The separation of liquid mixtures has been studied for a long time. Separation is based on the difference in physical properties including pore size and electrostatic interaction. Therefore, there are many difficulties in separation of materials having similar physical properties such as ethanol–water and 1,4-dioxane–water mixtures. While pervaporation based on a difference in transport rates by permeability through a membrane has been suggested, it still has many difficulties such as high energy consumption and huge facilities. Yet there is an existing technical gap to remove trace amounts of organics dissolved in water. Here, a novel separation strategy employing a metamaterial, called meta-separation using the exotic structural property of metamaterials rather than electrostatic characteristics is reported. The structural properties of metamaterials provide various functions of super-hydrophobicity based on roughness of surface, the strong capillary effect based on nanopore, and huge void for great absorption of organics. It exhibits a water contact angle of 151.3° and high adhesive property from nanopore. On the other hand, ethanol is immediately absorbed up to 93 wt%. These differences make it possible to quickly and easily eliminate organics dissolved in water. Furthermore, their applications are expected to achieve functions in environmental remediation, biofuel separation process, etc., without large-scale facilities.

1. Introduction

Controlling the wettability of solid surfaces has recently attracted great interest from both fundamental and practical perspectives by tailoring surface topography. Inspired by the microstructure of lotus leaf which forms superhydrophobic surface in nature, various superhydrophobic surfaces with different adhesive properties can be controlled from periodicity, pore size, and shape of nanopore. Adhesive force of nanopore is originated from capillary force of it. On the other hand, nanopore has widely been used for various applications. Above all, gas adsorption due to the large surface area formed by nanopores has been importantly developed. In addition, nanopore is actively used in various kinds of water treatment such as water/oil separation, desalination, and water remediation. In this case, most of them usually use only the pore size and their electrostatic characteristics on the surface. Therefore, it is difficult to find a way to use nanopores to separate mixtures of similar polarity and size.

To overcome these limitations, pervaporation employs a membrane for liquid separation, where a polymeric or zeolite membrane usually serves as the separating barrier. This process provides incomparable advantages for the separation of azeotropic mixtures due to its mild operating conditions, and lack of environmental discharge for additional species in the feed stream. Thus far, pervaporation has found viable application in the following three areas: dehydration of organic solvents, removal of dilute organic compounds from aqueous streams, and separation of organic–organic mixtures. Although the dehydration of organic solvents is a well-developed process, removing organic compounds from water still has certain limitations.

Herein, we show exotic separation based on metamaterial. The superhydrophobic surface, originated from the surface roughness, makes barrier for water. In contrary, the capillary force originated from the huge void of bulk and nanopore of surface absorbs organics. A separation method based on both the surface and bulk properties shows a new separation strategy that is completely different from the size exclusion and electrostatic force based separation strategy. Moreover, unlike conventional membranes for separation, which have been providing a passive separation barrier against external driving force, the new method provides an active driving force itself by capillary force.

2. Results and Discussion

2.1. Crystal Structure Characterization of HYLION-14

HYLION-12 with the building block for the Dirac metamaterial and metasurface was designed with two pairs of parallel strands of alkyl chains for the formation of the $xy$-plane and a pyrene moiety for stacking along the $z$-axis. This procedure is to fabricate the 3D porous structure assembled by nonpolar, noncovalent physical bonds. In order to utilize metamaterial for absorption, higher thermal stability was essential. HYLION-14 was designed and synthesized to increase the insufficient thermal stability of HYLION-12 (Figures S1–S3, Supporting Information). Crystallization through various solvents was
The orthorhombic structure was designated as \( \Phi_o \), belonging to the \( P2_22_1 \) space group. The cell parameters of \( \Phi_o \), were refined to \( a = 42.913 \) Å, \( b = 45.996 \) Å, and \( c = 9.270 \) Å (residuals: \( R_c = 8.532 \), \( R_w = 11.747 \), and \( \chi^2 = 1.37 \)) (Figure 1b). The XRD pattern of the plate-like crystal was solved in the same way as for \( \Phi_o \). It adopts a primitive monoclinic crystal structure designated as \( \Phi_m \). Classified in the \( P2 \) space group with cell parameters of \( a = 9.024 \) Å, \( b = 43.341 \) Å, \( c = 8.303 \) Å, and \( \beta = 105.30^\circ \) (residuals: \( R_c = 8.257 \), \( R_w = 11.242 \), and \( \chi^2 = 1.36 \)) and the unit cell of \( \Phi_m \) is 151% larger than that of \( \Phi_o \).

