

Muon Transfer Rates from Muonic Hydrogen Atoms to Gaseous Benzene and Cyclohexane

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The chemical effect of the muon transfer process from muonic hydrogen atom was examined for benzene and cyclohexane under low pressure gaseous conditions. The muon transfer rates were investigated by measuring the resulting muonic X-rays resulting from muon irradiation of hydrogen and neon gas mixtures. The muon transfer rates to benzene and cyclohexane carbon atoms were very similar in the gaseous state. The results differed from previous results obtained for muon and pion transfer rates to benzene and cyclohexane under high density liquid conditions. It was concluded that the difference originated from the atomic muon state of the muonic hydrogen atoms with a large radius.

1. Introduction

A muonic atom is an atomic system in which one atomic electron is replaced with a negatively charged muon. The mass of a muon is 207 times larger than that of an electron and the lifetime of a muon is 2.2 μs . Muonic atoms are formed when a muon is captured in the Coulomb field of a nucleus. Because a muon has a different mass from that of an electron, the size and energy of atomic orbitals of the muonic atom are completely different from those of electrons. The binding energy of the atomic muon is very large and the radius of the muon atomic orbital is very small and the muon is close to the nucleus. Usually in the first step of the muon capture and muonic atom formation processes, the muon can be found at a highly excited atomic muon level, and the muon immediately de-excites to the muonic 1s state by emitting Auger electrons or characteristic muonic X-rays. When a muon is captured by a hydrogen atom, a muonic hydrogen atom that consists only of a muon and a nucleus (a proton) is formed. Because a muonic hydrogen atom has no electrons, the radius of the muonic hydrogen atom is approximately 200 times smaller than that of an electronic hydrogen atom. Therefore, the charge of the muonic hydrogen nucleus is strongly shielded by the compact muon orbital and muonic hydrogen atom can behave like a neutron. As a result, muonic hydrogen atom can diffuse freely, it can even pass through the electron clouds of other atoms. When the muonic hydrogen atom approaches the nucleus of another $Z \geq 2$ atom, the muon moves from the muonic hydrogen atom to a deeper atomic muon level of the $Z > 2$ atom and forms a new $Z > 2$ muonic atom. This process in compounds and mixtures is called muon transfer.¹

To investigate the muonic atom formation process, determination of the muon capture probability by each atom in a substance is important. Multiple models have been proposed to explain the muon capture probability.²⁻⁴ In these studies, the

chemical environmental effects in the muon capture process have been singled out and named the “chemical effect.” The muon capture probability strongly depends on the molecular chemical structure of the muon-capturing atom.⁵⁻⁷ For example, muon capture ratios per atom of carbon compared to those of oxygen are different between CO_2 molecule and CO molecule.⁸ Similar studies have also been intensively carried out with a negative pion; and because the negative pion has a similar mass to that of the negative muon, the formation processes for pionic atoms and muonic atoms are thought to be very similar.⁹⁻¹¹

The chemical effects of muonic (or pionic) atom formation in a hydrogen-containing system have also been reported.¹²⁻¹⁴ The capture probability of hydrogen atoms is dependent on the molecular structure of the capturing molecule, and is strongly influenced by the strength of the hydrogen bond.^{13,14} Changes in the formation process originate from the difference in the initial state of the captured muon in the hydrogen atom. On the other hand, all of the chemical effects from the transfer processes have not been fully examined. Muon transfer processes for hydrogen gas mixtures and other gaseous compounds under various pressure conditions were observed and the following muon de-excitation processes were examined.¹⁵ The pressure dependence on muonic X-ray intensity patterns (X-ray structures) has already been reported, however, it is not clear whether such a dependence is due to a chemical effect from the muon transfer process or the difference in the muon cascading process after muon transfer by electron refilling. In condensed substances, Shinohara *et al.*¹² reported a chemical effect for the pion transfer process between benzene and cyclohexane with carbon tetrachloride mixture. They reported that the pion transfer rate from pionic hydrogen atoms to the carbon atoms of benzene was about twice that of the carbon atoms of cyclohexane. They concluded the difference originated from the difference between benzene and cyclohexane in steric hindrance around the carbon atoms.¹² Inagaki *et al.*¹⁶ examined the chemical effect for muon transfer process in the same system and found a small difference in muon transfer

