

Measurements of He accumulation effect on muon catalyzed fusion in the solid/liquid D-T mixtures

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An effect on the muon catalyzed fusion of the ^3He accumulated by the tritium decay was studied, by measuring time-dependent change of the fusion-neutron disappearance rate (λ_n) in the deuterium and tritium (D_{1-x}T_x) mixtures with various tritium concentration, $x = 0.1, 0.2, \dots, 0.7$. Clear difference between the solid and the liquid D-T mixture due to the ^3He accumulation effect was observed; in solid the λ_n increased time by time, besides in liquid the λ_n did not change. This suggests that ^3He created in the solid D-T mixtures is trapped in it.

The effect of helium impurities on muon catalyzed fusion (μCF) in a deuterium and tritium (D-T) mixture is an inevitable problem due to the following two processes: (1) ^3He originating from tritium β -decay accumulates time by time ($155 \times C_t$ ppm per day); (2) ^4He as a byproduct of μCF also accumulates although it is negligible in comparison with the above-mentioned process. Thus, for rigorous understanding of $\mu\text{-CF}$ phenomenon, it is necessary to study the effect of helium impurity.

On the other hand, the μ^- transfer phenomenon from hydrogen isotopes (p , d and t) to ^3He and ^4He is of special interest for the following reasons: this transfer reaction is known not only to be the most fundamental process but also to be anomalously fast compared to the conventional direct exchange process, suggesting a transfer through a formation of the muonic molecule, such as ($d^4\text{He}\mu$). 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 state like ($d^4\text{He}\mu$) has been proposed to explain the anomalously high μ^- transfer rate 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 ($d\mu$) + $^4\text{He} \rightarrow [({}^4\text{He}\mu)e^-] + d + e^-$, the molecular ion is formed through ($d\mu$) + $^4\text{He} \rightarrow [({}^4\text{He}\mu)e^-]^+ + e^-$, where ($d^4\text{He}\mu$) is preferentially formed at the bound state of $2p\sigma$, then deexcites to its unbound ground state, *i.e.* (${}^4\text{He}\mu + d$) system. In this case, the characteristic photon spectrum of the bound-free transition was predicted: an unique peak energy with a broad and an asymmetric shape.^{1,2)} 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, 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 ^3He accumulation effect in D-T μCF was conducted at the RIKEN-RAL Muon Facility⁴⁾ constructed at ISIS facility of Rutherford Appleton Laboratory. There, advanced tritium handling system (THS) with a ^3He removal capability (palladium filter) as well as a gas analysis capability (gas-chromatography) was installed.⁵⁾ Using the THS, one can prepare ^3He -free, *i.e.* pure, D-T mixture for the μCF experiment within 0.5 h after ^3He removal.

The D-T target was prepared by the following procedure.⁵⁾ The D-T mixture with an intended tritium concentration C_t (x) is made in a gas storage in the THS. The mixture gas was transferred to a constant-volume gas buffer, through the palladium filter, by which the D-T gas was purified and also the chemical equilibrium was attained. Subsequently, the purified gas is introduced into a 2-cm³ 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 ^3He concentration is below 1 ppm right after the removal procedure and that the chemical equilibrium is attained among D_2 , T_2 and DT .

The experiment was carried out by using 54.5-MeV/c backward-decay μ^- with a double pulse structure, which was introduced in the solid or liquid D-T mixture target placed at the center of 2.4-T confinement magnetic field of the su-

perconducting 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 and just behind 5-cm thick lead absorber. The detection efficiency of the neutron detectors was calibrated by 14-MeV standard neutron source at ETL.⁶⁾ The presently reporting experiment was mostly done by the fusion neutron detection, while the X-ray combined experiment is reported separately.⁷⁾

