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Visualized Characterization of Molecular Mobility for Water Species in Foods

Yasuyuki Konishi and Masayoshi Kobayashi

***Abstract*—**Six parameters, the effective diffusivity (*De*), activation energy of *De*, pre-exponential factor of *De*, amount (ASOW) of self-organized water species, and amplitude (α) of the forced oscillation of the molecular mobility ($1/\tau_C$) derived from the forced cyclic temperature change operation, were characterized by using six typical foods, squid, sardines, scallops, salmon, beef, and pork, as a function of the correlation time (τ_C) of the water molecule's proton retained in the foods. Each of the six parameters was clearly divided into the water species A_1 and A_2 at a specified value of τ_C $=10^{-8}$s (=Cτ_C), indicating an anomalous change in the physicochemical nature of the water species at the Cτ_C. The forced oscillation of $1/\tau_C$ clearly demonstrated a characteristic mode depending on the food shown as a three dimensional map associated with $1/\tau_C$, the amount of self-organized water, and τ_C.

***Keywords*—**molecular mobility, self-organization, hysteresis, water species A_1 and A_2, forced cyclic temperature change operation (FCTCO)

I. Introduction

IN the field of food engineering, as has been discussed by many researchers, the water activity of foods has been conveniently employed to create a stability map [1] for various reactions in foods illustrating the appearance of a specified water activity. Water activity (a_w) has commonly been used as a parameter to evaluate the deterioration of foods [2]. Water species retained in foods, as is well known, expose a multifunctional nature dynamically responding to environmental conditions, such as temperature, pressure, dehydration, and water content. For the scientific analysis of the multifunctional water species retained in foods, a recognized difficulty is related to its nonlinear dynamic change, which influences food quality.

To demonstrate the nonlinearity of the water species, the effective water diffusivity (*De*) in foods has frequently been used to evaluate the dehydration rate of food [3]. In previous studies [4]–[6], the water species retained in fish-past-sausage and squid were categorized into two species as a function of the water content. The two were water species A_1, recognized as the higher water content at W_0 >120%-d.b., with higher water diffusivity, *De*, and water species A_2, W_0 <120%-d.b., with lower *De*.

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For the quantitative evaluation of the water species in foods, the proton NMR technique was recently applied because of the need to use physicochemical data rather than the water activity [7], [8]. Focusing on a more direct identification of the water species on the molecular level, a proton nuclear magnetic resonance technique was applied to evaluate the food quality as demonstrated in the International Conferences on Applications of Magnetic Resonance in Food Science [9]. From the viewpoint of the molecular mobility of water species, the progress of research was recently accelerated to try to demonstrate the theoretical meaning [9], [10] and applications in various researches [11]–[14].

In the present study, a dynamic proton NMR technique is proposed as a new technique. The aims of this study are (1) to identify the two water species, A_1 and A_2, by using the effective diffusivity (*De*) as a three-dimensional mobility, the activation energy (E_D) of *De*, and the pre-exponential factor of *De*; (2) to visualize the self-organization of the two water species by using the dynamic NMR technique; (3) to visualize the forced oscillation of the molecular mobility ($1/\tau_C$); and (4) to demonstrate the dynamism of the amount of the self-organized water species.

II. Experimental Methods

A. Materials and Methods

Pork meat (P_H: produced in Hokkaido), beef meat (B_A: produced in Australia and B_H: produced in Hokkaido), squid, salmon, sardine, and scallop, were chosen as food samples. The pork and beef had an initial water content of 230~320%-d.b. and 230~280%-d.b., respectively; the initial water content of the squid and the salmon commonly ranged from 300~360%-d.b., and that of sardine and scallop, from 210~270%-d.b. and 180~230%-d.b. of the initial water content, respectively. To evaluate the effective diffusivity (*De*) of water species, each sample was placed in a stainless steel net tray (4 meshes) that was mechanically hung from a strain gage transducer in a dryer. To evaluate the dehydration rate, the sample weight was continuously recorded by the output of a strain-gage transducer using a data-logger.

To evaluate the correlation time (τ_C) of the water species for the samples, a nuclear magnetic resonance (NMR) technique was applied to measure the ^{1}H-NMR spectra and spin-spin relaxation time (T_2) of water protons. All the samples cut into 2×2×10 mm pieces were inserted into an NMR sample tube (4mm in inner diameter and 180mm in length). ^{1}H-NMR spectra

were obtained using a JEOL A-500 FT-NMR spectrometer operating at 500MHz for protons. The observed frequency width was 20 kHz. The 90º pulse width was 12.5 μs, and the number of pulse repetitions was 8. The proton chemical shifts were measured by using a small amount of water containing deuterium oxide as an external reference. Except the forced cyclic temperature change operation (FCTCO), all the NMR measurements were performed at 23.5±0.5ºC. The spin-spin relaxation times, T_2, were obtained by the spin-locking method.

