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The Structure of Water Induced by Specific Ceramics Treatment and Effects of Treated Water in View of Some Biological Aspects

Kazuishi SATO, Mariko AGO, Katsumi ISHIKAWA*, Tsuneo SATO** and Kunihiko OKAJIMA***

Department of Environmental and Systems Engineering, Faculty of Engineering,
Tokushima Bunri University, 1314–1 Shido, Sanuki, Kagawa 769–2193, Japan
** Faculty of Agriculture, Kochi University, B 200 Monobe, Nankoku, Kochi 783–8502, Japan*
*** Central Research Laboratories, Asahi KASEI Corporation, 2–1 Samejima, Fuji, Shizuoka 416–8501, Japan*

**** Department of Nano Material and Bio Engineering, Faculty of Engineering, Tokushima Bunri University,*
1314–1 Shido, Sanuki, Kagawa 769–2193, Japan

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The structure of water induced by a high temperature sintered kaolinite ceramics treatment was studied in view of thermally stimulated depolarized current (J) and temperature profiles (TSDC analysis) on the iced treated water and ^{1}H Nuclear Magnetic Resonance (NMR) spectrometry on the treated water as well as some physical properties of the sintered ceramics. In addition, some biological effects of the treated water were examined to give indirect evidences for the proposed water structure. The treated water were positively charged based on essentially the formation of electric double layer interface between the ceramics surface and water flow, without any absorption desorption phenomena to substances contained in water to be treated by the ceramics. The positive charge was indirectly affirmed by TSDC and ^{1}H NMR analyses on ion exchanged water and for the drinking water. The water structure derived by the existence of chlorine substances was apparently disappeared from ^{1}H NMR spectrum, probably by the apparent neutralization or accumulation of chlorine substances through interaction with positive charge. This phenomenon seemed to be closely related with the fact that absorption of chlorine ion through a vegetable root was greatly depressed when the treated water by the ceramics was used. The treated water was also proved to restrict the growth of *E. coli* in poor nutritional culture media and the production of bacterial cellulose by CFJ 002 (New bacterium cellulose producing microorganism, an enterobacter) stock, and to suppress the oxidation action of hydrogen peroxide.

Keywords : anti oxidation, electrokinetic treated waters, ceramics, *E. coli*, positive charge

INTRODUCTION

Authors have already reported the results on plant cultivation (Ishikawa et al., 1996; Tamura et al., 1999; Tamura et al., 1999; Abul et al., 2003) and agricultural production (Tanabe et al., 1998) by using a water treatment apparatus. The structure of the apparatus is simple cylinder in which kaolinite granular ceramics with average particles size 1–2 mm were packed in half volume

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Corresponding author : Kazuishi Sato, fax : ＋81–87–894–4201,
e mail : ksato@fe.bunri u.ac.jp

of the cylinder on flow controller support, and the water to be treated is passed though from the bottom inlet to the top outlet. The ceramics particles are floated up with mutual collision by water flow. The water treatment itself is quite simple, economical and quite effective at short times, compared with other outer force treatment such as magnetic field treatment, electro static field treatment, ultrasonic treatment, micro wave treatment and so on. The above apparatus has also been applied in a dialysate fluid production for blood dialysis, where the apparatus is placed before reverse-osmisis (RO) procedure, revealing that the duration of RO membranes is drastically prolonged and the obtained dialysate fluid contains practically no endotoxin (Nomizo, 2004). In this way, the water treated with the ceramics has some specific function in comparison with usual water. However, a detailed discussion on the water structure induced by the ceramics treatment has not done yet.

Recently, the method for deducting some water structure (Sato et al., 2004; Sato et al., 2005) has been proposed by the authors, which is based on TSDC analysis on several iced waters. The TSDC analysis revealed that 1) 5 major relaxation peaks (A to E from lower temperature side) representing the water motions formed by different strength or number of hydrogen bonds (dipole-dipole interactions) were detected at ca. −150 to −20°C, and 2) A peak shifted to higher temperature side by adding so-called positive salvation ions (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Li^{+}) and shifted to lower temperature side for drinking water which contains chlorine ion (negative salvation ion), with quite good reproducibility of the date.

In this paper, we will estimate the state of treated water obtained by the ceramics treatment with aid of TSDC method and ^{1}H-NMR analyses and will show the some evidences for the proposed water structure in view of biological and botanical aspects.