The number of measured neutrons was corrected for pileup and multi-hit effects, a solid angle of detector, neutron transmission efficiency and detection efficiency, to get total neutron yield (Y_n). The neutron disappearance rate (λ_n) can be obtained from the timing spectrum whose typical examples are shown in 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: $\lambda_n = \lambda_0 + \phi \lambda_c W_n + \lambda_{He}$, where ϕ is the density of the D-T mixture normalized by the liquid hydrogen density, λ_0 is the μ^- decay-rate ($0.455 \times 10^6 \text{ s}^{-1}$), λ_c is the cycling rate of the D-T μCF , W_n is the total muon loss probability per cycle except transfer to ^3He and λ_{He} is the rate of μ^- transfer or capture to ^3He . As shown in Fig. 1, we observed gradual, typical time-scale of hour, increase of the λ_n only in solid, and not in liquid. As described later, this phenomenon is explained by the accumulation of ^3He originating from tritium β -decay. In general, the solubility of helium in condensed hydrogen is estimated by Henry's law, and the contribution of the dissolved helium to the λ_{He} term is estimated to be negligible in the present case. However Henry's law is supposing the situation under pressurizing helium gas, and not that helium is created in hydrogen. Thus, supposing Henry's law may not be appropriate in this case.

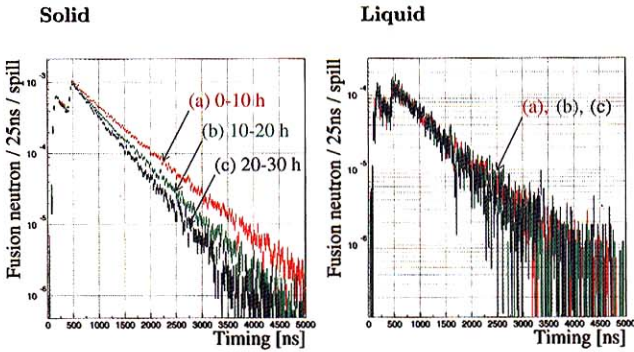


Fig. 1. Typical time spectra of fusion neutrons of muon catalyzed fusion in D-T mixture; liquid and solid with $C_t = 0.7$ at (a) $\tau = 0-10$ h, (b) $10-20$ h and (c) $20-30$ h after condensation.

The features of the phenomenon which we pay attention are summarized: (1) only in solid, the neutron disappearance rate increased time by time after condensation; (2) by melting solid, the neutron disappearance rate returned to the original value at the time right after solidification. We confirm these features by measurements in sequential purification, solidification and liquefaction. Our assumption that ^3He from tritium β -decay remains in solid did not conflict with all these features.

In Fig. 2, the data of the time-dependent change of $\lambda_n(\tau)$ are

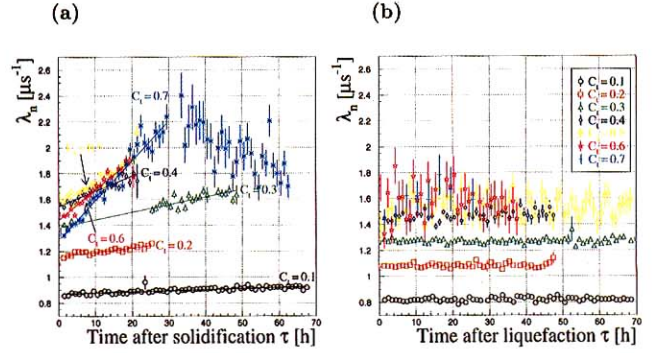


Fig. 2. The plot of $\lambda_n(\tau)$ in (a) solid and (b) liquid D-T mixture with various C_t .

summarized for both (a) in solid and (b) in liquid, where τ denotes the time after condensation. Also, the data presented as $\lambda_n(\tau) - \lambda_n(\tau = 0)$ are summarized in Fig. 3. From these data, the following conclusions can readily be drawn: (i) there is none of the significant ^3He accumulation effect in liquid D-T, while there is a visible effect in solid D-T; (ii) in solid D-T, the slope of λ_{He} increases with the C_t increasing. In the case of the solid $C_t = 0.7$, the increase of λ_n is suppressed at $\tau \sim 30$ h, at which the helium concentration is about 130 ppm. This phenomenon is explained by the nucleation of helium.