B. Evaluation of the correlation time (τ_C) of water proton

The spin-locking pulse technique used was effective to detect a very fast relaxation signal at a low water content. For the evaluation of the relaxation time, T_2 , the equation of $M_t = M_0\ exp(-ts/T_2)$ was used, where M_t is the magnitude of the magnetization vector after the spin locking pulse, M_0 is the magnitude of the macroscopic equilibrium magnetization vector, and *ts* is the spin locking pulse length. In the present study, the plot of $ln\ [M_t / M_0]$ vs. *ts* indicated a good linearity (which demonstrates a correlation coefficient higher than 0.99) through all water contents of the foods, suggesting that the evaluated T_2 value was reasonable. From T_2, the correlation time of a water proton, τ_C , was evaluated using Equation (3) as described by Abragam [15]:

$$\frac{1}{T_2}=\frac{\gamma^4\cdot\hbar^2\cdot I(I+1)}{5r^6}\left(3\tau_c+\frac{5\tau_c}{1+\omega_0^{\ 2}\cdot\tau_c^{\ 2}}+\frac{2\tau_c}{1+4\omega_0^{\ 2}\cdot\tau_c^{\ 2}}\right)\ , \qquad (1)$$

where γ is the gyromagnetic ratio of a proton (= 2.675×10^8 $rad\cdot T^{-1}\cdot s^{-1}$), $\hbar$ is the modified Plank's constant (6.63×10^{-34} J· s), I is the nuclear-spin quantum number of a water proton (= 0.5), r is the proton-proton distance of a water molecule (0.16 nm), ω_0 is the resonance frequency of NMR (= 3.14×10^9 s^{-1}), and τ_C is the correlation time of a water proton (s).

C. Forced cyclic temperature change operation (FCTCO)

To clearly distinguish the mode for the self-organization of water species among the foods used, a forced cyclic temperature change operation (FCTCO) was effectively conducted. The time schedule for the FCTCO was as follows: when the temperature was gradually decreased, a steep reduction of $1/\tau_C$ at a specified temperature (t_1) appeared, and, at the peak bottom of the $1/\tau_C$, when the temperature was gradually increased, the locus of the $1/\tau_C$ showed a typical hysteresis reaching the temperature (t_2) previously used. The FCTCO was carefully repeated between t_1 and t_2, and a forced oscillation of the $1/\tau_C$ was then clearly visualized. In the course of the FCTCO, as an expedient way, 2 min to increase or decrease the temperature and 13 min to evaluate the T_2 as the time for the operation of the NMR equipment were required; the total time of 15 min was thus needed as the measuring time at each temperature in the hysteresis period. The oscillating curves obtained were demonstrated as a function of the elapsed time including the 15 min.

III. Results and Discussion

A. Characterization of the water species due to De (the three-dimensional mobility: 3D mobility, m^2/s)

As has been demonstrated by a large number of researchers [3, 10], the three-dimensional mobility (3D mobility) of water species retained in the foods was evaluated by using an effective diffusion coefficient (*De*, m^2/h). The value of *De* at a given water content could be evaluated by using Equation (2).

$$\frac{W-We}{W_D-We}=\left(\frac{8}{\pi^2}\right)^3\cdot exp\left(\frac{\pi^2\cdot De\cdot t}{4}\cdot\left(L_a^{\ 2}+L_b^{\ 2}+L_c^{\ 2}\right)\right) \qquad (2)$$

where W is the water content at the drying time t (%-d.b.), W_e is the equilibrium water content (%-d.b.), W_D is the initial water content of the rectangular sample of drying flesh (%-d.b.), t is the drying time (s), and L_a , L_b, and L_c are the half distances of the rectangular sample (m).

Since *De* is strongly influenced by the water content of food, it should be evaluated as a function of the water content (W_0). The water content of food consequently contributes to the correlation time (τ_C) evaluated from the proton NMR technique and could reasonably be replaced by τ_C instead of the water content [16].