MATERIALS AND METHODS

Water samples

A pure water was prepared by using an apparatus (Mill-Q Academic A10 model, Millipore Inc., USA). An ion-exchanged water prepared by a Shimadzu apparatus (SWAC-520) and drinking waters supplied in Fuji city (Factory in Asahi KASEI Corporation) and Nankoku city (Kochi University) were utilized.

The treatment of water by specific ceramics

A water treatment apparatus (see Fig. 1) and the ceramics particles (globular with diameter of ca. 2 mm) were both supplied from Nishijima, Tatsuhisa (Pursanova-Japan). The water was treated in two ways: Into a cleaned PET bottle with 1 liter content containing 20 g of the sintered ceramics, waters to be treated was poured, then shaken 10 times in an action of up-side-down of the bottle (batch method). The treated water was subjected to ^{1}H-NMR analysis. Using an apparatus shown

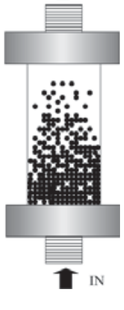


Fig. 1 Water treatment device using granular ceramics.

Table 1 Characteristics of granular ceramics.

Compositions (wt%)				Specific surface (m^2/g)		Apparent specific gravity
quartz	anorthite	alumina	mullite			
SiO_2	$CaAl_2Si_2O_8$	Al_2O_3	$Al_6Si_2O_{13}$	surface	inside	3.2
57	19	12	12	0.47	0.75	

in Fig. 1, the water stocked in a stock vessel were circulating 3 times at the rate of 3 L min^{-1} (consecutive method). The treated ion exchanged water was used as medium for oxidation reaction of hydrogen peroxide and cultivation of *E. coli*, as well as original water. The treated drinking water from Fuji city and its original one were also subjected to ICP mass analysis. Using a similar water treatment apparatus, the treated drinking water from Nankoku city was obtained with one pass treatment, subjecting to botanical experiment. During the consecutive treatment, the ceramics are collided with each other probably to give local temperature rise in the surface of the ceramics. It is well known that when water passes on the surface of silica, so called electrical double layer is formed between interfaces of silica and water, giving negative and positive charges, respectively.

Extraction of ionic inorganic substance from the sintered ceramics

A 20 g of the high temperature (1,200°C) kaolinite sintered ceramics (see Table 1 for their some physical properties) was boiled with 200 cm^3 of pure water for 1h and the water used was subjected to ICP mass analysis.

A quantitative analysis of ionic substances

A quantitative analysis of ionic substances (41 elements from Li to Ba, 29 Lantanoid elements) in sample waters was carried out by a ICP mass spectrometer (model X7, Thermoelectron Co., Ltd., USA), using calibration coefficient for In (115) with the blank value for a super ultra pure water (TAMAPURE AA, Tama Chemical Ind. Co., Ltd., Japan). For Si, a flameless atomic absorption method (Atomic absorption photometer Model AA 6800, SHIMADZU Co., Ltd., Japan) was applied using Rh as a modifier.

Wide angle X ray scattering and surface area measurements for sintered ceramics powder

The sample was powdered by crushing the sintered ceramics in a pulverizer. Chemical constitution analysis of the ceramics was made using Wide angle X ray diffractometer RINT 2200 (Rigaku Corporation, Japan) for reflection method and the data analysis was carried out by using the compound retrieval software. Surface area measurement of globular ceramics surface part and central part were measured by the BET method using a type AUTOSORB 1 (Yuasa Ionics Inc., Japan) instrument.

Thermally stimulated depolarized current (TSDC) measurements

TSDC measurements were made using TSC/RMA spectrometer 91000 (Rigaku Corporation / Solomat) on water samples aligned between electrodes in He gas filled sample chamber. Global measurements were carried out according to the procedure described in the previous study (Sato et al., 2005). Since the equilibrium molecular motions attained at polarization temperature Tp was abruptly cooled down to below -160°C, Tp was chosen at 20°C here.

