

# Formation Process of Pionic Atoms and Their Behavior in Material

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Exotic atoms are the interesting object as a new probe in material science and a new chemical species in future chemistry. Our group ( $\pi$ AX) has recognized the mesoatomic and mesomolecular phenomena as a subject of chemistry, and continued the experimental study for the capture process of negative pions since 1982. In the recent study, we have investigated the pion capture process mainly in liquid and gas phases, focusing to the behavior of pionic hydrogen atoms. A novel method to measure selectively the pion capture for a particular hydrogen in a molecule revealed the effect of the chemical state of the hydrogen on the transfer process. A proposed model, which combines a LMM model with a transfer process, was useful to wholly understand the capture process observed. The outline of the formation process of pionic atoms and its behavior in material is described briefly, and the main results of our studies are reviewed.

## 1. Background of the Present Studies

Recent progress of an accelerator has allowed us to apply various exotic atoms to material science, analytical chemistry, medical science, etc, other than particle physics. Especially, the use of muons has been increasing, such as  $\mu$ SR, muonium chemistry,  $\mu$ CF, and so on, in addition to the use of positrons. Muonic or pionic atoms, which consist of a negative muon or pion and an ordinary atom (nucleus), have been studied since the discovery of the atoms, and the outline of the formation and decay process of the atoms has been established.<sup>1-5</sup> However, more detailed and fundamental studies for formation mechanism and behavior of such atoms are required to develop a new chemistry of the 2nd generation substance (muonic or pionic atoms and molecules) and new application of the phenomenon.

It is accepted that the formation and decay of a pionic atom take place according to the following step. A negative pion in the first stage of the slowing down in matter loses the energy by ionization, and it is slowed down by collision with electron of comparable velocities until finally it is captured by the Coulomb field of an atom. The pion captured in an outer Bohr orbit cascades down accompanied by the emission of Auger electrons and by the pionic X rays. In the capture process, "chemical effects" are observed because electrons are considered to affect the Coulomb field in the capturing stage. In the lower levels, the strong interaction affects the cascade scheme and finally causes the nuclear absorption of the pion. Complete understanding has not yet been attained for the mechanism of molecular and atomic capture of pions.

A large mesomolecular (LMM) model is more useful to describe the capture process including the chemical effects among various capture models proposed. This model was proposed by Ponomarev et al.<sup>1</sup> to explain the obvious chemical effect observed in hydrides, and extended to the capture phenomenon in general molecules by Scheuwly et al.<sup>3</sup> In the first step (probability P) of the model, it follows that the pion is captured on the LMM orbital when the valence electron emission occurs, and on the atomic orbital when the core electron of the corresponding atom is emitted as Auger electron. The pion on the LMM orbital moves to an atomic state of any atom in the molecule and then the pionic atom forms in the second

step (probability Q). If the pion is captured by hydrogen, the capture probability of hydrogen ( $W_H$ ) is expressed by

$$W_H = PQR, \quad (1)$$

where R means the probability that the pion in the pionic hydrogen is finally captured by the proton without transfer to other atoms.

Ponomarev well explained the hydrogen effect by assuming the second step to be radiative process. Scheuwly could take the chemical effects into account in the LMM model by treating the probability P as a localization factor defined by the ionicity parameter  $\sigma$  introduced by Paurling. In our previous works,<sup>6-9</sup> we proposed a modified LMM model, in which the second step proceeds via Auger transition, based on the large chemical effects observed for the pion capture ratio in a series of beryllium borides ( $Be_xB_y$ ) and in  $MBH_4$ , MOH, and MF ( $M=Li, Na, K$ ).