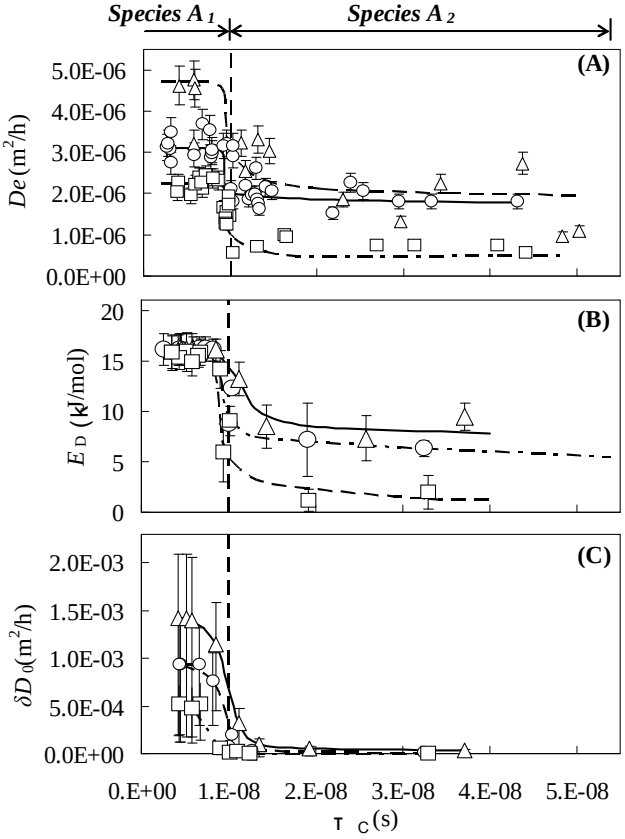

Fig. 1 De, ED, and δ D0 as a function of τ C .Key: ○, BA; △, BH; □, PH

The value of τ_C means a restriction in the strength of water species due to the surrounding macro-molecule as protein. The

De described as a function of τ_C provides information as to how it will be influenced by τ_C. Fig. 1(A) illustrates *De* for B_A, B_H, and P_H as a function of τ_C. As is evident from the results, two water species A_1 and A_2 regions were clearly distinguished at $\tau_C = 10^{-8}$s (designated as a critical τ_C, $C\tau_C$) indicating a stepwise reduction of *De* and an identical dependency as a function of τ_C within each of the two regions. This anomalous change of *De* for the three foods at the $C\tau_C$ strongly supports the idea of a physical change of the food structure derived from the progress of dehydration, such as porosity (ε) and labyrinth factor (χ) of food solids. This presumption would also be supported by the following discussion.

As is well known, *De* can be expressed as a function of the diffusibility (δ) and activation energy (E_D) of diffusion by Equation (3).

$$De = \left(\frac{\varepsilon}{\chi}\right)\cdot D = \delta \cdot D_0 \cdot exp\left[\frac{-E_D}{R\cdot(T_D+273)}\right] \quad , \qquad (3)$$

where ε is the porosity of solid food, χ is the labyrinth factor of food, δ is the diffusibility ($=\varepsilon/\chi$), and D_0 is the pre-exponential factor of the diffusion coefficient (D).

Although the Arrhenius plot of *De* evaluated from the data obtained at 30, 40, and 50°C was widely scattered, the activation energy (E_D) of *De* can roughly be evaluated as a function of τ_C, as shown in Fig. 1(B). The obtained values of E_D again demonstrate a drastic reduction at the $C\tau_C$ as a function of τ_C, indicating an identical value of 16 (±1.5) kJ/mol within the water species A_1 region independently of the kind of foods; on the other hand, in the water species A_2 region, the evaluation was different depending on the kind of foods, i.e., 6.1 (±0.6) kJ/mol for B_A, 8.1 (±0.4) kJ/mol for B_H, and 2.0 (±0.8) kJ/mol for P_H, indicating no change depending on τ_C .

Focusing on Eq. (3), the pre-exponential factor (δD_0) can easily be evaluated by using the extrapolation of the Arrhenius plot line against the perpendicular axis, and the values obtained are plotted as a function of τ_C, as shown in Fig. 1(C). The obtained value of δD_0 clearly demonstrated a remarkable reduction at $C\tau_C$. From this anomalous change of δD_0, the values of ε and D_0 can be presumed to decrease and those of χ to steeply increase because of the progress of dehydration. The drastic change of the three parameters (ε, D_0, and χ) indicates that there is a steep change in the physical structure of the solid food influencing the reduction of *De* in the water species A_2 region, as seen in Fig. 1(A). In this region, regarding the significance of the reduction of E_D, it may be presumed that the energy barrier for water species A_2's diffusion is reduced due to the reduction of the value of D_0 in the hydration water. On the basis of this presumption, the following discussion may be offered.

In a heterogeneous catalysis [17], as an empirical principle, the compensation effect between the pre-exponential factor (A_0) and activation energy (E) for the rate constant in a heterogeneously catalyzed reaction has been widely accepted. The linear relation between the log A_0 and E gives another straight line when the reaction occurs on a different active center of the catalyst surfaces associated with some difference in the reaction mechanism. This empirical principle is possibly applied to the water diffusion process in the foods. Fig. 2 demonstrates the ln (δD_0) as a function of E_D for P_H, B_A, squid, salmon, and sardine. Although the data obtained includes an appreciable number of experimental errors, except for the case of P_H and B_A, each straight line among the three foods clearly gives a different tendency. This evidence strongly demonstrates that the diffusion mechanism of water is different among the three foods. Since D_0 is possibly associated with the nature of the adsorption sites for the water species, the protein structure, as an adsorption site, involved in the foods strongly influenced the value of D_0. The difference among the straight lines of the $\ln(\delta D_0) \sim E_D$ obtained for the five foods in Fig. 2, therefore, strongly suggests that D_0 changes depending on the foods. Although it is difficult to describe the physical meaning of the variation of D_0, it may be presumed that the number of adsorbed water molecules and the structure of the hydration water on the adsorption sites derived from the mechanical structure change of the food solid will change depending on the foods, as discussed in the above section using Fig. 1(C).

