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Measurements of ^{3}He accumulation effect on muon catalyzed fusion in the solid/liquid DT mixtures

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Abstract

An effect on the muon catalyzed fusion (μCF) of the ^{3}He originating from the tritium decay was studied by measuring the time-dependent change of the fusion-neutron disappearance rate (λ_n) in the deuterium and tritium (DT) mixtures with various tritium concentrations, $C_t = 0.1, 0.2, \ldots, 0.7$. A clear difference between the solid and the liquid DT mixtures due to the ^{3}He accumulation effect was observed: in solid λ_n increased with the time after solidification, whereas in liquid λ_n did not change. This indicates that ^{3}He produced in the solid DT mixtures is trapped. Admitting that all the ^{3}He remain in solid, the muon transfer rate from tμ to ^{3}He is determined to be about 4×10^9 s^{-1}, consistent with the theoretical prediction.

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The effect of helium impurities on muon catalyzed fusion (μCF) in a deuterium and tritium (DT) mixture is an inevitable problem due to the tritium β-decay. The ^{3}He originating from tritium β-decay accumulates with time at the rate of $155 \times C_t$ ppm per day, where C_t denotes the tritium concentration. In addition to this, ^{4}He as a byproduct of μCF also accumulates, although the amount of ^{4}He produced in the present scale of μCF experiment is negligible small in comparison with the ^{3}He from tritium β-decay. Thus, for accurate understanding of μCF phenomenon, it is necessary to study the effect of helium impurity.

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On the other hand, the μ^- transfer phenomenon from hydrogen isotopes (p, d and t) to helium is of special interest for the following reasons: this transfer reaction is one of the most fundamental muon transfer reactions, and it is known to be anomalously fast compared to the rate expected for the conventional direct exchange process, suggesting a transfer through formation of the muonic molecule, such as $d^4He\mu$. As described in the following paragraph, this transfer reaction gives us an opportunity to learn experimentally about the structure of a muonic molecule by X-ray spectroscopy.

Historically, the formation of a muonic molecule like $d^4He\mu$ has been proposed to explain the anomalously high μ^- transfer rates from hydrogen isotopes to helium [1,2]. According to the proposed model, the following process is expected to take place: instead of a direct exchange reaction of $d\mu + (^4He\,e^-e^-) \rightarrow [(^4He\mu)e^-] + d + e^-$, the molecular ion is formed through $d\mu + (^4He\,e^-e^-) \rightarrow [(d^4He\mu)e^-]^+ + e^-$, where $d^4He\mu$ is preferentially formed in the bound state of $2p\sigma$, then deexcites to its unbound ground state, i.e. $^4He\mu + d$ system. In this case, the characteristic photon spectrum from the bound-free transition was predicted; a unique peak energy with a broad and asymmetric shape [1–4]. At UT-MSL/KEK, such an experiment was first carried out for a liquid D_2 target with helium impurity (430 ppm) [3], and the characteristic broad and asymmetric photon peak was observed with a central energy of 6.85 ± 0.04 keV and a width of 0.74 ± 0.04 keV (FWHM), agreeing quite well with the theoretical prediction. The result confirmed the model of the muon transfer mechanism through the formation of a muonic molecule state.

In the present work, the systematic study of the ^{3}He accumulation effect in DT μCF was conducted at the RIKEN-RAL Muon Facility [5] constructed at ISIS facility of Rutherford Appleton Laboratory (UK). There, an advanced tritium handling system (THS) with ^{3}He removal capability (palladium filter) as well as gas analysis capability (gas-chromatography) was installed [6]. Using the THS, one can prepare a ^{3}He-free, i.e. pure, DT mixture target in liquid or solid phase within 0.5 h after ^{3}He removal.