**2.2. Thermal Property of Crystals of HYLION-12 and -14**

Differential scanning calorimetry (DSC) with temperature ramping from 45 to 95 °C at a heating rate of 1 °C min\(^{-1}\) revealed two peaks at 74.01 and 81.11 °C, corresponding to phase transitions for \( \Phi_o \), and an exothermic peak for crystallization at 70.60 °C upon cooling to 45 °C at a rate of 1 °C min\(^{-1}\) (Figure 2a,b). The first heating cycle of \( \Phi_o \) had a similar profile with \( \Phi_o \) but the transition temperatures increased by 13.65 °C. In addition, the first and the second phase transition energies have been increased from 10.05 to 11.70 kcal mol\(^{-1}\) and 26.59–29.11 kcal mol\(^{-1}\), respectively. It can be understood through the effect of van der Waals interaction energy difference with increasing the number of carbons. The second heating cycle curve showed only the second phase transition, which implies that the phase formed after annealing from the isotropic liquid is different from \( \Phi_o \). The second heating cycle shows only one phase transition at 81.11 °C and it means that \( \Phi_o \) is a metastable phase.

The phase transition was investigated in depth by using high-temperature XRD. The theta/2theta peaks at 9.87°, 10.73°, 11.48°, and 19.48° in the profile of \( \Phi_o \) were indexed to the (400), (330), (240), and (002) faces, respectively (Figure 1c). After heating to 74.01 °C, the peak at 10.25° appeared, providing meaningful and important information of the phase transition. This can be interpreted as a change in the d-space of the (010) plane from 3.58 nm to 0.8 nm. During the phase transition, HYLION-14 in the middle layer of the unit cell of \( \Phi_o \) rotates 90° from the x-axis to the y-axis. As originally intended, the structure with larger inner pores and higher thermal stability is obtained from HYLION-14.
2.3. Surface and Void Property of \(\Phi_{o14}\)

The \(N_2\) gas adsorption of \(\Phi_{o14}\) depicts the porosities of it. \(\Phi_{o14}\) produced the nitrogen adsorption and desorption isotherms of a typical type IV with H3 hysteresis loops, according to the International Union of Pure and Applied Chemistry classification (Figure 3a). This behavior indicates the predominance of mesopores.[15] Brunauer–Emmett–Teller (BET) surface area measurements and the \(t\)-plot analysis were carried out to determine the specific surface area of the as-prepared material. The BET surface area plot of \(\Phi_{o14}\) corresponded to the data from the BET equation[16] (Figure 3b). The specific surface areas of \(\Phi_{o14}\) were found to be 45.25 m\(^2\) g\(^{-1}\). These small surface areas indicate that alkyl chains of \(\Phi_{o14}\) is only suitable sites for adsorption.[17] It represents a plot of the volume of adsorbed (Q) nitrogen at different relative pressures against the thickness of the adsorbed layer, \(t\), for \(\Phi_{o14}\) (Figure 3c). The average pore diameter from the Barrett–Joyner–Halenda desorption analysis was 11.3 nm, with a very wide pore size distribution, and the corresponding single-point total pore volume at \(P/P_0 = 0.975\) was 0.05 cm\(^3\) g\(^{-1}\). The experimental points in the \(t\)-plot method agreed with the data from the Harkins–Jura isotherm equation. It is clear from the plot that a set of experimental data points falls in a straight line for \(t = 0.35–0.45\) nm. Thus, \(\Phi_{o14}\) was confirmed to be porous in nature, as the \(t\)-plot curves did not pass through the origin. The intercept of the fitted linear line was positive, which confirms the presence of mesopores.