Considering usual cycling-reaction diagram in $dt-\mu\text{CF}$, we must consider possible three routes for the μ^- to be captured to ^3He : (i) $\mu^- \rightarrow (d\mu) \rightarrow (^3\text{He}\mu)$; (ii) $\mu^- \rightarrow (t\mu) \rightarrow (^3\text{He}\mu)$ including $\mu^- \rightarrow (d\mu) \rightarrow (t\mu) \rightarrow (^3\text{He}\mu)$; (iii) $\mu^- \rightarrow (^3\text{He}\mu)$. Thus the rate of $\lambda_{He}(\tau) \equiv \lambda_n(\tau) - \lambda_n(\tau = 0)$ in solid D-T can be decomposed as:

$$\lambda_{He}(\tau) = \phi (f_{d\mu} \lambda_{dHe\mu} + f_{t\mu} \lambda_{tHe\mu} + f_a \lambda_a) C_{He}(\tau), \quad (1)$$

where C_{He} denotes the helium concentration, $f_{d\mu}$, $f_{t\mu}$ and f_a are the fraction of the muon staying time in $(d\mu)$, $(t\mu)$ and free muon, and $\lambda_{dHe\mu}$, $\lambda_{tHe\mu}$ and λ_a are the transfer rate to ^3He through a formation of $(d^3\text{He}\mu)$, $(t^3\text{He}\mu)$ molecule and capture rate from free muon (atomic capture), respectively. Since the amount of ^4He originated from the μCF phenomenon itself was negligible in comparison with the ^3He from tritium β decay, C_{He} is expressed with the tritium decay rate $\lambda_t (= 1.55 \times 10^{-4} / \text{day})$: $C_{He}(\tau) = (1 - e^{-\lambda_t \tau}) C_t$. The experimental period was enough short to satisfy $\lambda_t \tau \ll 1$, and thus $C_{He} = \lambda_t \tau C_t$ is good approximation. Finally, the λ_{He}

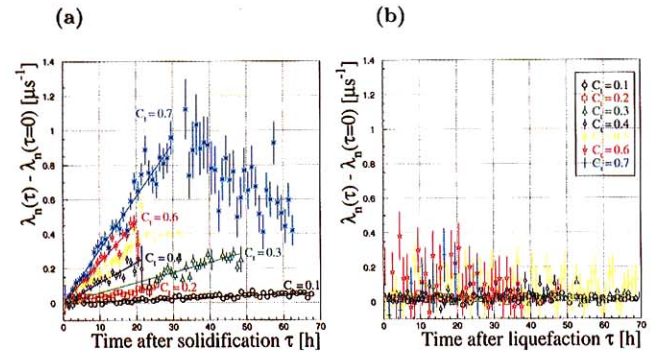


Fig. 3. The plot of $\lambda_n - \lambda_n(\tau = 0)$ in (a) solid and (b) liquid D-T mixture with various C_t .

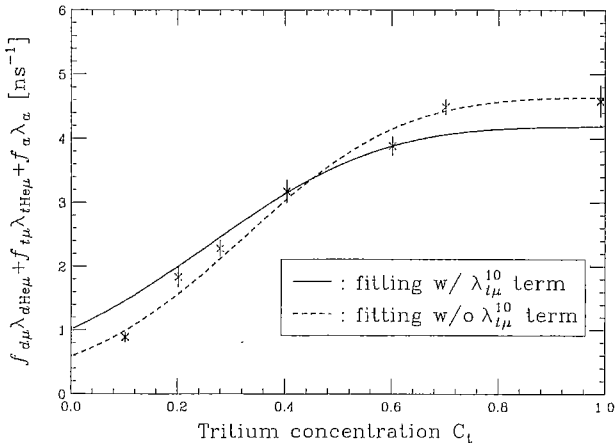


Fig. 4. The slope of $\lambda_{\text{He}}(\tau)$ in solid D-T divided by $\phi\lambda_t C_t$, i.e. $f_{d\mu}\lambda_{d\text{He}\mu} + f_{t\mu}\lambda_{t\text{He}\mu} + f_a\lambda_a$ is plotted against C_t . The solid line shows the fitting result with the $\lambda_{t\mu}^{10}$ term, and the dashed line is without the $\lambda_{t\mu}^{10}$ term.

is proportional to τ as shown in Fig. 3. In Fig. 4, the slope of λ_{He} divided by $\phi\lambda_t C_t$, i.e. $(f_{d\mu}\lambda_{d\text{He}\mu} + f_{t\mu}\lambda_{t\text{He}\mu} + f_a\lambda_a)$, is plotted against various C_t .