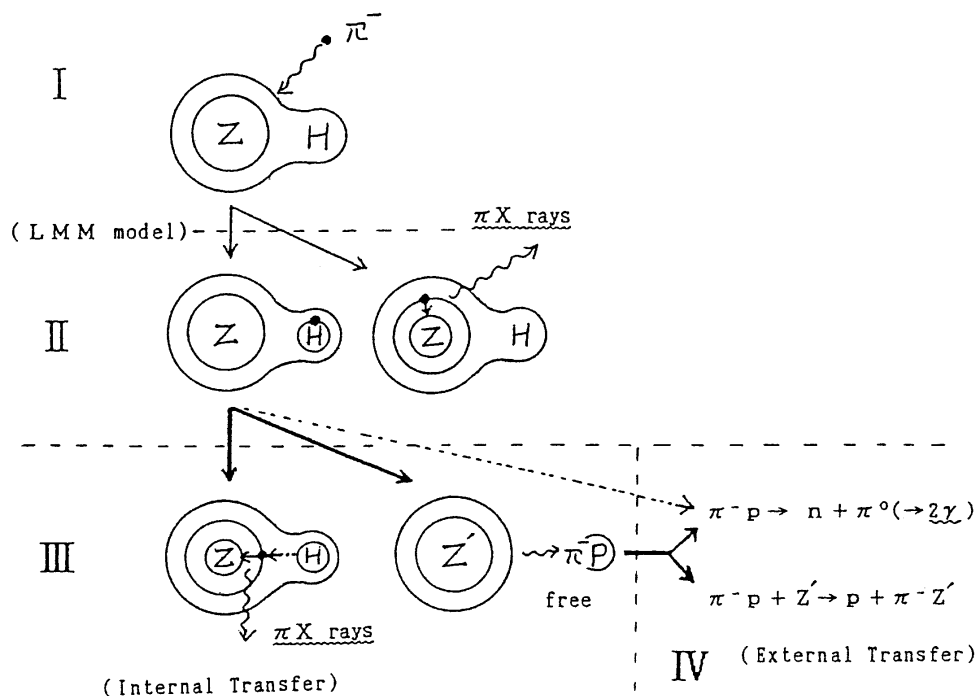
As for the pion transfer phenomenon, many gas phase experiments performed mainly by Russian group showed that the pion transfer from pionic hydrogen atoms to other heavier atoms occurs in gas phase in spite of its short life time. The semi-empirical formulae for the transfer rate were also derived from the systematic measurements.<sup>2,4,10,11</sup> The influence of chemical environment on the transfer process, however, was scarcely observed and was not involved in such phenomenological treatment.

## 2. Theme and Goal of the $\pi$ AX Project

The goal of the  $\pi$ AX project lies in comprehensively understanding the formation process and behavior of pionic atoms and molecules in order to apply the phenomenon to material science, analytical chemistry, bio and medical science and to develop a new chemistry of 2nd generation substance (muonic or pionic atoms). Especially, the quantitative treatment for pionic hydrogen atoms will provide a unique probe for hydrogen in material. Figure 1 shows the capture and decay processes of negative pions in hydrogen-containing molecule (Z-H). In the present studies (Visiting Researcher's Program of KEK-PS: E202, E262, E360), we focused the formation and behavior of pionic hydrogen atoms, as shown with the bold lines in the figure.

The observable quantities are the pionic X rays and two  $\gamma$  rays from  $\pi^0$  decay (indication of the final capture by hydro-

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**Figure 1.** Pion capture process in hydrogen-containing molecules (Z-H).

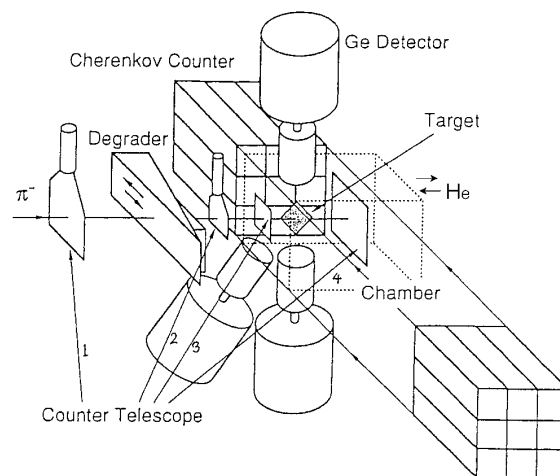
gen). The former includes the information on the internal transfer, and the change of the latter against the concentration of Z'-atom reflects the external transfer rate. The selection of the bond nature and structure of the Z-H molecule is also additional degree of freedom in the research technique from the viewpoint of chemistry.