^1H NMR measurement

Each one drop of the treated distilled water (batch method) and corresponding original water was put into heavy water with TMS (0 ppm) as standard and they measured by a NMR spectrometer (Model JNM ECA400, JEOL, Japan) under the following operating conditions: single pulse width, 5.6 μs (45°); pulse waiting time, 5.0 s; accumulation, 8; and temperature, 25°C. The reproducibility of the data was ascertained by 5 times measurements. For the treated drinking water (batch method) and corresponding original water, samples were put into inner sample tube with outer tube containing TMS/d3 chloroform as an outer standard. The measurement was carried out under the following conditions: single pulse width, 5.0 μs (45°); pulse waiting time, 2.90 s;

accumulation, 100; and temperature, 35°C.

Antioxidation assessment

According to the procedure of antioxidant effect proposed by Takehara and Takashima (Takehara and Takashima, 2000), for categorizing the several waters treated by outer forces (magnetic field, static electric field and so on), a given amount of hydrogen peroxide was added into a given amount of water (treated and original ones) in glass vessel controlled at 25.0 (±0.1)°C, then the reaction was started by poring a given amount of pottacium ferrocyanide at 25.0(±0.1)°C. Pottacium ferrocyanide is oxidized to ferricyanide giving yellow color development. Absorption at 420 nm was measured by ultraviolet and visible spectrophotometer (Model V570, JASCO, Japan) after 40 min reaction. The samples with desired stock days were also subjected to the above reaction

Growth of E. coli in cultivation media with poor nutrition

Each treated ion exchanged water and its original one was filtered with filter having average pore size of 0.2 μm for bacteria elimination. Into each treated water, a condensed M 9 culture media sterilized before use, thiamine and D glucose were mixed to give culture media composed of 79 wt% treated water, 20 mg/l of thiamine and 20 g of glucose. An *E. coli*, BL21/TB1 stock was inoculated to the culture media and cultured under mild shaking for desired intervals at 37°C. As a function of culture interval the turbidity of the system and the number of colonies were measured in conventional manners. That is, absorption of culture media taken at desired cultivation intervals was measured at 600 nm by ultraviolet and visible spectrophotometer (Model V570, JASCO, Japan) and at the same time a given amount culture media taken was spread over gelatin plate and the number of colonies were measured by colonies counter (Model MCL 1000C, Masuda Sci. Co., Ltd., Japan).

Production of bacterial cellulose by culturing of CFJ 002, an Enterobactor

The bacterial cellulose production experiment using CFJ 002 stock was carried out according to the procedure described in a literature (Shimada, 1995; Ago et al., 2005). The waters used for nutrient media are an ion exchanged water and the treated water, respectively. After each nutrient medium were sterilized with the use of high pressure steam, five hundred ml of that were poured in a two liter flask, respectively. And CFJ 002 were inoculated at concentration of 10^5CFU ml^{-1} on the culture media. Culturing CFJ 002 was carried out initial pH as 7.5 and shaking condition (105 strokes min^{-1}. (29°C, 48 h)).

Absorption of ionic substances from plant roots

Nursery plants of komatsuna (*Brassica campestris* L. rapifera group) of which raising seedling was done in tap (drinking) water were used for the absorption experiment. A cultivation apparatus (longitude 53 cm × transverse 36 cm × depth 19 cm) equipped with a fix planting panel (fix planting pore diameter; 2.5 mm, pore interval; 2.0 × 3.5 cm, pore numbers; 40) in which the nursery plants were set out. The cultivation media was prepared adding MIKADO SUIKOU No. 1 and No. 2 (Mikado Seed Growers Co., Ltd., Chiba, Japan) into tap water so that pH and electro conductivity (EC) of the media become 6.0 and 3.0 dS/m, and used as a reference. The cultivation media was treated by using an apparatus described before for 5 min. These two cultivation media were introduced into clearly identifiable sections of plant panel and exchanged every 1 week. A part of media was sampled in arbitral intervals and absorption of target substances from the plant root was estimated based on material balance.

$$Q_s = Q_w[M]_s + \frac{V(M_{t1} - M_{t2})}{\Delta t} Q_s \qquad (1)$$

Here, Q_s (mg h^{-1}) is the absorption speed of substance M, Q_w(L h^{-1}) is the flux of the supplemental solution, $[M]_s$ (mg L^{-1}) is the concentration of substance M in the supplemental solution, V is the volume of nutrient medium in circulating path, $[M]_{t1}$(mg L^{-1}) is the concentration of nutrient medium in circulating path at t_1, and $[M]_{t2}$ is the concentration of substance M at $t_2(t_1+\Delta t)$.