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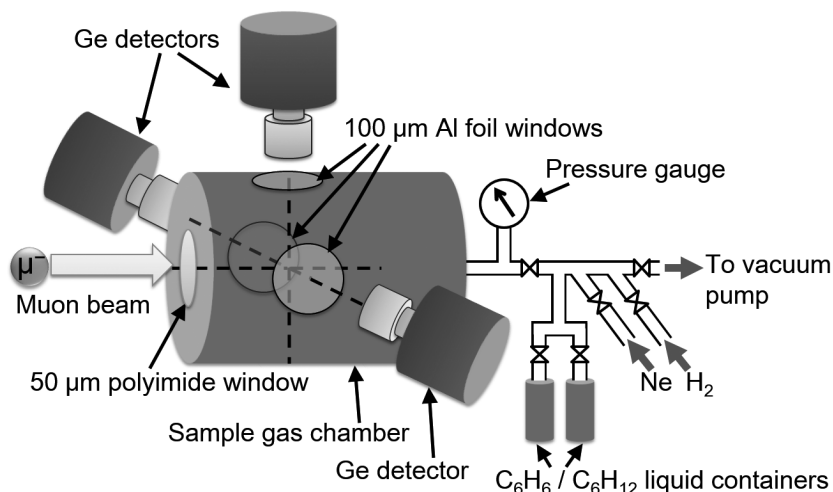


Figure 1. Schematic view of the experimental setup.

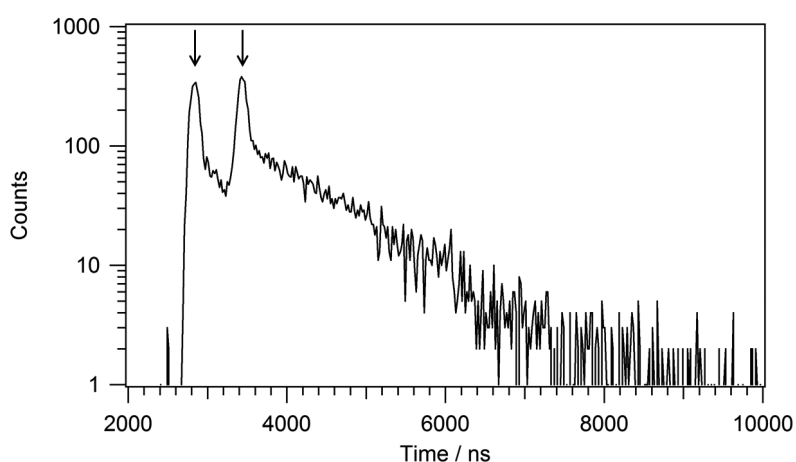


Figure 2. Time deviation spectrum between the accelerator operation signal and muonic X-ray detection signal. The two peak components (2830 ns and 3430 ns) correspond to the muon beam injection timing in the sample.

rates.

The chemical effects in the transfer processes have only been reported using condensed phase samples.^{12,16} In such high-density conditions, muon transfer occurs in a very short time scale. This fact implies that the chemical effects in the transfer processes are apparent only from the excited state of the muonic hydrogen atoms that can exist immediately after capturing a muon in a hydrogen atom. This is consistent with the existence of a chemical effects in the pion transfer processes because the pionic hydrogen atoms in the 1s state are unstable and the pion transfer processes occur only from excited state.

In this study, an examination was made on the chemical effect in the muon transfer in benzene and cyclohexane in a low-density gaseous state and the obtained results were compared with these in high-density conditions.¹⁶ Under low pressure conditions, the muon transfer rate should be low and thus, the muon transfer process component was separated from that of direct muon capture. Additionally, under such conditions, it was possible to extract the ground state muonic hydrogen atom contribution.

2. Experimental

The muon irradiation experiment was performed at the MUSE D1 beam line in the Materials and Life Science Experimental Facility of J-PARC, Ibaraki Prefecture, Japan in 2014. The experimental equipment consisted of a gas cham-

ber, a diaphragm-type pressure gauge, a sample vessel, and a gas handling system. A schematic view of the experimental setup is shown in Figure 1. A thin polyimide film of 50 μm thickness was used for the muon entering window. The measured samples were: C_6H_6 (0.2 kPa) + Ne (3.0 kPa) + H_2 (96.8 kPa), labeled as the C_6H_6 sample; C_6H_{12} (0.2 kPa) + Ne (3.0 kPa) + H_2 (96.8 kPa), labeled as the C_6H_{12} sample; and Ne (3.0 kPa) + H_2 (97.0 kPa), labeled as the blank sample. The measurement times for each sample were 9.5 h, 9.1 h, and 3.7 h respectively. Each sample was prepared by introducing the gases into the chamber to the requisite target pressure in the order of benzene or cyclohexane, neon, and hydrogen.