Many experiments were carried out with a newly developed apparatus, which allows us to measure simultaneously two  $\gamma$  rays and low-energy pionic X rays, to achieve the goal mentioned above. These experiments are classified into the following theme.

- (1) Selective measurement for the capture probability on the hydrogen at a particular site in the molecule.<sup>14,15</sup>  
Systems: R(alkanes), ROH/ROD, RCOOH/RCOOD.
- (2) Measurement of the external transfer rates by two-component systems in liquid and gas phases.<sup>16,17</sup>  
Systems:  $C_6H_{12}/C_6H_6 + CCl_4/CS_2$ ,  $H_2/CH_4 + Z$  (rare gas).
- (3) Measurement of pressure dependence of transfer rate in gas phase (in progress).  
Systems:  $H_2/CH_4 + Ar$  (chaining pressure).
- (4) Transfer and structure effects appearing in the intensity patterns of pionic X rays.<sup>18,19</sup>  
Systems: Fullerenes,  $H_2/D_2/CH_4 + SF_6$ , & All samples studied.
- (5) Measurement of the fundamental data (Pionic X-ray yield, correction data,...).<sup>20</sup>  
Systems: Various elements,  $H_2O + D_2O$ ,...
- (6) Application of the LMM model.<sup>21</sup>  
Systems:  $M_x(acac)_y$ ,  $MH_x$ .
- (7) Proposal for the combined LMM model (LMM + transfer).<sup>14</sup>  
Based on (1) and (2).
- (8) Proposal for a new model including the dynamical behavior of pionic hydrogen (in progress).

### 3. Experimental Method

The experimental setup was installed at the  $\pi\mu$ -channel<sup>22</sup> of the 12-GeV proton synchrotron of the High Energy Accelerator Research Organization (KEK). The channel comprises a D4Q2D2Q lens system and has a length of 11.0 m from the production target to the sample position. The apparatus comprised an energy degrader, a collimator, a radiation shield, a defining counter system, and a measuring chamber, as



**Figure 2.** Schematic view of the experimental setup.

shown in Figure 2. Negative pions were slowed down by the graphite degrader so as to stop in the target. Incident and stopping pions in the sample were counted using a conventional counter telescope comprising four plastic scintillation counters (PS1, PS2, PS3, and PS4 (veto)).

The pionic X rays were measured with two or three Ge detectors in coincidence with the stop events ( $1 \times 2 \times 3 \times 4$ ). Two coincident  $\gamma$  rays from a charge-exchange reaction,  $p + \pi^- \rightarrow n + \pi^0$ ,  $\pi^0 \rightarrow 2\gamma$  (70 MeV each, 60% branching ratio), were detected using a pair of Cherenkov-detector arrays. The beam size at the sample position was defined as  $4(H) \times 3(V) \text{ cm}^2$  by the collimator and the defining counters (PS2 and PS3 in Figure 2).

The solid or liquid sample was set in the chamber filled with helium gas in order to avoid any disturbance from air.<sup>14</sup> A rectangular container [ $5.5(H) \times 7.0(V) \text{ cm}^2$  in area and 2.5, 5.0, or 7.5 mm in thickness] made of thin beryllium or aluminum foil was used for liquid and powder samples. The self-supporting sample was also prepared for some compounds as a pressed method. A high-pressure gas chamber was specially designed for measurement of low-energy pionic X rays and  $\pi^0$  decays from a gas sample.<sup>18</sup> The chamber had cylindrical form to bear high pressure ( $50 \text{ kgf/cm}^2$ ) and had the sample volume of  $125 \text{ cm}^3$ . The 2.5 mm thick chamber wall was made of berylli-

um to measure low-energy pionic X rays (ex. 18keV for carbon). PS-3 and PS-4 counters were mounted inside the chamber and used as the phoswich type counter system.