RESULTS AND DISCUSSION

Table 1 collects the some physical parameters of sintered ceramics (composition of mineral substances, specific surface area, apparent density, solubility). According to the X ray analysis, the ceramics is composed of four crystalline minerals (quartz, corundum, mulite, anorthite) of which weight ratio of quartz, corundum, mulite and anorthite is 57%, 19%, 12% and 12%, respectively. That is, major components of sintered ceramics are oxides of silicon, aluminium and calcium. The specific surface area for the outer most layer and inner part is quite low, being less than 1 $m^2 g^{-1}$ (outer, 0.47; inner, 0.75), which is far lower than that of active carbon (ca.$>$10,000 $m^2 g^{-1}$). This means that water treatment is not done based on absorption desorption of substances contained in waters treated. The ceramics has a gradient structure in view of specific surface area. Any sub stances were not dissolved out from the ceramics even by ion exchanged water reflux treatment (see Table 2). Both Si and Al contents in original and treated waters were practically in a same level, as well as B, Na and K. Similar results were obtained by circulating treatment of drinking water trough the ceramics. These results again suggest that water treatment is not done based on absorption desorption of substances contained in waters treated.

Figure 2 shows TSDC profiles an ion exchanged water and its treated one by the ceramics. TSDC profiles for an ion exchanged water show five major peaks (A–E) below 0°C, indicating that there are plural numbers of molecular state of ice water constituted of different hydrogen bonding. The ceramics treatment resulted in considerable higher temperature shifts for peaks A–E and of these D and E become too strong in depolarized current J, denoting that water molecules are strongly restricted in their molecular motion. The higher temperature shift of peak A has not been observed by other outer force treatments of water, such as direct magnetic field, electro static field,

Table 2 Dissolved elementals in the boiled water with granular ceramics and original pure water.

Chemical symbol	Pure water	Boiled water	Chemical symbol	Pure water	Boiled water	Chemical symbol	Pure water	Boiled water
Li	1.2	1.2	Rb	0.09	0.05	Eu	Nd	Nd
B	640	540	Sr	0.15	0.03	Gd	Nd	Nd
Na	490	470	Y	Nd	Nd	Tb	Nd	Nd
Mg	0.60	0.24	Zr	Nd	Nd	Dy	Nd	Nd
Al	17	15	Nb	0.01	0.01	Ho	Nd	Nd
K	160	150	Mo	0.03	0.02	Er	Nd	Nd
Ca	31	24	Ru	Nd	Nd	Tm	Nd	Nd
Sc	0.18	0.16	Rh	Nd	Nd	Yb	Nd	Nd
Ti	0.37	0.13	Pd	Nd	Nd	Lu	Nd	Nd
V	0.19	0.18	Ag	Nd	Nd	Hf	Nd	Nd
Cr	0.69	0.70	Cd	0.01	0.00	Ta	Nd	Nd
Mn	0.19	0.17	Sn	0.02	0.01	W	0.01	0.01
Fe	29	30	Sb	0.46	0.40	Re	Nd	Nd
Co	0.01	0.01	Te	0.05	0.04	Os	Nd	Nd
Ni	0.28	0.19	I	0.18	0.11	Ir	Nd	Nd
Cu	0.40	0.25	Cs	0.01	0.01	Pt	0.02	0.01
Zu	0.25	0.14	Ba	1.1	0.22	Au	0.01	Nd
Ga	0.05	0.04	La	Nd	Nd	Hg	0.08	0.08
Ge	0.03	0.02	Ce	Nd	Nd	Pb	0.02	0.07
As	0.16	0.20	Pr	Nd	Nd	Bi	Nd	Nd
Se	0.92	1.1	Nd	Nd	Nd	Th	Nd	Nd
Br	14	13	Sm	Nd	Nd	U	0.01	0.01

unit : ppb
Nd : not detected

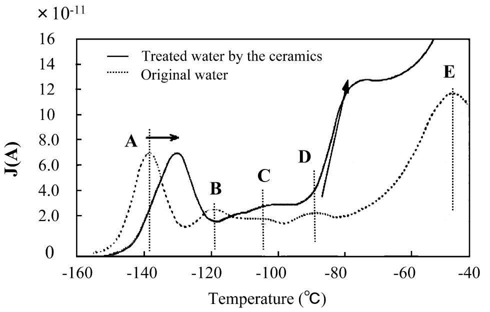


Fig. 2 TSDC spectra corresponding to the water treated with the ceramics and original pure water.