A pulsed muon beam with a momentum of 19 MeV/c was irradiated on each sample gas, and the emitted muonic X-rays were measured using three germanium semiconductor detectors (Canberra GL0515R, Ortec Loax 36300, and Ortec GLP-16195). All muonic X-ray events were recorded for both energy and timing information, which is the elapsed time from the frequency signal supplied from the accelerator facility to the X-ray detection. The EGS5 Monte Carlo simulation code¹⁷ was used to determine the detection efficiencies of the germanium detectors based on a standard radiation source measurement.

3. Results

The time spectrum between the accelerator signal and muonic X-ray detection for the C_6H_6 sample is shown Figure 2.

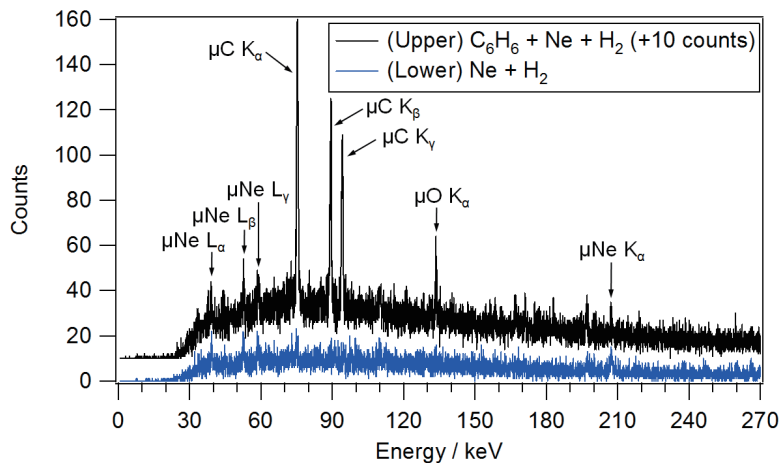


Figure 3. Muonic X-ray spectra for the C_6H_6 and blank samples.

TABLE 1: Muonic X-ray intensity ratios of muonic carbon Lyman series to muonic neon Lyman series and Balmer series for C_6H_6 and C_6H_{12} samples. The intensities of muonic X-rays were determined from the sum of series X-rays. For example, the μNe Balmer was obtained from the sum of L_{α} , L_{β} and L_{γ} X-rays of muonic neon. The ratios correspond to the ratios of muon transfer rates.

	C_6H_6 sample	C_6H_{12} sample	C_6H_6 sample / C_6H_{12} sample
μC Lyman / μNe Balmer	6.4 ± 0.7	5.7 ± 0.5	1.1 ± 0.2
μC Lyman / μNe Lyman	8.0 ± 1.3	8.8 ± 1.6	0.9 ± 0.2

Around 2830 ns and 3430 ns, a peak component derived from the muon direct capture process was observed just after the muon injection timing (prompt component) and a decreasing signal over time was identified as the muon transfer process (delayed component). The prompt component consisted of a double peak approximately 100 ns in width with a 600 ns interval time that was coincident with the incident muon beam cycle of J-PARC/MUSE. The events originating from the muon transfer process were obtained delayed from the muon beam pulse because the muon transfer process occurs after muonic hydrogen atom formation and collision with another nucleus. Therefore, investigate the muon capture event was investigated from the muon transfer process by extracting the delayed component.

Figure 3 shows the energy spectra of the delayed components from 2980 ns to 3280 ns and from 3580 ns to 12230 ns of the C_6H_6 and the blank samples. In the C_6H_6 sample, X-rays of the neon Balmer series ($\mu Ne L_{\alpha}$, $\mu Ne L_{\beta}$ and $\mu Ne L_{\gamma}$), carbon Lyman series ($\mu C K_{\alpha}$, $\mu C K_{\beta}$ and $\mu C K_{\gamma}$), and neon Lyman series ($\mu Ne K_{\alpha}$) were identified. A similar spectrum was obtained from the C_6H_{12} sample. Small signals from muonic oxygen, nitrogen, and carbon atoms were observed in the spectrum of the blank sample. These signals were derived from the polyimide film used for the muon entrance window of the chamber. The X-ray intensity ratio of carbon and oxygen in the blank sample spectrum was used, and the muonic carbon X-rays contributed from the polyimide film were corrected in the spectra of the C_6H_6 and C_6H_{12} samples.