In Eq. (1), the last term can be relatively small; because (i) the atomic capture process (λ_a) is about 100 times faster than the other μCF processes (λ_{dt} etc. described later), so that f_a is 100 times smaller than $f_{d\mu}$ or $f_{t\mu}$, and (ii) the λ_a is 10 times faster than the $\lambda_{d^3\text{He}\mu}$ or $\lambda_{t^3\text{He}\mu}$, finally the last term is about 10 times smaller than the first two terms. Thus we neglect this term in the following discussion.

The fraction of the muon staying time in $(d\mu)$ and $(t\mu)$ are expressed by $f_{d\mu} = \frac{\tau_{d\mu}}{\tau_{d\mu} + \tau_{t\mu}}$ and $f_{t\mu} = \frac{\tau_{t\mu}}{\tau_{d\mu} + \tau_{t\mu}}$, where $\tau_{d\mu}$ and $\tau_{t\mu}$ are the muon staying time in each muonic atom. Assuming the $(dt\mu)$ formation rate being dominant from $F = 0$ state of $(t\mu)_{1s}$, according to the knowledge of the μCF cycle, $\tau_{d\mu}$ and $\tau_{t\mu}$ are obtained:

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

$$\tau_{t\mu} = \frac{3}{4} \frac{1}{C_t \lambda_{t\mu}^{10}} + \frac{1}{(C_d)^2 \lambda_{dt\mu-d}^0 + 2C_d C_t \lambda_{dt\mu-t}^0}, \quad (3)$$

where $C_d (= 1 - C_t)$ is the deuterium concentration, q_{1s} is the probability of $(d\mu)$ arriving at $1s$ ground state before transfer reactions, λ_{dt} is the transfer rate from $(d\mu)$ to t , $\lambda_{t\mu}^{10}$ ($= 1300 \mu\text{s}^{-1}$ ⁸⁾) is the hyperfine transition rates from $F = 1$ state to $F = 0$ state in $(t\mu)_{1s}$ and $\lambda_{dt\mu-d}^0$ and $\lambda_{dt\mu-t}^0$ are the $(dt\mu)$ formation rate through $(t\mu)_{1s}^{F=0} + D_2$ and $(t\mu)_{1s}^{F=0} + \text{DT}$, respectively. The parameters determining $\tau_{d\mu}$ and $\tau_{t\mu}$, i.e. q_{1s} , λ_{dt} , $\lambda_{dt\mu-d}^0$ and $\lambda_{dt\mu-t}^0$, was determined by analyzing the C_t dependence of the muon cycling rate, $\lambda_c (= (\tau_{d\mu} + \tau_{t\mu})^{-1})$.⁷⁾ The λ_c is associated with the total neutron yield and the neutron disappearance rate, $Y_n(\tau) = \int y_n(t, \tau) dt = \phi \lambda_c / \lambda_n(\tau)$, and is confirmed to be independent of C_{He} .

Using $q_{1s} (= 1/(1 + aC_t))$ with $a \sim 1$, $\lambda_{dt} \sim 600 \mu\text{s}^{-1}$, $\lambda_{dt\mu-d}^0 \sim 600 \mu\text{s}^{-1}$ separately reported,⁷⁾ the $f_{d\mu}$ and $f_{t\mu}$ was determined as a function of C_t and fixed in the fitting for the