ultrasonic, micro-wave treatments which shift peak A to lower temperature side more or less (Sato et al., 2005). In this sense, the treatment of water by sintered ceramics might be quite different from other treatments in its treating mechanism. In contrast to this, the drinking water gives three peaks (ca. −143°C(A), ca. −118°C(B), ca. −110°C(C)) and a strong upswing at higher temperature than −75°C, owing to the large content of Ca^{2+}, Mg^{2+}, Na^{+} and K^{+} (Sato et al., 2005).

Figure 3 shows TSDC profiles for a Fuji-drinking water and its treated one by the ceramics. The ceramics treatment of the drinking water gives much the same TSDC profile to its original one, but clearly shifted peak C to higher temperature side (3–4°C). This means that the ceramics treatment could gives similar effect on the state of water molecules even if a large amount of cation is present in the system. The TSDC analysis of water is essentially carried out on iced water, therefore it is still questionable whether the results reveal the molecular state of the common water. This feature will be discussed with the present results of ^{1}H-NMR spectra. Since the water treatment by sintered ceramics particles is simple enough just to pass water over the ceramics surface, an electric double layer formation in interface between the ceramics surface and water flow should be considered as one of most probable water treatment mechanisms. In this case, water flow side might be positively charged. In this sense, the present treated water is more preferably named as the electrokinetic treated water (hereafter, referred to as EKT water). If EKT water is positively charged true, the situation might be detected by simple ^{1}H-NMR analysis.

Figure 4a and b show ^{1}H-NMR spectra for the original/EKT ion-exchanged waters and the original /EKT drinking waters. The original ion-exchanged water gives one broad peak at 4.670 ppm with half value width of 3.0×10^{-3} Hz. However, the EKT water clearly shows a lower mag-

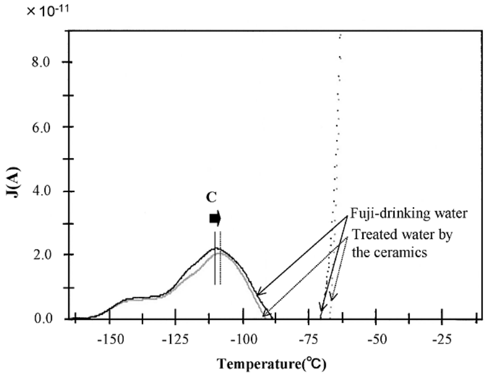
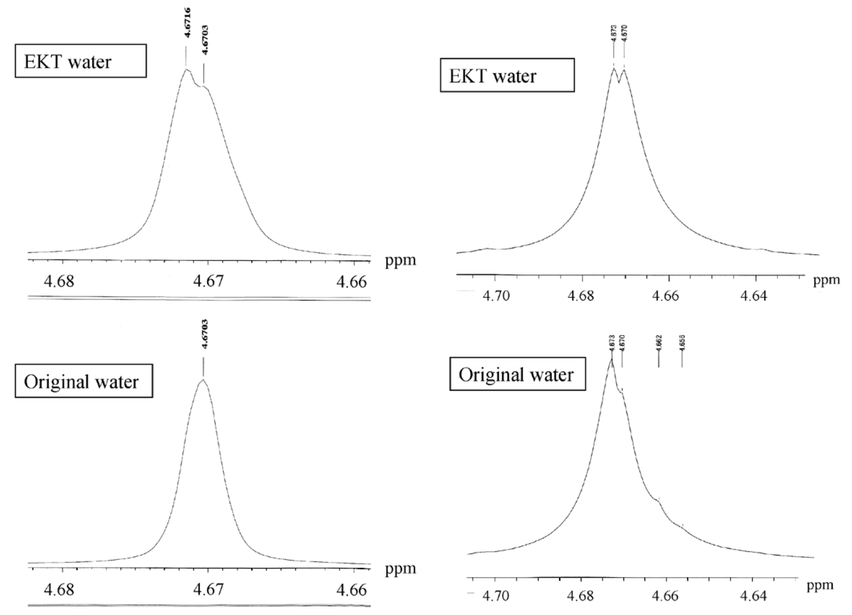