The DT target was prepared by the following procedure [6]. The deuterium and tritium gases were mixed with an intended tritium concentration in a gas storage of the THS. The mixed gas was transfered to a constant-volume gas buffer through the palladium filter, by which the DT gas was purified and also the chemical equilibrium was attained. Subsequently, the purified gas was introduced into a 2-cm^3 target cell followed by either solidification or liquefaction by adjusting the temperature of the cell to about 16 K or 20 K, respectively. In a separate off-beam experiment, by the gas-chromatography analysis, we confirmed that ^{3}He concentration was below 1 ppm right after the purification procedure.

The experiment was carried out by using a 54.5-MeV/c backward-decay μ^- beam with a double pulse structure (each 70-ns width with a 320-ns separation and 50-Hz repetition rate), which was introduced in the solid or liquid DT mixture target placed at the center of 2.4-T beam-focusing magnetic field generated by a superconducting Helmholtz coil pair. Two neutron detectors composed of NE213 liquid scintillator (2'' in diameter and 2'' in length) were placed at 82 cm from the target along the μ^- beam axis and just behind 5-cm thick lead absorber. The detection efficiency of each neutron detector was calibrated by a 14-MeV standard neutron source at ETL (Tsukuba, Japan) [7]. Although this article is based upon the fusion neutron detection, we also conducted combined X-ray detection [8]. For detection of X-rays associated with μCF phenomenon against a huge bremsstrahlung background from tritium β-decay, utilization of an intense pulsed muon beam drastically improved the signal to noise ratio by synchronized detection with the muon beam [5].

The total neutron yield (Y_n) was determined from the number of measured neutrons by taking into account pileup and multi-hit correction, a solid angle of the detector, neutron transmission efficiency and detection efficiency. The neutron disappearance rate (λ_n) was obtained from the neutron emission time spectrum (Fig. 1). The fusion neutron yield at time t after muon beam injection and the neutron disappearance rate are associated with each other; $y_n(t) = \phi\lambda_c e^{-\lambda_n t}$, and λ_n is decomposed; $\lambda_n = \lambda_0 + \phi\lambda_c W + \lambda_{He}$. In these equations, ϕ is the density of the DT mixture normalized by the liquid hydrogen density, λ_0 the μ^- decay-rate (0.455×10^6 s^{-1}), λ_c the cycling rate of the DT μCF, W the total muon loss probability per cycle except for the effect of ^{3}He, and λ_{He} refers to the muon loss due to μ^- transfer