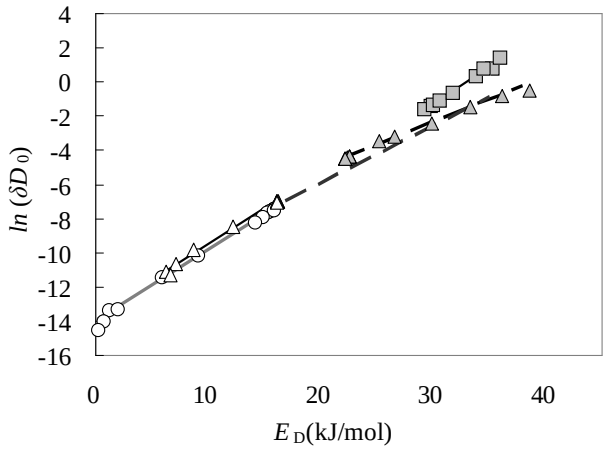

Fig. 2 Compensation effect derived from the ln (δ D0) as a function of ED. Key: ○, PH ; △, BA ; ●, squid ; ▲, salmon ; ■, sardine.

B. Characterization of the water species due to the molecular mobility ($1/\tau_C$, s^{-1})

As has been described in the above section, the molecular mobility ($1/\tau_C$, s^{-1}) can be easily evaluated by using the proton NMR technique. The value of $1/\tau_C$ obtained for all the foods demonstrates the self-organization of the water species retained in the food, which was caused by a programmed reduction in temperature. Fig. 3 demonstrates the $\ln(1/\tau_C)$ as a function of $1/T$ for the scallop as a typical example. The self-organization of the water species is clearly evident at a specified temperature, which influences the water content of the sample (this can be replaced by the initial correlation time, τ_C, of the food sample). Since the molecular mobility ($1/\tau_C$, s^{-1}) is the rotation rate of the water proton, the $1/\tau_C$ obtained at the steeply reduced value can be plotted as a function of $1/T$. The Arrhenius plots obtained for various initial τ_C of the food samples evaluate *E*so(I) = 29.5

(±3.0) for the water species A_1 and *E*so(II) = 57.6 (±2.6) kJ/mol for the water species A_2. All the values of *E*so (I) and *E*so (II) for the six foods were evaluated as presented in Table I. Each of the *E*so obtained gave a characteristic value depending on the kind of foods, indicating the self-organization of the water species to be influenced by different environmental conditions due to the macromolecules of each type of food, such as proteins.

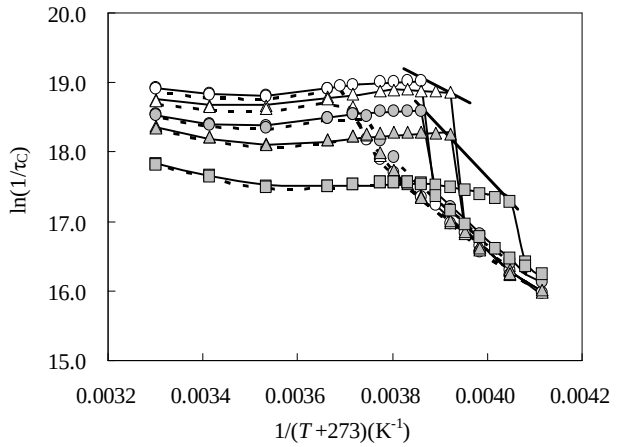

Fig. 3 Evaluation of the activation energy for the self-organization of scallop derived from the ln(1/ τ_C) as a function of $1/T$. W_0 and the initial τ_C are respectively evaluated as the Key: ○, 197%-d.b., 6.53×10^{-9}s ; ∆, 162%-d.b., 7.58×10^{-9}s ;●, 126%-d.b., 1.01×10^{-8}s ; ▲, 104%-d.b., 1.23×10^{-8}s ;■, 64%-d.b., 4.37×10^{-8}s.

TABLE I
ACTIVATION ENERGIES OF SELF-ORGANIZATION FOR WATER SPECIES A_1 AND A_2 RETAINED IN B_A, B_H, P_H, SQUID, SALMON, SARDINE, AND SCALLOP

	E_{SO} (kJ/mol)	
	Species A_1	Species A_2
B_A	31	42
P_H	49	98
Squid	65	67
Salmon	160	63
Sardine	183	116
Scallop	30	58