Fig. 3 TSDC spectra corresponding to the water treated with the ceramics and original Fuji-drinking water.



a

b

Fig. 4 a ^{1}H-NMR spectra of the EKT water and original water; ion exchanged water. b ^{1}H-NMR spectra of the EKT water and original water; Fuji-drinking water.

netic field peak at 4.672 ppm besides the original peak. The reproducibility of data was ascertained by repeated measurements (5 times). This result is quite reasonable by assuming that a part of water molecules are positively charged, leading to lower electron density on hydrogen atoms of EKT water, probably though the interaction of positive charges with oxygen atoms in H_2O, as will be shown in Fig. 9. The lower magnetic field shift might also relate to the mutual interaction of water molecules probably through a kind of pseudo hydrogen bond formation, which might restrict the mobility of water molecules against such as temperature rise as shown in TSDC analysis. This means that the results obtained for iced water by TSDC might reflect the common state of water in parts. In contrast to this, ^{1}H NMR for a drinking water reveals a main peak at 4.673 ppm with a shoulder peak at 4.670 ppm (main peak for ion exchanged water) and small but distinct peaks at 4.662, 4.656 ppm. It is astonishing to know that water molecules are distributed in several order of associations without averaging of the association states even at NMR time scale. The main peak might represents the water molecules which are orderly associated through interaction (positive salvation) with cations (Arakawa et al., 1979). It is important to note that 4.673 ppm peak is quite near to that newly observed for the EKT ion exchanged water, which might be positively charged. This means that positively charged state of water molecules behave like the water molecules with cations in view of NMR spectra. The two peaks at higher magnetic field might represent the water molecules which are disorderly reorganized through negative salvation of anions such as chlorine substances. These two peaks apparently disappeared and seemed to be included in the 4.670 ppm peak for the EKT drinking water. This phenomenon could be explained if we again assume that the EKT water is positively charged. That is, the positive charge might neutralize or draw anion (such as chlorine substances, chlorine ion) and deprived the ability of chlorine ions which might be associated each other through interaction with positive charges to apparently give large mass. This might relate to the results on reduced up take by vegetable root, shown later. As a result, the EKT drinking water gives two NMR peaks at 4.673 and 4.670 ppm. Although TSDC analysis on the

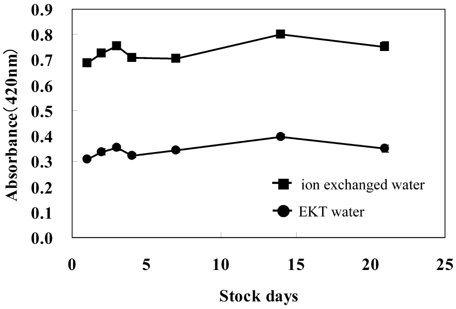


Fig. 5 Antioxidant effect of the EKT water.

EKT ice drinking water still gives higher temperature shifts of peak C, it is not clear whether 4.673 ppm peak include or not the water molecule assembly restricted in its motion by positive charge in water.

The following experimental facts might provide the indirect evidences that the EKT water by sintered ceramics is positively charged. Figure 5 show the antioxidant effect of the EKT ion exchanged water, compared with original water, where absorbance at 420 nm is plotted against the stock days of the waters. The absorbance at 420 nm suggests the degree of oxidation for potassium ferrocyanide to potassium ferricyanide in hydrogen peroxide aqueous solution. When the reaction medium is changed from original ion exchanged water to the EKT one, the degree of oxidation is lowered by ca. 50% and the restriction effect of oxidation lasts at least 30 days after the treating instance. How the oxidation of ferrocyanide is restricted is not clear at present. However, if hydrogen peroxide itself is reduced by pulling out electron to give O_2 having higher oxidation state, as described in literature (Sawyer et al., 1995), the produced O_2 might be captured by positive charge or the hydrogen atom with reduced electron density (see Fig. 5), resulting in the lowering of oxidation ability of peroxide.