The surface area of 45.25 m\(^2\) g\(^{-1}\) means that it has a surface area of 79209 Å\(^2\) per \(\Phi_{o14}\). If any sphere has this surface area, the volume of it is 2.096 Å\(^3\). This is only 0.001% of unit cell volume. Alkyl chains floating in \(\Phi_{o14}\) are difficult to act as a wall for gas adsorption and only a very small amount of gas is absorbed. Therefore, huge space in \(\Phi_{o14}\) is ready for absorption. It has an inner space of 78 nm\(^3\) through a pore with a 2 nm radius. This shows that strong absorption occurs with very strong nanocapillary force (see the Supporting Information).

Materials for water treatment have been designed with physical properties such as the polarity and pore size.[6b] Therefore, it acted as sieves, and only surface properties were importantly treated. However, in order to overcome this approach, various nanomaterials have been newly proposed. Typically, oil–water and water–water soluble dye separation is performed using MoS\(_2\) derivatives.[18] In addition, it has been reported that the wettability is controlled by using the surface patterned polydimethylsiloxane (PDMS).[19]

While traditional separation materials are passively operated with external energy, nanocapillary force and surface properties of novel separation materials serve a new driving force to reject and uptake solute itself.[4] Although a number of separation strategies using novel materials and patterned surface have been proposed, no case has yet been announced in which different characteristics are given by separating the surface and bulk of a material. Also, separation of 1,4-dioxane and ethanol in water is recognized as a challenging task. To confirm this, the contact angle (CA) of the various phases of HYLION-14 was...
measured. The water CA of soda lime glass is 65.1° (Figure S4, Supporting Information) and water CA of $\Phi_{m,14}$ is 106.7° (Figure 4a). This is similar to water CA observed for the conventional long alkyl chain hydrocarbons such as wax. However, water CA of $\Phi_{o,14}$ is 151.3° (Figure 4b). It demonstrates superhydrophobic surface $\Phi_{o,14}$ and it can be thought as superhydrophobic surface based on their structural characteristics. At the same time, $\Phi_{o,14}$ exhibited a very strong adhesive property against water. Because the nanocapillary attraction occurs by the nanopore of $\Phi_{o,14}$. As a result, the water droplet was concretely pinned on the surface without any movement once the sample was upside down (Figure 4c). It verified that the surface of $\Phi_{o,14}$ may have liquid–solid area contact that makes high adhesive property.[3a] On the other hand, ethanol CA of $\Phi_{m,14}$ is 23.1° (Figure 4d). This value conforms to usual ethanol CA of hydrophobic surfaces.[21] Notably, as a characteristic feature of the $\Phi_{o,14}$, no ethanol droplet is formed on the surface. Instead, rapid wetting occurs as can be seen in Video S1 in the Supporting Information. Therefore, for water–organic mixture, only $\Phi_{o,14}$ can selectively reject water and uptake organics (Figure 1e,f). From the structural difference of the crystal structure of HYLION-14, the result of water–organic separation can be more clearly understood through the total scheme of meta-separation process (see Supporting Information).

2.4. Spontaneous Separation of Water–Organic Mixtures

Thermogravimetric analysis (TGA) demonstrates the extremely high absorption capacity of the $\Phi_{o,14}$ (Figure 5a). The absorbed amount was 92.6 wt% for ethanol and 98 wt% for 1,4-dioxane. $\Phi_{o,14}$ maintained their structure after soaking up with the guest molecules, as confirmed by PXRD patterns (Figure 1d). With the dropwise addition of the solvent to $\Phi_{o,14}$, the PXRD peak at 19.48° was gradually broadened, indicating twisting of HYLION-14 along the z-axis, while the other peaks remained unchanged during the soaking process.[22]

Moreover, the rate of absorption of various organics by $\Phi_{o,14}$ is measured by modified edge immersion tests.[23] 1,4-dioxane, acetone, butanol, and ethanol are absorbed in 1.5 s (Figure 5b) and the maximum velocity ($V_{\text{max}}$) of absorption was calculated using the hill function.[24] The $V_{\text{max}}$ values for 1,4-dioxane, acetone, butanol, and ethanol were 95.32, 98.68, 96.19, and 93.81, respectively (Figures S5–S8, Supporting Information).