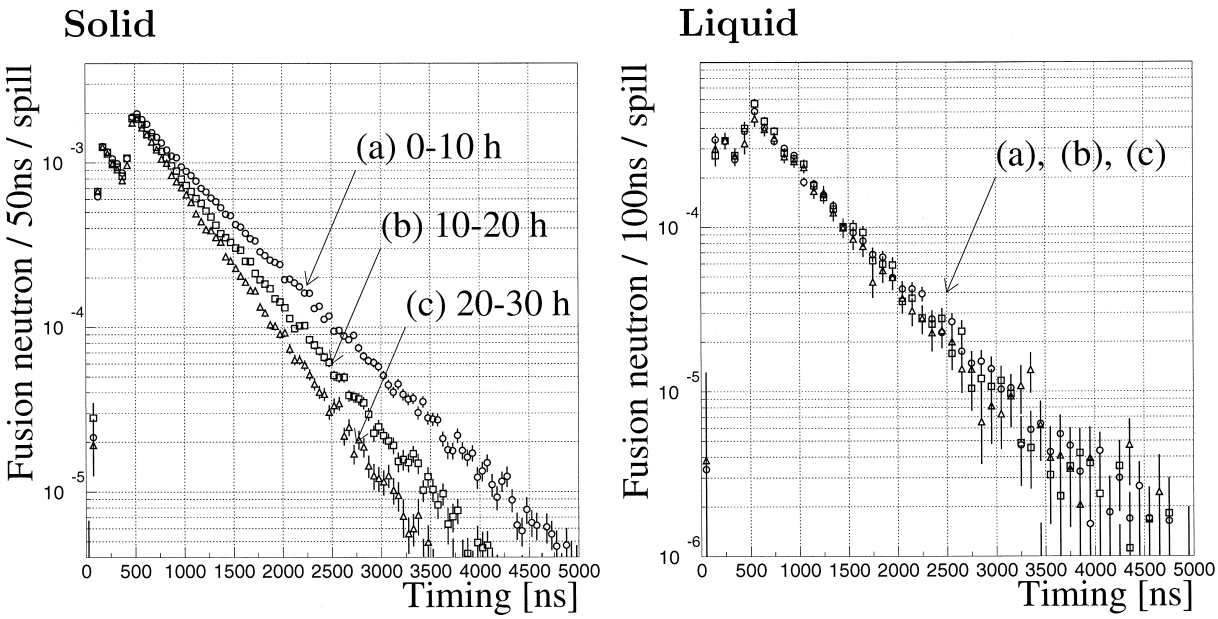

Fig. 1. Typical time spectra of fusion neutrons produced by μCF in the DT mixture; liquid and solid with $C_t = 0.7$ at (a) $\tau = 0$–10 h, (b) 10–20 h and (c) 20–30 h after condensation.

or capture to ^{3}He. The λ_c is associated with the total neutron yield and the neutron disappearance rate as: $Y_n = \int y_n(t)dt = \phi\lambda_c/\lambda_n$, and λ_c is independent of helium impurity. As shown in Fig. 1, it was observed that λ_n increased gradually (typical time-scale of hours) with the time after condensation only in solid, and not in liquid. As described later, this phenomenon is explained by the accumulation of ^{3}He originating from tritium β-decay. In general, the solubility of helium in condensed hydrogen is estimated by Henry's law. Assuming this law, the dissolved helium is expected not to change λ_n visibly in the present case, and it is inconsistent with the present work. However it is not appropriate to assume Henry's law in the present case, since Henry's law deals with the solubility of helium pressurizing on hydrogen, and not helium produced in hydrogen.

The features of the phenomenon which we pay attention to are summarized:

- Only in solid, the neutron disappearance rate increased with the time after solidification.
- By melting the solid, the neutron disappearance rate returned to the original value at the time right after solidification.

We confirmed these features by measurements in sequential purification, solidification and liquefaction. Our assumption that ^{3}He from tritium β-decay remains only in solid explains all these features.

In Fig. 2, the time-dependent changes of $\lambda_n(\tau)$ are summarized for both (a) in solid and (b) in liquid, where τ denotes the time after condensation. The increase of λ_n and $\lambda_n(\tau) - \lambda_n(0)$ are shown in Fig. 3. In addition to the above-mentioned features, the following feature is clearly seen in these figures:

- In solid, the slope of $\lambda_n(\tau) - \lambda_n(0)$ increases with an increase of C_t.

In the case of the solid $C_t = 0.7$, the increase of λ_n is saturated at $\tau \sim 30$ h, at which time the helium concentration is about 130 ppm. This phenomenon can be explained by nucleation of helium, i.e. several helium atoms gathering to make a cluster.

Now, let us consider the possible sublimation phenomena induced by the radiation of tritium β-decay in solid DT mixtures [9]. In a series of present experiments, the muon stopping number was found to increase by about 10 % within 5 hours after solidification, and to stay like a saturation afterward. This is expected be due to the sublimation effect. In