$\lambda_{d\text{He}\mu}$ and $\lambda_{t\text{He}\mu}$ in Eq. (1). The result of the present work is $\lambda_{d\text{He}\mu} = (6 \pm 2) \times 10^8 \text{ s}^{-1}$ and $\lambda_{t\text{He}\mu} = (4.6 \pm 0.3) \times 10^9 \text{ s}^{-1}$. Here it should be noted that the better fitting, of which result is mentioned above, was obtained by ignoring the effect of the hyperfine transition in $(t\mu)_{1s}$, i.e. by rejecting the $\lambda_{t\mu}^{10}$ term. This may imply that the thermalization of $(t\mu)$ is slow, and $(dt\mu)$ molecule formation from $(t\mu)_{1s}^{F=1}$ can not be neglected. Since $(t\mu)$ with the kinetic energy of the order of 10^{-1} eV has several resonant formation mode even if in $F = 1$ state,⁹⁾ $(t\mu)_{1s}^{F=1}$ may form a $(dt\mu)$ molecule before the hyperfine transition, and the contribution of $\lambda_{t\mu}^{10}$ term in Eq. (3) becomes smaller. The same analysis procedure was applied on the $tt\text{-}\mu\text{CF}$ data which is reported separately,⁵⁾ and its result was confirmed to follow the present result. Theoretically, $\lambda_{d\text{He}\mu}$ and $\lambda_{t\text{He}\mu}$ were calculated by Kravtsov et al. above $\sim 50 \text{ K}$.¹⁰⁾ For instance, supposing their simple-approach approximation, we forced to extrapolate from their result to the temperature of the present work, $\sim 16 \text{ K}$, to estimate the $\lambda_{t\text{He}\mu}$ and $\lambda_{d\text{He}\mu}$ as $\sim 7 \times 10^9 \text{ s}^{-1}$ and $\sim 4 \times 10^8 \text{ s}^{-1}$, respectively. The present result is in agreement with their theory in the same order of magnitude.

As already mentioned, the muonic molecule, like $(d^3\text{He}\mu)$, may have a chance to emit a characteristic X-ray with a broad and characteristic shape,^{2,3)} and such photons were also observed in the solid D-T mixtures. This fact is one of evidences supporting that the above-mentioned subject is due to the accumulated ^3He .

These quantitative understanding of experimental results on ^3He accumulation effect in the solid D-T mixture supports the picture of trapping of ^3He after tritium decay in the solid D-T mixture. Furthermore the present work shows the application of this effect to the study of the muon transfer phenomenon from hydrogen isotopes to ^3He .

Finally, let us discuss some possible mechanism of helium trapping in solid hydrogen. For instance, taking the case in which β -decay occurs in T_2 molecule, the kinetic energy of a recoil ^3He nucleus is $\sim 0.35 \text{ eV}$ at the average, and $\sim 3.4 \text{ eV}$ at the maximum. Since the binding energy of T-T bond is $\sim 4.6 \text{ eV}$, the ^3He forms a $(^3\text{HeT})^+$ ion with the tritium in the original T_2 molecule, rather than a $^3\text{He}^{++}$ ion or a $^3\text{He}^+$ is formed. In solid, this ion may be stucked to the original lattice point. On the contrary, in liquid, the ^3He in this ion may have a chance to neutralize in the collision with the other hydrogen molecules, and neutralized ^3He goes out of liquid easily. This model is simplified too much, and the practical trapping process may be more complicated. For example, in gas $(^3\text{HeT})^+$ ion is known to make a cluster with the surrounding hydrogen molecules.¹¹⁾ Thus the lattice around $(^3\text{HeT})^+$ may be distorted in solid. As the ^3He concentration increase, this distortion may overlap each other and cause the nucleation of ^3He , in which case the number of isolated $(^3\text{HeT})^+$ may be decreased as shown in the case of the solid $C_t = 0.7$ at $\tau \sim 30 \text{ h}$.

On the hand, different theoretical approach has been done. The recent calculation using Hatree-Fock approximation expects the existence of the bound position of $^3\text{He}^+$ ion in solid.¹²⁾

The authors acknowledge contributions to the μCF facil-

ity construction by Messrs. P. Wright, R. Hall, D. Haynes, W. A. Morris, G. Thomas and H. J. Jones. Contributions at the earlier stage of the μ CF facility construction by Dr. H. Umezawa and Professor H. Kudo are also acknowledged. Messrs. T. Hashimoto and H. Sugai are also acknowledged due to the THS construction. The authors would like to thank Professors A. Arima, M. Oda and other related persons at RIKEN and Drs. P. Williams, A. D. Taylor and W.G. Williams and others related persons at RAL for their kind supports and encouragements. Helpful discussions with Professor M. Kamimura, T. P. Das, Dr. J. S. Cohen and Dr. W. Higemoto are also acknowledged.

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