Gas chromatography was carried out to verify high separation capability of $\Phi_{o,14}$ for an aqueous azeotropic mixture. It should remove 1,4-dioxane, acetone, butanol, and ethanol from the water mixtures. To confirm this, water–organic mixture was prepared with a 100 µg L$^{-1}$ for each solvent and treated with separation cartridge which is $\Phi_{o,14}$ was packed in the perforated Al-foil to prevent diffusion of $\Phi_{o,14}$ in water mixture and to uptake solvents. The separation cartridge was dipped in water–organic mixtures and shaken a minute for separation (see Supporting Information). The results of gas chromatograph markedly show the peaks of water and individual compounds in the azeotropic mixtures (Figure 5c). The profile of each sample showed a water peak at 2.5 s and the signals in the order of acetone, butanol, ethanol, and 1,4-dioxane were detected thereafter, respectively. $\Phi_{o,14}$ was well packed into a porous metal pipe and immersed in an agitated azeotropic mixture for 1 min. There were no peaks of organic compounds in the results of gas chromatography from the treated azeotropic mixtures (Figure 5d). This demonstrates the huge
potential of $\Phi_{o,14}$. Above all, the mixture of water and 1,4-dioxane is one of the most notorious positive azeotropes because of its infinite miscibility and similar boiling point. Nevertheless, the molecular metamaterial perfectly separates them without any kind of energy consumption. Moreover, acetone, butanol, and ethanol azeotropes are also very important mixtures that must be separated in the biofuel industry. The selective process for separating acetone, butanol, and ethanol from water is still the major bottleneck for the biofuel industry and the most expensive procedure regarding energy efficiency and productivity.][25] Herein, $\Phi_{o,14}$ can remove acetone-butanol-ethanol (ABE) from water spontaneously. Meta-separation can provide a cheap, convenient, and complete separation method without energy consumption.

3. Conclusion

A novel strategy of separation exploiting the structural properties, rather than polarity, of a metamaterial was demonstrated. The regular and periodic structure of the molecular metamaterial provides low surface energy and super-hydrophobicity with high adhesive forces against water, whereas the nanopores confer a capillary effect for absorption of organics. The molecular metamaterial can uptake various organic solvents from 92 to 98 wt%. Based on this exotic concept, azeotropes and various organic–water mixtures that are difficult to separate by pervaporation were easily separated in a few seconds. In particular, the separation using a capsule containing the molecular metamaterial opens up new possibilities for water treatment or organic solvent capture anywhere without large facilities. This approach has the potential to lead to great innovations not only in the biofuel industry but also in the elimination of various endocrine disruptors from environmental system.

4. Experimental Section

Reagents: Pyrene, NaIO₄, Bu₄NBr, Na₂S₂O₄, and dodecyl bromide were purchased from Sigma-Aldrich and used as received. CDCl₃ was purchased from Cambridge Isotope Laboratories and was used for the ¹H NMR spectroscopic studies.

Synthesis: A mixture of pyrene-4,5,9,10-tetraone (10 mmol), Bu₄NBr (13 mmol), and Na₂S₂O₄ (115 mmol) in H₂O (50 mL) and tetrahydrofuran (THF) (50 mL) was shaken for 5 min. Then the color of mixture was changed from dark brown to pale yellow. Bromo tetracane (60 mmol) was added and followed by aqueous potassium hydroxide (306 mmol, in 50 mL H₂O).
The mixture was stirred for overnight, poured into a mixture solution of H$_2$O (50 mL) and ethyl acetate (30 mL). The yellow solid was filtered and washed with ethanol. After drying in a vacuum, the solid was recrystallized from ethyl acetate resulting in a white solid with a yield of 85%.