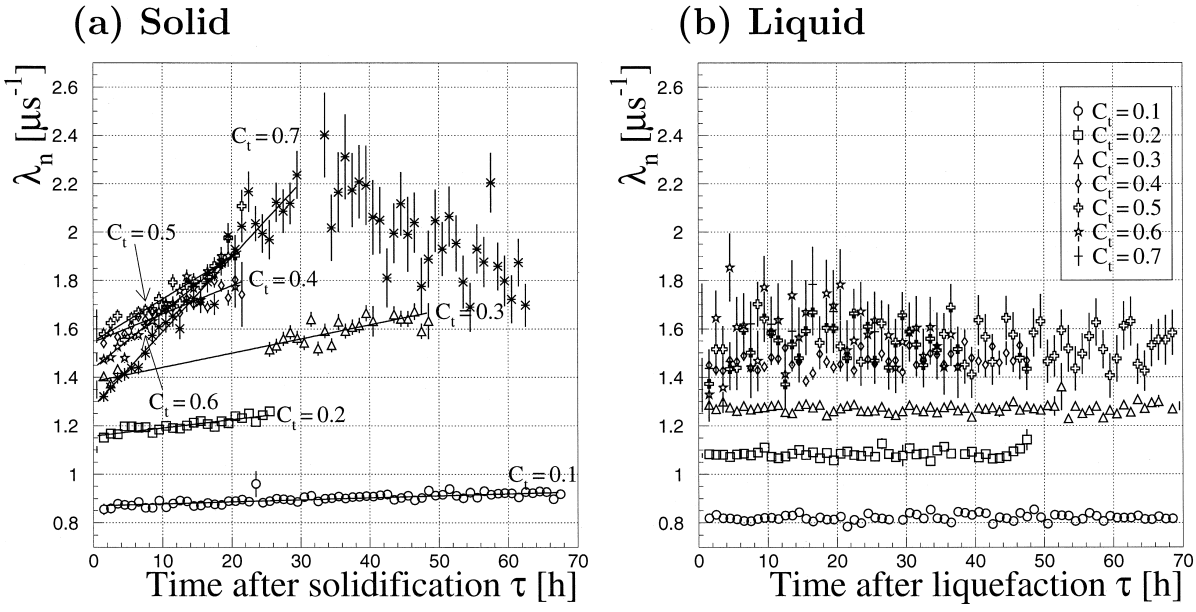

Fig. 2. The plot of $\lambda_n(\tau)$ as a function of the time after condensation; (a) in solid and (b) in liquid with various C_t.

the target cell partially filled with solid DT mixture, hydrogen molecule sublimated from warmer interior surface preferentially solidifies at cooler outside area, leading to a change of shape. In the present experimental arrangement, the shape of the DT target may have more muon stopping. However it did not affect