#### 4. Main Results of the Present Project

Among the above theme, the results of (1), (3), and (7) were the noticeable ones in our work. Especially, the selective measurement for hydrogen was successfully done by using the deuterated compounds.<sup>15</sup> The dependence of the transfer rate on gas pressure was also observed at the first time in the CH<sub>4</sub>+Ar system. The former will provide a useful technique to investigate individually the behavior of the pionic hydrogen atom originating from a hydrogen atom at different chemical states in a molecule or material. These results require a new model for the capture and transfer processes, which include the life time of the pionic hydrogen atom and the chemical effects in the transfer process. Main results of the project are briefly described below.

**4.1. Measurements for External Transfer in Alkanes and a Combined LMM Model.**<sup>14</sup> We attempted to reveal the contribution of the external transfer process in the pion capture for hydrogen-containing compounds in the condensed phase. We then measured precisely the capture process in a series of hydrocarbon (liquid alkanes), which had been analyzed with no contribution of the external transfer so far.<sup>23</sup> To explain the result obtained, we proposed a new model involving a modified LMM model and an external transfer process as mentioned below.

In the modified LMM model,<sup>7,9</sup> we introduced a parameter  $a_\gamma$  to express the capture probability on hydrogen, where  $a_\gamma$  is defined as a ratio of the sum of the radiative and the nuclear absorption rates to the Auger transition rate. The probability of pion capture by hydrogen  $W_H$  is obtained from eq 2:

$$W_H = \frac{\nu(1+\sigma) a_\gamma}{(N+\nu)(1-\sigma)Z_{\text{eff}}}, \quad (2)$$

where  $N$  is the sum of the core electrons relevant to the capture process ( $N=kn_2+ln_1$ ,  $n_1=0$ ),  $\nu$  presents the total valence electrons ( $\nu=kn_2+l$ ).<sup>9</sup> The factor  $a_\gamma/Z_{\text{eff}}$  is thought to correspond to the the product of the parameter  $Q$  and  $R$  from a comparison with eq 1. In this model, therefore, we approximate the parameter  $a_\gamma$  as the product of two factors,  $a_\gamma'$  and  $R'$ , where  $R'$  is the non-transfer probability associated only with the external transfer. The capture probability  $W_H$  is then rewritten as

$$W_H = \frac{\nu(1+\sigma) a_\gamma'}{(N+\nu)(1-\sigma)Z_{\text{eff}}} R'. \quad (3)$$

The probability  $R'$  can be expressed approximately by

$$R' = 1/(1 + \Lambda_Z C_Z), \quad (4)$$

based on a phenomenological model,<sup>2</sup> where  $\Lambda_Z$  is the reduced pion-transfer rate relative to the charge-exchange rate in the collision with  $Z$ -atoms, and  $C_Z$  is the ratio of the number of the  $Z$ -atom to the hydrogen atom. The  $a_\gamma'$  value should be recognized as being the probability of isolation of a  $\pi p$  from the  $Z$ -H bond or of absorption by the proton without experiencing an internal transfer (the latter probability is assumed to be negligibly small in eq 2) and hence is determined as a particular parameter for the kind of  $Z$ -H bond.

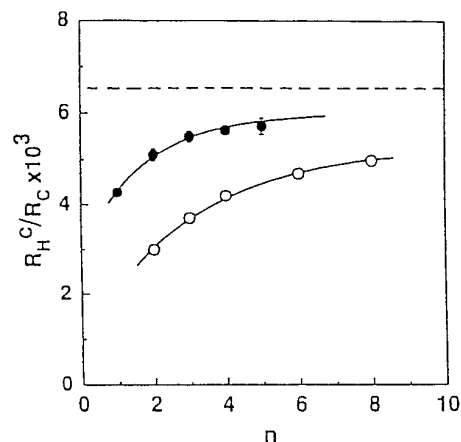
In conclusion, we found the contribution of the external transfer in liquid alkanes and obtained the transfer rate parameter for carbon as  $\Lambda_C=1.7\pm 0.2$  and the parameter for isolation process from the molecule as  $a_\gamma'=0.140\pm 0.008$  based on the proposed model. The obtained transfer parameters are summarized in Table 1 together with the semi-empirical estimates.<sup>4</sup>