Focusing on the character difference of the self-organization between water species A_1 and A_2 , each of which produces a different activation energy, it is noteworthy that the two water species can be identified from their molecular states associated with the pre-exponential factor, As_0, in the Arrhenius equation, ln ($1/\tau_C$)= As_0 exp (-*E*so/*RT*). To respond to a question how the adsorption sites for the two water species are discriminated, the compensation effect would be considered as discussed in Fig.1(C). Fig. 4 demonstrates ln (As_0) as a function of *E*so for squid, salmon, sardine, P_H, and B_H. All the data for each food agreed, indicating that an identical compensation effect was in effect between the ln (As_0) and *E*so. This result strongly demonstrates that there is no difference in the water species as hydration water among the foods because of the similarity of the hydration water even though water species A_1 and A_2 can be distinguished by the difference in their self-organization temperature. This evidence differs from the compensation effect between the δD_0 and E_D for the 3D mobility (*De*), indicating a clear difference among the foods evaluated at 30~70ºC, as described in Fig. 2. From these results, it is evident that the self-organization of the water species occurred as a result of an identical physical mechanism that was independent of the difference between water species A_1 and A_2 and among the foods.

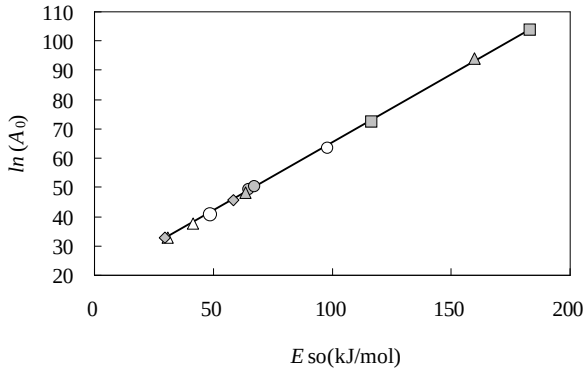

Fig. 4 Compensation effect derived from the ln(A_0) as a function of E_{SO} . Key: ○, P_H ; ∆, B_A ; ●,squid ;▲, salmon ; ■,sardine ;♦, scallop.

C. Visualization of the forced oscillation of the molecular mobility ($1/\tau_C$, s^{-1})

Focusing on the hysteresis behavior of the scallop in Fig. 3, the forced oscillating behavior of the molecular mobility ($1/\tau_C$) can be easily visualized by using a forced cyclic temperature change operation (FCTCO) between two specified temperatures (designated as t_1 and t_2) at which the hysteresis behavior of $1/\tau_C$ appears. Fig. 5 demonstrates a schematic explanation for the FCTCO. The temperature was cyclically scheduled by decreasing and increasing t_1 and t_2, respectively. To evaluate the value of τ_C using the proton NMR spectra obtained at the given temperature, about 15min was needed for the temperature to

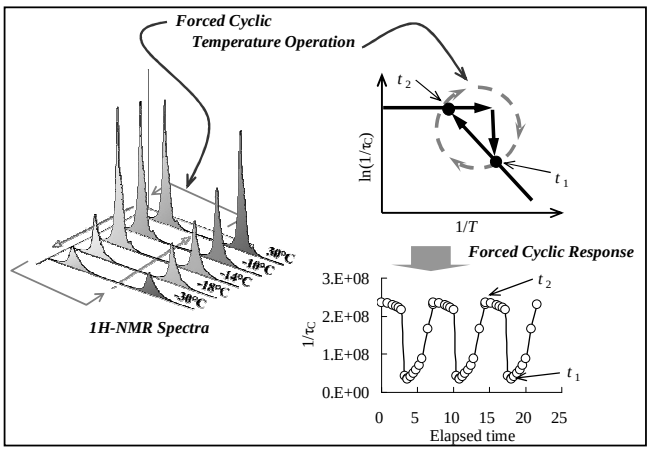

Fig. 5 Schematic visualization of the forced cyclic temperature change operation (FCTCO), the NMR-spectra dynamism, and the $1/\tau_C$-oscillation obtained

decrease and increase and for the evaluation of T_2. The values of $1/\tau_C$ obtained at each temperature were visualized while taking the 15min into the elapsed time as an oscillating curve. The oscillating curve obtained was visualized as a function of time, as demonstrated in the figure.

The forced oscillations of the $1/\tau_C$ obtained were visualized for all the foods taken in this study. Fig. 6 demonstrates the forced oscillating curves for sardine, scallop, and P_H as typical examples. As is evident in the oscillating response curves obtained, the amplitude, period, and waveform were changed depending on the kind of foods and the water content, which can be replaced by the initial τ_C of the sample. The amplitude (α) is drastically reduced in the water species A_2 region, indicating the reduction of the molecular mobility ($1/\tau_C$). This reduction of α, in the case of scallop, can be easily recognized from the diagram in Fig. 7 by demonstrating a width of $\ln(1/\tau_C)$ given as a distance