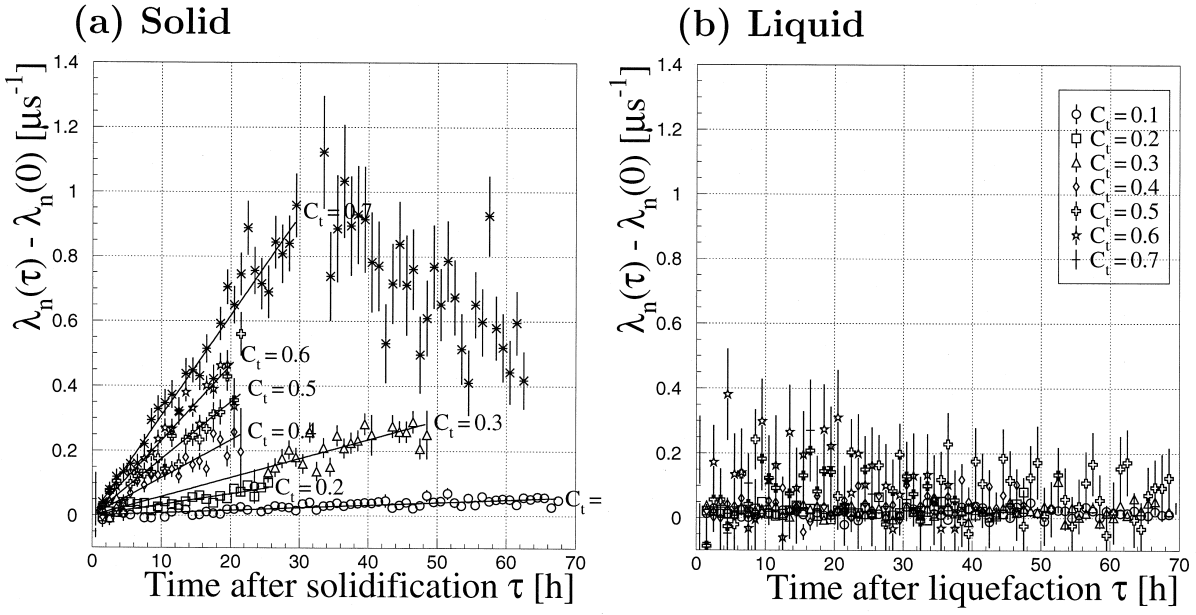

Fig. 3. The plot of the increase of λ_n, $\lambda_n(\tau) - \lambda_n(0)$; (a) in solid and (b) in liquid with various C_t.

the extracted basic parameter, such as λ_n, which increased linearly for more than a few tens of hours while the muon stopping number stayed constant.

Considering the usual cycling-reaction diagram in dt–μCF, we must take into account three possible routes for the μ^- to be captured by ^{3}He: (i) $\mu^- \rightarrow d\mu \rightarrow {}^3\mathrm{He}\mu$; (ii) $\mu^- \rightarrow t\mu \rightarrow {}^3\mathrm{He}\mu$ including $\mu^- \rightarrow d\mu \rightarrow t\mu \rightarrow {}^3\mathrm{He}\mu$; (iii) $\mu^- \rightarrow {}^3\mathrm{He}\mu$. Thus $\lambda_{\mathrm{He}}(\tau)$ in solid DT, which solely depends on the amount of accumulated ^{3}He, is decomposed as:

$$\lambda_{\mathrm{He}}(\tau) = \lambda_n(\tau) - \lambda_n(0) = \phi\left(f_{d\mu}\lambda_{d\mathrm{He}\mu} + f_{t\mu}\lambda_{t\mathrm{He}\mu} + A\lambda_c\right)C_{\mathrm{He}}(\tau) \quad (1)$$

where C_{He} denotes the helium concentration, $f_{h\mu}$ $(h = d, t)$ is the fraction of the muon staying time in hμ, and $\lambda_{h\mathrm{He}\mu}$ is the transfer rate from $h\mu$ to ^{3}He through formation of $h^3\mathrm{He}\mu$ molecule. The third term in this equation, $\phi A\lambda_c C_{\mathrm{He}}$, stands for the muon loss process due to the atomic capture of a liberated free muon by a helium, where A is the ratio of the muon captured by helium to that by a hydrogen atom. Since the amount of ^{4}He originating from the μCF phenomenon itself is negligible in comparison with the ^{3}He from tritium β-decay, C_{He} is expressed with the tritium decay rate $\lambda_t(= 1.55 \times 10^{-4}\ /\mathrm{day})$; $C_{\mathrm{He}}(\tau) = (1 - e^{-\lambda_t \tau})C_t$, where we assume that all ^{3}He remain in solid. The typical experimental period is short enough to satisfy $\lambda_t\tau \ll 1$, and thus $C_{\mathrm{He}} = \lambda_t\tau C_t$ is a good approximation. Therefore, the linear correlation between λ_{He} and τ, which appears in Fig. 3, can be understood. In Fig. 4, the slope of λ_{He} divided by $\phi\lambda_t C_t$, i.e. $\left(f_{d\mu}\lambda_{d\mathrm{He}\mu} + f_{t\mu}\lambda_{t\mathrm{He}\mu} + A\lambda_c\right)$, is plotted against various C_t.

The atomic capture term in Eq. (1) is relatively small. The atomic capture ratio A is about 2 according to the Fermi-Teller law, and λ_c is at most 150 μs^{-1} [8], and thus the atomic capture term, $A\lambda_c$, is less than 10 % of the values shown in Fig. 4. Therefore muon transfer from hydrogen to helium contributes to λ_{He} mainly.