#### 4.2. Selective Measurement for the Capture on Hydrogen.

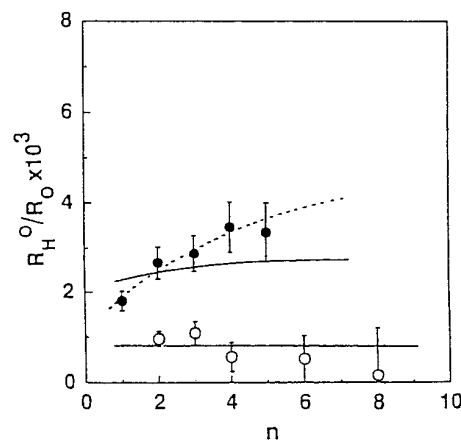
In the analysis of the pion capture process, the main problem is the fact that the capture probability for a particular  $Z$  atom is

given as an average over the chemically different states of the element in the molecule. Observation for the pion capture probability on a particular atom in the molecule, if possible, will give significant information for understanding the pion capture mechanism. We then tried to extract the capture rate for a particular hydrogen atom using deuterated compounds. The capture events of pionic hydrogen undergoing no transfer can be measured by two- $\gamma$ -rays of  $\pi^0$  produced by the charge exchange reaction. On the contrary, the reaction of a pionic deuterium ( $\pi d$ ) is strongly suppressed (about  $10^{-4}$ ).<sup>24</sup> The measurement for deuterated molecules therefore provides the capture probability for hydrogen except for hydrogen at the deuterated site. Thus we can obtain the capture rate for a particular hydrogen atom with a comparison between the ordinary compound and the deuterated one.

We selected a series of alcohols, carboxylic acids, and the corresponding compounds with a deuterated hydroxyl (-OD) or carboxyl (-COOD) group to study the behavior of the pionic atoms originating from hydrogen in different chemical states. As a result, we found that pionic hydrogen atoms from hydroxyl and carboxyl groups show different behavior as demonstrated in Figures 3a and 3b. The pionic hydrogen from hydroxyl group strongly favors the transfer to oxygen and that from carboxyl group shows almost no transfer phenomenon. The transfer rate parameters  $\Lambda_O$  of individual pionic hydrogen for oxygen were determined based on the combined LMM model using  $\Lambda_C=1.7$  mentioned above. The result indicates that the chemical state of the hydrogen in the molecule affects the life or



**Figure 3a.** Capture ratios per atom of the hydrogen bound to carbon relative to the carbon,  $R_H^C/R_C$ , as a function of the number ( $n$ ) of carbon atoms for alcohols (●) and carboxylic acids (○). The dashed line represents the capture ratio for polyethylene. Solid curves are the model calculations.



**Figure 3b.** Capture ratios per atom of the hydrogen bound to oxygen relative to the oxygen,  $R_H^O/R_O$ , as a function of the number ( $n$ ) of carbon atoms for alcohols (●) and carboxylic acids (○). Solid curve is the model calculations with  $\Lambda_C=1.7$  and  $\Lambda_O=4.1$ , and the dotted curve represents those with  $a_\gamma'=0.14$  for alcohols. No external transfer was found for carboxylic acids.

velocity of the isolated pionic hydrogen atom and causes the chemical effect in the transfer process.

**4.3. Measurement of the External Transfer by Two-Component Systems in Liquid and Gas Phases.**<sup>16,18</sup> The external transfer process is explicitly observed in two-component system consisting of the hydrogen-containing molecule and the hydrogen-non-containing one. For the liquid phase, we selected  $C_6H_{12}$  or  $C_6H_6 + CCl_4$  or  $CS_2$  systems which don't react with each other and mix with each other in any concentration. The external transfer rate was derived from the dependence of two- $\gamma$ -events on the capture fraction of  $C_6H_{12}$  or  $C_6H_6$  molecule, which was obtained from the pionic X-ray measurement. In this study, we revealed the structure effect for the transfer process by comparing between the transfer rate for carbon of alkaline ( $sp^3$  structure) and that for carbon of benzene ( $sp^2$  structure) as shown in Table 1. The transfer rates at 51kgf/cm<sup>2</sup> were also measured for the  $H_2 + Z$  and  $CH_4 + Z$  ( $Z=He, Ne, Ar, Kr, Xe$ ) systems to compare with the systematics for the atomic number  $Z$ . The obtained external transfer rate parameters are listed in Table 1 and plotted as a function of  $Z^{1/3}$  in Figure 4 together with the predictions by the semi-empirical formulae.<sup>4,10,11</sup>