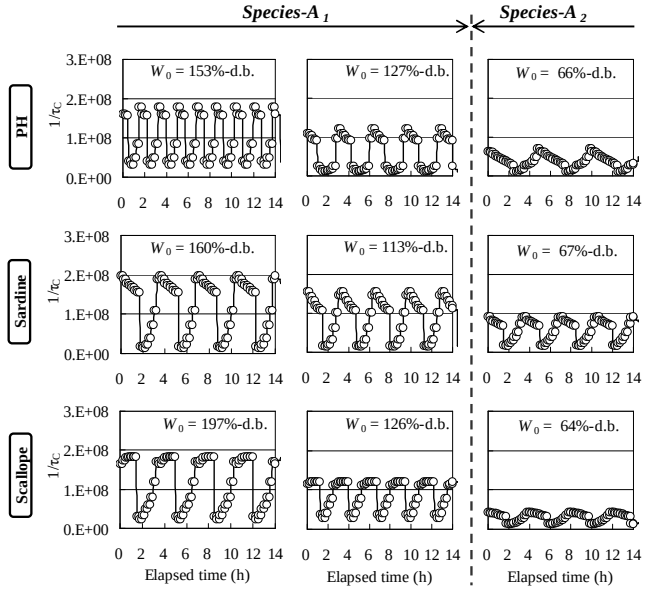

Fig. 6 Oscillating dynamism characterized by the kind of foods

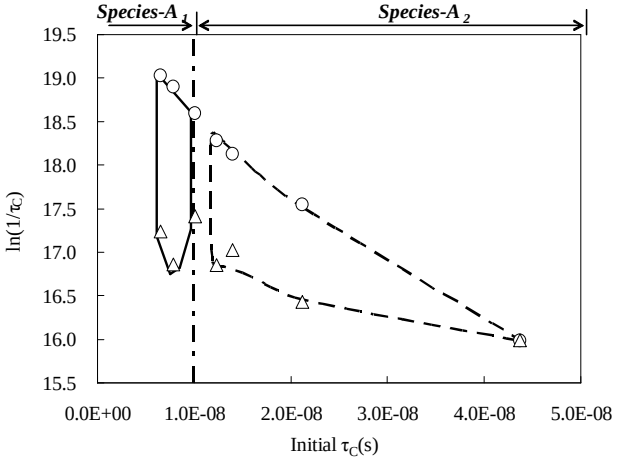

Fig. 7 Width of $\ln(1/\tau_C)$ for the forced oscillation as a function of the initial τ_C for scallop

between the peak top and peak bottom of the oscillating curves in Fig. 6. Since the α is demonstrated as a function of the initial τ_C of the sample, as shown in Fig. 8, the forced oscillations for squid, sardine, and scallop disappeared at the initial τ_C $=3.1\times10^{-8}$, 6.9×10^{-8}, and 4.4×10^{-8} s, respectively. In addition, since the slopes of the $\alpha\sim\tau_C$ straight lines in the species A_2 region gave a specified value depending on the food, designated as $\Delta\alpha\sim\tau_C$, the $\Delta\alpha\sim\tau_C$ obtained for each of the foods can be used to visualize the difference among the characteristic modes of the foods, as is presented in Fig. 11.

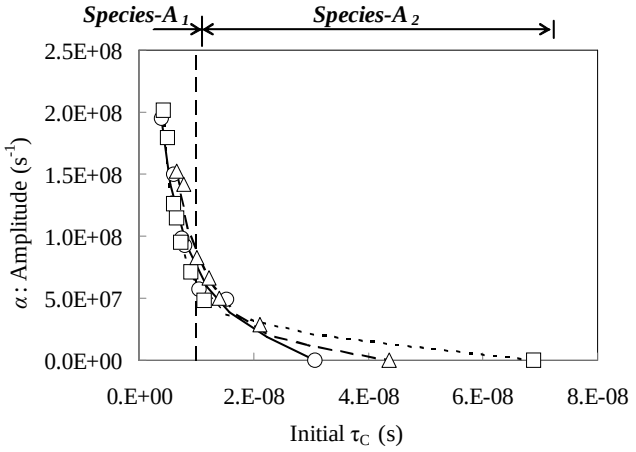

Fig. 8 α as a function of the initial τ_C . Key: ○, squid ; □, scallop ; □, sardine

D.Visualization for the hysteresis behavior of the self-organized water amount (ASOW)

The amount of the self-organized water species (ASOW) can be evaluated by an examination of the graphical integration of the proton NMR spectrum presented in Fig. 5 and expressed as a modified water content (MWC, %-d.b.). The MWC was evaluated by using the initial water content of the sample food corresponding to the peak area of the proton NMR spectra. Fig. 9 demonstrates the MWC as a function of $1/T$ and the initial τ_C. It is clearly evident that the hysteresis of the MCW is similar to the $1/\tau_C$ hysteresis presented in Fig. 3, indicating the steep reduction of MWC at the same temperature as the $1/\tau_C$.

Since the reduced amount of the MWC clearly indicates the ASOW, the amount is linearly reduced by increasing the initial τ_C of the sample. Fig. 10 demonstrates, as an example, the ASOW as a function of the initial τ_C for scallop. All the other foods showed similar results. Based on the results obtained, the linear dependency of the ASOW~τ_C straight line indicates a inflection point at τ_C $=10^{-8}$s, pointing again to the existence of two water species as water species A_1 and A_2, as shown in Fig. 1.