The fraction of the muon staying time in $h\mu$ $(h = d, t)$ is expressed by $f_{h\mu} = \tau_{h\mu}/(\tau_{d\mu} + \tau_{t\mu})$, where $\tau_{h\mu}$ is the muon staying time in the $h\mu$ atom. With the knowledge of the μCF cycle, $\tau_{d\mu}$ and $\tau_{t\mu}$ are obtained as:

$$\tau_{d\mu} = \frac{q_{1s}C_d}{\lambda_{dt}C_t}, \quad (2)$$

$$\tau_{t\mu} = \frac{3}{4}\frac{1}{\lambda^{10}_{t\mu}C_t + \lambda^{1}_{dt\mu}C_d} + \frac{1/4 + 3/4\cdot\chi}{\lambda^{0}_{dt\mu}C_d},$$

$$\chi = \frac{\lambda^{10}_{t\mu}C_t}{\lambda^{10}_{t\mu}C_t + \lambda^{1}_{dt\mu}C_d}, \quad (3)$$

where $C_d(= 1 - C_t)$ is the deuterium concentration; q_{1s} the probability of dμ arriving at $1s$ ground state before muon transfer to tμ; $\lambda_{dt}(= 280\ \mu s^{-1}\ [10])$

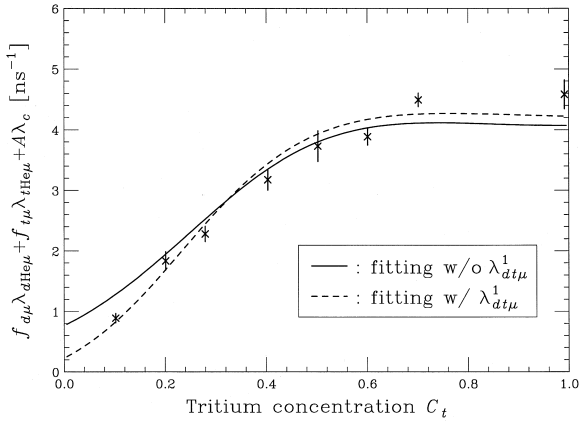

Fig. 4. The slope of $\lambda_{\mathrm{He}}(\tau)$ in solid DT divided by $\phi\lambda_t C_t$, i.e. $f_{d\mu}\lambda_{d\mathrm{He}\mu} + f_{t\mu}\lambda_{t\mathrm{He}\mu} + A\lambda_c$ is plotted against C_t. The solid line represents the fitting without the effect of the the molecular formation from $(t\mu)^{F=1}_{1s}$, and the dashed line is the fitting with it.

the ground-state transfer rate of muon, $(d\mu)_{1s} \rightarrow (t\mu)_{1s}$; $\lambda_{t\mu}^{10}(=1300\,\mu s^{-1}$ [11]) the hyperfine transition rate, $(t\mu)_{1s}^{F=1} \rightarrow (t\mu)_{1s}^{F=0}$; $\lambda_{dt\mu}^{F}$ $\left(\equiv \lambda_{dt\mu}^{F,D_2} C_d + \lambda_{dt\mu}^{F,DT} C_t\right)$ the dtμ molecule formation rate from each state of $(t\mu)_{1s}^{F}$ statistically averaged over collisions with D_2 and DT molecules; and χ is the branching ratio of the hyperfine transition to the molecular formation from $(t\mu)_{1s}^{F=1}$.