Growth of an *E. coli* stock under poor nutrient culture media is considerably reduced when the EKT ion exchange water is used instead of non treated ion exchanged water (see Fig. 6). The reason is not clear at presents, but might relate to the cell structure of Bacteria. The cell is essentially a vesicles structure of which outer membrane is surrounded by phosphate anion extending hydrophobic part into inner, therefore the positively charged water might affect the state of the phosphate of phospholipid. In addition, CFJ 002 stock did not produce bacterial cellulose at all when the

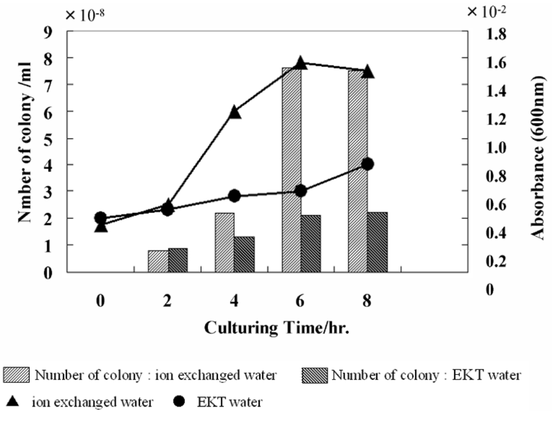


Fig. 6 Breeding of *E. coli* under oligotrophic condition.

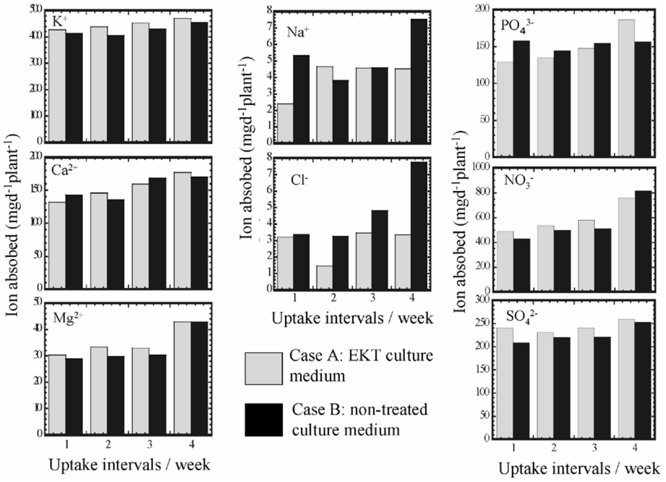


Fig. 7 The uptake of various ions during komatsuna culture.

EKT ion-exchanged water is used. Since CFJ-002 is an aerobic bacterium requiring oxygen for its growth, if oxygen is not taken in effectively by the bacterium, the growth of the bacterium might be reduced. Such circumstance could be realized when the system is positively charged. This is equivalent to the fact that CFJ-002 stock hard to produce bacterial cellulose under acidic condition (Takahashi et al., unpublished result), which is almost the non-aerobic state.

Figure 7 shows the uptake (mg day^{-1} plant^{-1}) of various ions by vegetable roots using the EKT culture (case A) or non-treated culture (case B) media as a function of uptake intervals. This experiment is meaningful since the sintered ceramics treatment is not base on absorption-desorption of substances in water. The uptake of cations are follows: K^+(case A, 1784.3; case B, 1700.9), Ca^{2+}(case A, 614.7; case B, 612.1), Mg^{2+}(case A, 138.9; case B, 131.6), Na^+(case A, 16.0; case B, 21.2). For mono-valent cation, a quite contrast results were obtained for Na^+ and K^+ for which the EKT water restricted the uptake of Na^+ by 24.5% and enhanced the uptake of K^+ by 4.9%. The reason is not clear, but might relate to the difference in the dissolving states of these ions in water. Na^+ is categorized as positive hydration ion (Ohsaka and Uedaira, 1989), but K^+ is said to be a negative hydration ion (Emeliyanov and Valiev, 1964). The treated water might change their hydration states which influence the uptake mechanism by vegetable root. This aspect is far beyond the scope of the present experiment, requiring more essential and detailed study. Similar discussion could be applied for the uptake difference for Mg^{2+} and Ca^{2+}. The uptake of anions are follows: NO_3^-(case A, 2382.23; case B, 2253.3), SO_4^{2-}(case A, 969.9; case B, 901.9), PO_4^{3-}(case A, 596.7; case B, 612.7), Cl^-(case A, 11.5; case B, 19.2). Most conspicuous result is seen for uptake of chlorine ion where the EKT water restrict it as much as by 40%. This phenomenon might be clearly related to the positive charge of EKT water as was described before. As a possible reason, it can be said that the positive charge may gather chlorine ions so as to make large mass and the resultant associate mass could not pass through the plant cell membranes. Inactivation of chlorine substance in drinking water by the ceramics treatment is easily known from the fact that the non-treated drinking water develops dense yellow color by putting orthotolidine but the color is quite pale for the treated drinking water (Okajima et al.,unpublished results). Since the ceramics treatment is not based on absorption-desorption principle as mentioned before, chlorine substances should be inactivated in another way such as neutralization by positive charge and so on.