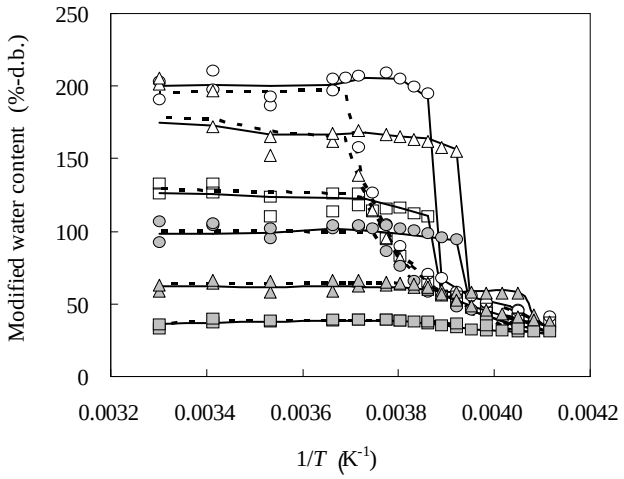

Fig. 9 The converted water content for scallop as a function of 1/*T* and the initial τ_C . Key: ○, 6.5×10^{-9}s ; □, 7.9×10^{-9}s ; □, 1.0×10^{-9}s ; ●, 1.2×10^{-8}s ; ▲, 2.1×10^{-9}s ; ■, 4.4×10^{-9}s.

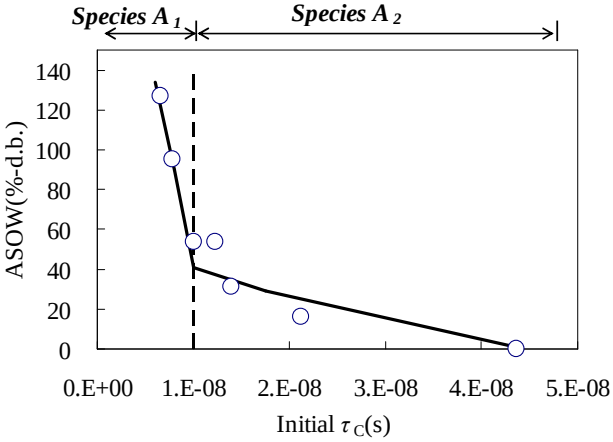

Fig. 10 ASOW as a function of the initial τ_C for scallop.

In the water species A_2 region in Fig. 10, the slope of the ASOW~τ_C straight line clearly changes depending on the food used. The slope may be designated as ΔASOW~τ_C. The values of the ΔASOW~τ_C for P_H, B_A, squid, salmon, sardine, and scallop are evaluated to be -1.15×10^{9}, -8.19×10^{8}, -1.41×10^{9}, -5.49×10^{9}, -4.61×10^{9}, and -1.18×10^{9} %-d.b./s, respectively. The difference of the ΔASOW~τ_C among the six foods would be recognized as the difference in the acceleration of the self-organization of water species in the foods, strongly associated with the α's in Fig. 6. It may be presumed that the ΔASOW~τ_C is larger with a higher α. This assumption can be visualized by Fig. 11. Fig. 11 demonstrates a three-dimensional plot of $\ln(1/\tau_C)$ as a function of the initial τ_C and the ΔASOW~τ_C for P_H, squid, and salmon as typical foods. The α can be expected to sensitively change depending on the kind of food, indicating a characteristic behavior. These results show that the ASOW strongly contributes to the mode of the oscillation wave and α. From these results, the mode of the oscillating width can be used as the difference of the water species in the foods indicating a characteristic dynamism of the hydration water depending on the value of the initial τ_C.

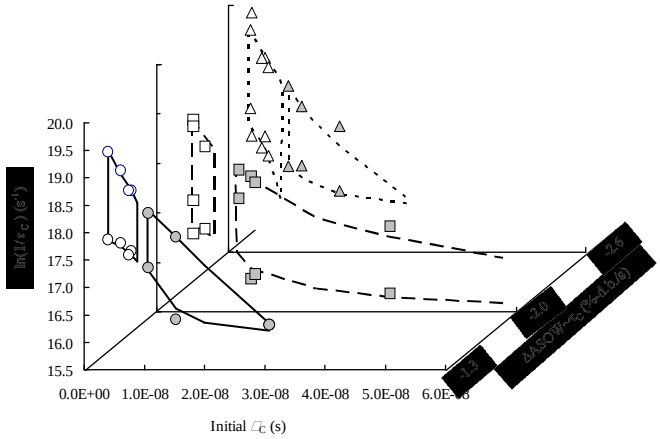

Fig. 11 Three-dimensional visualization for the forced oscillation of the molecular mobility as a function of the ΔASOW~τ_C and the initial τ_C . Key in the A_1 region: ○, squid; △, salmon; □, P_H ; and Key in the A_2 region : ●, squid; ▲, salmon; ■, P_H .