From the series of muonic X-ray intensities, the number of captured muons in each element were determined. Table 1 shows the ratio of the muonic X-ray intensity of the delayed component of carbon to that of neon in the C_6H_6 and C_6H_{12} samples. Since the muon transfer rates to neon is reasonably assumed to be equal in both samples, the muonic X-ray intensity ratio shows the relative values of the muon transfer rates to carbon based on the muon transfer rate to neon. The values using the neon Lyman series were smaller than the values

using the neon Balmer series because K_{β} and the subsequent events on the neon Lyman series could not be observed due to the low detection efficiency. The muon transfer rates to carbon in the C_6H_6 and C_6H_{12} samples were within the experimental error derived from peak count statistics, as shown in Table 1. This indicated that no clear difference was found in on the muon capture for carbon in these two molecules.

4. Discussion

In this experimental system, no significant difference in the muon transfer rate to carbon atoms was observed between benzene and cyclohexane. This result differed from those obtained in the liquid system previously.¹² For pion transfer, the transfer rate to the carbon atom of benzene is two times higher than that of cyclohexane.¹² Such a difference would be caused by the difference in muonic hydrogen atom states between the two systems. The sample densities of benzene and cyclohexane in this experiment were 5 orders of magnitude lower than those in the corresponding liquid system. The muon transfer rate to carbon in the present experimental system was estimated to be on the order of $10^5 s^{-1}$ from the decay constant of the delayed component shown in Figure 2, and the muon transfer was completed with an average time on the order of $10^{-6} s$. This time scale is slower than the muon cascading process in a hydrogen atom; de-excitation to the 1s or 2s state is completed within $10^{-7} s$, and most of the muons in hydrogen atom reach the 1s state while the muon transfer proceeds.¹² On the other hand, considering the density ratio in gas and liquid samples, the muon transfer rate in the liquid system was estimated to be on the order of $10^{10} s^{-1}$, and the transfer was finished with a mean time on the order of $10^{-11} s$. At $10^{-11} s$, most of the captured muons have not reached the muonic 1s state and 70% of the captured muons remain in a state with a principal quantum number $n \geq 3$.¹⁸ Therefore, in the liquid system, muon transfer was considered to mainly occur from the excited state with a principal quantum number $n \geq 3$.

Likewise, in the case of pion transfer in a liquid system, the transfer occurs only from the excited state, because in pionic hydrogen atoms, due to the strong interaction between the pion and proton, the lifetime in the ground state is extremely short ($\sim 10^{-15}$ s). On the other hand, in the case of muonic hydrogen atoms, a strong interaction does not exist between the muon and proton and the ground state lifetime is long (10^{-6} s). In this way, pion transfer in the liquid state occurs only from the excited state of pionic hydrogen atoms, most of the muon transfer in the liquid state occurs from the muonic hydrogen atoms at the principal quantum number $n \geq 3$, and muon transfer in the gas state occurs from the ground state muonic hydrogen atoms.

The difference in steric hindrance around the carbon atom of benzene and cyclohexane causes a chemical effect in the transfer process, that is, a difference in the transfer rates to carbon atoms.¹² When a muonic hydrogen atom, or a pionic hydrogen atom, approaches a cyclohexane carbon atom, the steric hindrance is larger than in the case of benzene. In the case of benzene, only one hydrogen atom is bound to each benzene carbon atom, whereas two hydrogen atoms are bound to the cyclohexane carbon atom. Such a steric hindrance may strongly influence the transfer from the excited state of muonic or pionic hydrogen atoms. For example, a pionic hydrogen atoms with the principal quantum number $n = 3$ has a radius approximately 9 times larger than that of a muonic hydrogen atoms in the ground state. As a result, a significant chemical effect on the muon transfer process was not observed in the present experimental system, in which muon transfer occurred only from the ground state muonic hydrogen atoms.

4. Conclusion

The muon transfer rates in gaseous benzene and cyclohexane were investigated from the relative muonic X-ray intensities of carbon to those of neon. Although a chemical effect on the muon and pion transfer processes were reported between benzene and cyclohexane in a liquid system, such a chemical effect was not observed within the measurement accuracy of this experiment. In gaseous state muon transfer, most of the muon transfer occurred from the muonic hydrogen atoms in the ground state. Ground state muonic hydrogen atom has a small radius and the effect of steric hindrance was very small. On the other hand, in a liquid system, most of the muon and pion transfer processes occur from the excited states. As a result, steric hindrance during transfer is large and a difference in the transfer process between benzene and cyclohexane is observed.

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