It was predicted that the dtμ molecule formation is dominant from $(t\mu)_{1s}^{F=0}$ below about 100 K [12], i.e. $\lambda_{dt\mu}^{1} \ll \lambda_{t\mu}^{10}, \lambda_{dt\mu}^{0}$, and thus $\tau_{t\mu}$ approximates to $\tau_{t\mu} \cong \frac{3}{4}(\lambda_{t\mu}^{10} C_t)^{-1} + (\lambda_{dt\mu}^{0} C_d)^{-1}$. The parameters determining $\tau_{d\mu}$ and $\tau_{t\mu}$, i.e. $q_{1s}, \lambda_{dt\mu}^{0}$, were determined by analyzing the C_t dependence of the muon cycling rate, $\lambda_c(=(\tau_{d\mu}+\tau_{t\mu})^{-1})$ [8]. In the case of $q_{1s}\left(=1/\left(1+a_q C_t\right)\right)$ with $a_q \cong 5$, $\lambda_{dt\mu}^{0,D_2} \cong 770$ μs^{-1}, $\lambda_{dt\mu}^{0,DT} \cong 0\ \mu s^{-1}$, the result of $\lambda_{tHe\mu} = (4.1 \pm 0.1) \times 10^9\ s^{-1}$ is obtained, while $\lambda_{dHe\mu}(\cong 230\ \mu s^{-1}$ [13]) is fixed. As shown in Fig. 4, the fitting result, which is represented by the solid line, qualitatively reproduces the experimental value, however small but systematic disagreement is seen especially at low and high C_t. The dashed line shows the fitting result to improve the disagreement by introducing the $\lambda_{dt\mu}^{1}$ term. The best fitting result is obtained in the case that $\lambda_{dt\mu}^{1}$ corresponds to about 50 % of $\lambda_{dt\mu}^{0}$, and $\lambda_{tHe\mu}$ is slightly modified to $(4.3 \pm 0.1) \times 10^9\ s^{-1}$. The improvement by the $\lambda_{dt\mu}^{1}$ term may imply that the thermalization of tμ is slow. Since tμ with the kinetic energy of the order of 10^{-1} eV has several resonant formation modes even in the $F=1$ state [12], $(t\mu)_{1s}^{F=1}$ may form the dtμ molecule easily if tμ spends enough time with the kinetic energy corresponding to the resonant energy.

The same analysis procedure is applied for the tt–μCF data, which is reported separately [14], and its result is consistent with the present result. The muon transfer rate from hydrogen to helium was theoretically calculated by Kravtsov et al. above ~ 50 K [15]. Applying their ''simple-approach approximation'', we extrapolate from their result to the temperature of the present work, ~ 16 K, to estimate $\lambda_{tHe\mu}$ as $\sim 7 \times 10^9\ s^{-1}$. The present result is in agreement with their theory to the same order of magnitude.

As already mentioned, the muonic molecule, like $d^3He\mu$, has a radiative decay branch by emitting a characteristic X-ray with a broad and asymmetric shape. Also in the present work and in the above-mentioned tt–μCF experiment, such X-rays were observed. The observed X-ray yield of radiative decay give the consistent result with the present argument based upon a full ^{3}He capture in solid DT mixture [14].

In the present work, we found that ^{3}He originating from tritium β-decay was trapped only in the solid DT mixture, which increased the neutron disappearance rate. The mechanism of ^{3}He trapping in solid and de-trapping in liquid is still an open question. According to the article [16], about 60 % of the T_2 decay product is $(^3HeT)^+$ ion, and the rest is mainly $^3He^+$. The evolution of these states may be different in liquid and solid. The article [17] indicated the possibility that the $(^3HeT)^+$ ion forms the cluster with surrounding hydrogen molecules. A recent calculation expects that $^3He^+$ ion is bound at the octahedron interstitial center of the solid hydrogen hcp structure [18]. However, these theory can partially explain the trapping mechanism. In addition to comprehensive trapping mechanism, the mechanism of de-trapping and diffusion in liquid is still an open question.

In the present work, we derived $\lambda_{tHe\mu}$ from the ^{3}He accumulation effect under an assumption that all the ^{3}He remain in solid. However, the fraction of ^{3}He remaining in solid should be determined more precisely. In such a sense, the ^{3}He evolution after tritium decay is important to solve this problem.

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