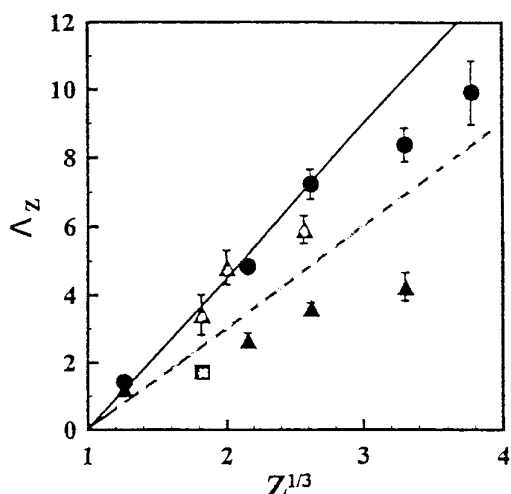
**TABLE 1: Transfer Rate Parameters Obtained in the Present Study**

System (liq.)	$\Lambda_Z$					
	$Z=$	C	O	Cl( $CCl_4$ )		
R(alkanes)		1.7±0.2	—	—		
ROH		1.7*	4.1±0.5	—		
RCOOH		1.7*	4.5±0.5	—		
$C_6H_{12}+CCl_4$		1.7*	—	5.9±0.4		
$C_6H_6+CCl_4$		3.4±0.6	—	5.9*		
Systematics**		2.5	3.0	4.7		
System (gas)	$Z=$	He	Ne	Ar	Kr	Xe
$H_2+Z$		1.39±0.10	4.83±0.22	7.21±0.43	8.35±0.49	9.89±0.95
$CH_4+Z$		1.2 ±0.1	2.64±0.22	3.59±0.17	4.23±0.42	—

\* The parameter was fixed in the analysis procedure.

\*\* Estimated from the semi-empirical formula for  $C_mH_n+Z$  (Ref. 4).

**4.4. Pressure-Dependence of the Transfer Rate in the Gas Phase (in Progress).** The dependence of the transfer rate on the gas pressure has not been observed so far in the muon or pion capture process for gas mixtures. The pressure-dependence may be observed for the pionic atoms from the bound hydrogen, which may have a short life as mentioned above, if the running path of the pionic hydrogen is short and comparable to the mean free path in the material. We then measured the change of the transfer rates at the pressure 50kgf/cm<sup>2</sup>, 13.5kgf/cm<sup>2</sup>, and 6.0kgf/cm<sup>2</sup> for the  $H_2 + Ar$  and  $CH_4 + Ar$  systems.



**Figure 4.**  $Z$ -dependence of the transfer rate parameters ( $\Lambda_Z$ ). Solid line is the semi-empirical estimates for  $H_2+Z$  system,<sup>4,10</sup> and dashed line for  $C_nH_m+Z$  system.<sup>4,11</sup> ●:  $H_2+Z$ , ▲:  $CH_4+Z$ , ○:  $C_mH_n+Z$ (liq.), □:  $C_6H_6+Z$ (liq.),  $Z$ =alkanes.

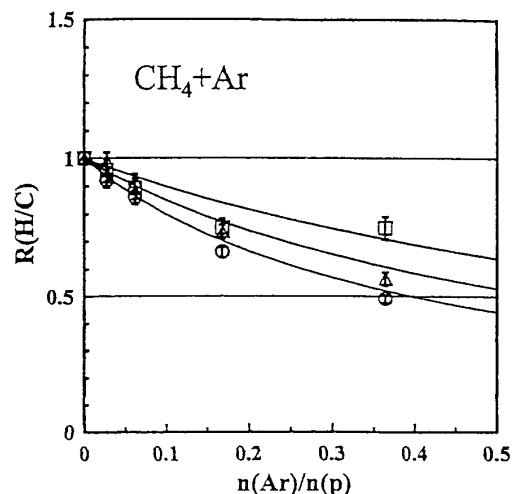
Figure 5 shows the pion capture ratios of hydrogen to carbon against the fraction of Ar for different pressures for the  $CH_4 + Ar$  system. One can see small change but apparent change of the transfer rate with pressure in the figure. On the other hand, we can observe no change of the transfer rate for the  $H_2 + Ar$  system.