IV. Conclusions

The water species retained in the six foods was clearly separated into two categories as species A_1 and A_2. The two species were characterized by two points of view, the effective water diffusivity (*De*, three-dimensional mobility, m^2/s) and the molecular mobility ($1/\tau_C$, reciprocal correlation time, s^{-1}). Based on the experimental evidence obtained, the following conclusions were demonstrated.

(1) *De* and the activation energy (E_D) of *De* were drastically changed at Cτ_C, at which the water species was divided into A_1 and A_2 in all the foods. This anomalous change was understood as a remarkable change in the physical structure of the food solids derived from the progress of the dehydration. This change brought about a change in a diffusion mechanism.

(2) The molecular mobility ($1/\tau_C$) in all the foods clearly demonstrated a characteristic hysteresis at a specified temperature lower than 0ºC, indicating a self-organization of the water species (SOW). The hysteresis of the SOW visualized a characteristic forced oscillation derived from the forced cyclic temperature change operation (FCTCO) indicating a reproducibility of the SOW.

(3) The obtained forced oscillation of the $1/\tau_C$ clearly demonstrated a characteristic amplitude (α), period, frequency, and oscillation wave depending on the kind of foods and the two water species. The α demonstrated a drastic reduction at Cτ_C.

(4) The ASOW was quantitatively evaluated by using the peak area of the proton NMR spectrum and determined as a function of the initial τ_C of the sample foods. The amount was changed depending on the kind of foods and understood as the difference of the hydration water amount retained in the foods.

(5) The oscillating behavior of $1/\tau_C$ clearly demonstrated as a three-dimensional visualization using two parameters, the initial τ_C and the ΔASOW~τ_C. The results interestingly visualized the

water species to be clearly divided into two water species and to have a variety of oscillations depending on the foods.

NOMENCLATURES

A_0 *pre-exponential factor of k (kJ/mol)*
As_0 *pre-exponential factor of the molecular mobility* $1/\tau_C$ (kJ/mol)
ASOW *the amount of the self-organized water* (%-d.b.)
$\Delta ASOW \sim \tau_C$ *the slope of the ASOW~*τ_C *straight line* (%-d.b./s)
B_A *beef meat produced in Australia* (-)
B_H *beef meat produced in Hokkaido, Japan* (-)
D *water diffusion coefficient* (m^2/h)
D_0 *frequency factor of D* (m^2/h)
De *effective water diffusion coefficient* (m^2/h)
De^0 *pre-exponential factor of De* ($=\delta \cdot D_0$, m^2/h)
E *activation energy of the heterogeneous catalysis* (kJ/mol)
E_D *activation energy of water diffusivity* (kJ/mol)
*E*so *activation energy of the self-organization* (kJ/mol)
FCTCO *forced cyclic temperature change operation* (-)
I *nuclear spin quantum number of water proton* (=0.5) (-)
L_a *half distance of a-axis of the rectangular sample* (m)
L_b *half distance of b-axis of the rectangular sample* (m)
L_c *half distance of c-axis of the rectangular sample* (m)
M_t *magnitude of magnetization vector* (-)
M_0 *magnitude of macroscopic equilibrium magnetisation vector* (-)
MWC *modified water content* (%-d.b.)
P_H *pork meat produced in Hokkaido, Japan* (-)
R *gas constant* (=8.314J/K·mol)
r *proton-proton distance of water molecule* (= 0.16 nm)
SOW *self-organization of the water species* (-)
ΔS *activation entropy* (kJ/K·mol)
T *temperature at the time to evaluate* T_2 (K)
T_2 *spin-spin relaxation time of water proton* (s)
T_D *drying temperature* (°C)
t *drying time* (s)
t_1 *the initiation temperature of the self-organization of water species in the course of the FCTCO* (°C)
t_2 *the returning temperature of the self-organization of water species in the course of the FCTCO* (°C)
ts *spin locking pulse length* (s)
W *water content at the drying time t* (%-d.b.)
W_0 *initial water content for the given sample* (%-d.b.)
W_D *initial water content of drying flesh sample* (%-d.b.)
We *equilibrium water content* (%-d.b.)

Greek letters

α *amplitude of the forced oscillation response* (-)
ε *porosity of the food tissue* (-)
π *the ratio of the circumference of a circle to its diameter* (=3.14)
γ *gyromagnetic ratio of proton* ($=2.675\times10^8$ rad·T^{-1}·s^{-1})
$\hbar$ *modified Plank's constant* ($=6.63\times10^{-34}$ J·s)
ω_0 *resonance frequency* ($=3.14\times10^9$ s^{-1})
τ_C *correlation time of water proton* (s)
$C\tau_C$ *critical correlation time of water proton* (s)
χ *labyrinth factor of the food tissue* (-)
δ *diffusibility* ($=\varepsilon/\chi$) (-)

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