A quite tentative mechanism for water treatment by sintered ceramics and a tentative water structure deducted from TSDC analysis, ^{1}H-NMR and some biological aspects before and after the

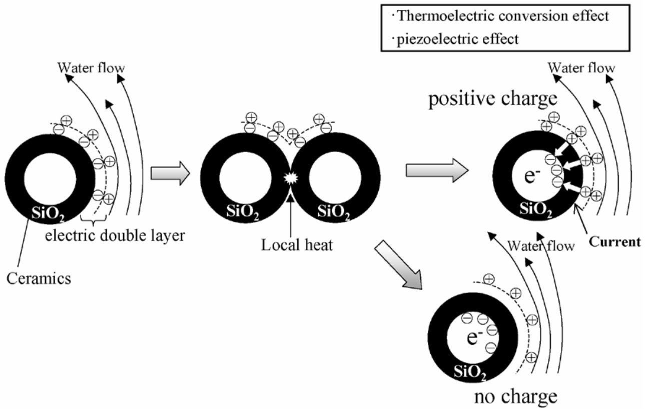


Fig. 8 The water treatment principle by granular ceramics fluidization.

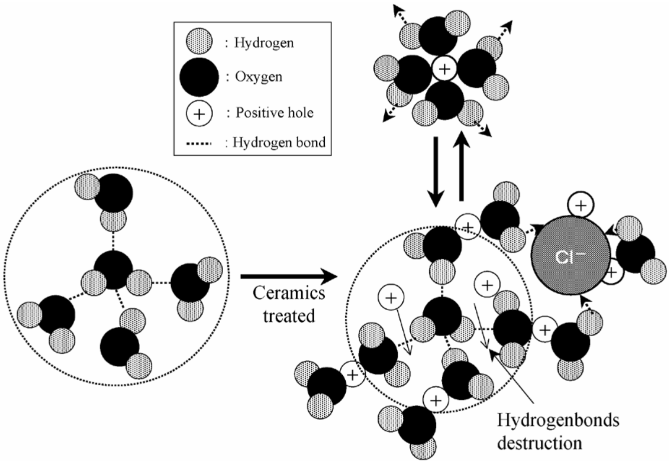


Fig. 9 A structural change of the water presumed from ^{1}H-NMR, TSDC measurement and some biological aspects before and after the ceramics treatment.

ceramics treatment are shown in Figs. 8 and 9. As is well known in colloidal chemistry, silica surface is negatively charged in aqueous silica colloid suspension, requiring pH 1–2.8 for the neutralization. Similar value is ca. pH 9 for alumina colloid. Since major component of the present sintered ceramics is oxide of silicon and the next one is oxide of aluminium, its surface is considered to be also rich in quartz (silica). Therefore, the principle mechanism for the structure change in water resides in the positive charge acceptance through electric double layer formation on interface between silica and water flow, as mentioned before. However, the higher temperature shifts for peaks A–E by as much as 10°C and the strong increase in depolarized current J of peaks D and E could not be understood only by the positive charge flow into water molecules through electric double layer formation. We have confirmed that the far infra red irradiation in water and the boiling of water give only slight higher temperature shift (4°C)(Okajima and Koizumi, 1998). The boiling treatment protonizes water in part giving lower magnetic field peak component on ^{1}H NMR (Okajima and Hisatani, unpublished results). Therefore, we should seek for another possible reason. Since the treatment works more effectively when water flow rate is higher than some threshold value so as to collide ceramics particles mutually, then the local heat or impact pressure development on silica surface might be one of key phenomena. Thermoelectric conversion effect or piezoelectric effect is one of possibilities (see Fig. 8), but of course requires more scientific evidence.

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