We can roughly estimate for the mean free path in Ar to be about  $10^{-8}$  m in gas phase at the pressure of 50kgf/cm<sup>2</sup>, assuming that the effective interaction radius is an atomic dimension, and for the traveling length ( $t_v$ ) of the pionic hydrogen atom to be  $6 \times 10^{-10}$  to  $2 \times 10^{-8}$  m, providing that the velocity ( $v$ ) of it is estimated from the kinetic energy of 1–2 eV.<sup>25</sup> Then overlapping between the ranges of the mean free path and the traveling length suggests the possibility that the transfer rate changes depending on the gas pressure and between the gas and condensed phases. If the observed result is attributed to the mechanism mentioned above, the fact that the pressure-dependence was observed only for the  $CH_4$  system is consistent with the difference in the life time ( $t$ ) between the excited states of the pionic hydrogen atom from  $CH_4$  and that from  $H_2$ .

**4.5. Transfer and Structure Effects Appearing in the Intensity Patterns of Pionic X Rays.**<sup>13,18</sup> Change of the intensity pattern and yields of pionic X rays will be caused by the contribution of the pion transferred from a pionic hydrogen as well as the different angular momentum distribution on the initial stage of the capture process. The relation of the change of intensity ratio was investigated from the viewpoint of the molecular structure and the number of hydrogen for the pionic X rays of carbon and oxygen for most samples studied. It was found that the pion brought by the internal transfer mainly contribute the atomic cascade for the pionic X rays of the atom bound to hydrogen. The pure structure effect was observed in the measurements for carbon allotropes including fullerenes and for various single substances. We discussed about the atomic levels concerned with the transfer based on the cascade calculations including the contribution of the transferred pion.

## 5. Summary

Figure 4 shows the dependence of the transfer rate parameters obtained on the atomic number and a comparison between the transfer rates in the liquid phase and those in gas phase. One can see that the present results slightly deviate from the  $Z^{1/3}$ -dependence which is assumed in the empirical formulae, and that the transfer rates in liquid phase are rather faster than those in gas phase for alkane systems. The pressure-dependence of the transfer rate was also found for alkane systems. It is generally accepted that the difference between the  $H_2$  system



**Figure 5.** Pion capture ratios of hydrogen to carbon atoms against the fraction of Ar at different gas pressure. ○: 51kgf/cm<sup>2</sup>, △: 13.5 kgf/cm<sup>2</sup>, □: 6.0kgf/cm<sup>2</sup>.

and CH<sub>4</sub> system in gas phase is attributed to the fact that the pionic hydrogen atom from the bound hydrogen (CH<sub>4</sub>) lies in a lower excited state compared with that from H<sub>2</sub> molecule.<sup>4</sup> If this means that the life of the pionic hydrogen from alkanes is shorter than that from H<sub>2</sub>, the results of the pressure dependence is qualitatively consistent with the difference between the liquid and gas phases. The present result also indicates that a dynamical model, in which the life, velocity, molecular structure, and so on are taken into consideration, is required for the pionic hydrogen (with short life) originating from bound hydrogen.

In conclusion, the combined LMM model and the cascade calculation including transfer process successfully explained the results of the capture rates, transfer rates, and pionic X-ray intensity patterns for most sample systems studied. Here we can conclude that these model calculations are very useful to understand the formation and decay process of most pionic atoms in various materials. After this, quantitative discussion about the discrepancy between the results of liquid and gas phases and further accumulating the data of  $a_1'$  and  $\Lambda_Z$  for various chemical systems are needed to achieve the goal of our study.

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