, R_n) \approx \frac{1}{G_0^2(E, R_n)} = \frac{\pi}{4\rho K_1^2(x)} e^{-2\pi \eta}
$$

(A.8)

and the compound nucleus formation width, $\Gamma_{i,0}(E)$, is

$$
\Gamma_{i,0}(E) = \frac{\pi h^2}{2\mu R_n^2} \frac{1}{K_1^2(x)} \theta_i^2 e^{-2\pi \eta}
$$

(A.9)

where $\theta_i^2$ is called the reduced width of a nuclear state, which is generally determined experimentally and contains the nuclear structure information. $\theta_i^2$ is dimensionless and $\theta_i^2 \leq 1$ (called Wigner limit) [42]. $\theta_i^2$ is normalized as $\sum \theta_i^2 = 1$.

The $S(E)$ factor, equation (A.4), near zero energies can now be written as [47]:

$$
S(E) = \frac{\pi^2 h^4}{4\mu^2 R_n^2} \frac{1}{K_1^2(x)} w \theta_i^2 F_{BW}(E)
$$

(A.10)

where

$$
F_{BW}(E) = \Gamma_f \frac{E - E_R}{(E - E_R)^2 + (\Gamma/2)^2}.
$$

(A.11)

References

Y.E. Kim: Theoretical interpretation of anomalous tritium and neutron productions...


23. A. Kitamura et al., Phys. Lett. A 373, 3109 (2009), and references therein

24. Y.E. Kim, Naturwissenschaften 96, 803 (2009), and references therein


30. Q.M. Barer, Diffusion in and through Solids (Cambridge University Press, New York, 1941)


40. N. Bogoliubov, J. Phys. 11, 23 (1966)


44. R.G. Thomas, Phys. Rev. 81, 148 (1951); R.G. Thomas, Phys. Rev. 88, 1109 (1952)


47. C.E. Rolfs, W.S. Rodney, Cauldrons in the Cosmos: Nuclear Astrophysics (University of Chicago Press, 1988), Chap. 4


50. J.R. Oppenheimer, J.S. Schwinger, Phys. Rev. 56, 1066 (1939)