Characterization: $^1$H NMR (600 MHz) spectra were recorded on a Varian DRX VNMRS 600 instrument. The purity of the products was determined by a combination of thin-layer chromatography on silica gel coated aluminum plates (with F254 indicator; layer thickness, 200 µm; particle size, 2–25 µm; pore size 60 Å, Sigma-Aldrich) Infrared spectra were taken in a KBr disc on a Jasco Fourier transform infrared spectroscopy 460 plus spectrometer (Figures S1 and S2, Supporting Information).

Crystallization: HYLION-14 was crystallized by various solvents and the morphology of HYLION-14 is divided into two types of needle and plate shapes. Crystallization was carried out through gradual cooling and an evaporation method. The HYLION-14 solution was prepared with 1 × 10^{-3} M with various solvents and in the case of gradual cooling, the temperature was gradually dropped by 1 °C min^{-1} from 50 °C to 24 °C. In the case of evaporation, the prepared solution was allowed to stand in an 8 mL vial at room temperature.

Thermal Property Analysis: Thermal phase transitions were measured on TA Instrument SDT Q 600 ver. 20.9 Build 20 system DSC integrated an 8 mL vial at room temperature.

Pressure adsorption isotherms measured at 77 K for N$_2$ were carried out by the GC oven was held isothermal at 150 °C, respectively, J&W scientific, Folsom, CA, USA) was used for the separation. A sample was prepared with a 100 µg L^{-1} for each solvent. The azeotropic mixture of water and solvents was treated for each solvent. The azeotropic mixture of water and solvents was treated with a refrigerated cooling system. In all cases, the heating and cooling rates were 1 °C min^{-1}. The transition temperatures were measured as the rates were 1 with a refrigerated cooling system. In all cases, the heating and cooling temperatures were measured as the maxima and minima of their endothermic and exothermic peaks.

X-Ray Diffractometer Analysis: Theta/2theta and in-plane X-ray diffraction patterns of all samples were measured on a SMARTLAB (Rigaku Co. Ltd.,) diffractometer using monochromatized Cu-K$_\alpha$ ($\lambda = 0.15418$ nm) radiation under 40 kV and 100 mA. High temperature XRD was measured on Bruker D8 Advance with a high temperature attachment in theta/2theta geometry. The high temperature stage allows the sample to be measured at tightly controlled temperatures from room temperature to 85 °C in a purge N$_2$ gas.

Porosity Analysis: Low-pressure (<1 atm) adsorption measurements were performed using a Quantachrome Autosorb-iQ2ST/MP analyzer. Ultrahigh purity grade N$_2$ was used for adsorption measurements. Low-pressure adsorption isotherms measured at 77 K for N$_2$ were carried out in liquid nitrogen. Prior to analysis, the HYLION crystals were typically degassed at room temperature for 16 h to remove the solvent molecules.

Contact Angle: Contact angle were measured using a KSV CAM 200 goniometer. The static contact angle values were calculated by fitting the water droplet baseline in the instrument software utilizing the Young–Laplace model.

Solvent Uptake and Gas Chromatography Experiment: To confirm the purifying ability of the $\Phi_{34}$ quantitatively, the gas chromatography/mass spectroscopy (GC/MS) executed. The sample was prepared with a 100 µg L^{-1} for each solvent. The azotropic mixture of water and solvents was treated with the $\Phi_{34}$. The $\Phi_{34}$ was packed in the Al-foil to prevent diffusion in azoeterpe and to uptake solvents the Al-foil was drilled by 23G needle. The Al-foil package was dipped in an azeotrope and to uptake solvents the Al-foil was drilled by 23G needle.

The GC oven was set to 150 °C for 2 min, and then temperature programmed to 300 °C at a rate of 6 °C min^{-1} with a final hold time of 33 min. The entire GC run was 60 min long. A 10 µL Hamilton gas tight syringe with a polytetrafluoroethylene (PTFE) tip plunger was used for sample injection. A volume of 3 µL of sample was injected splitless for 2 min into a deactivated double tapered splitless liner. A 20 min. ml^{-1} split flow was maintained after the splitless time. A gold coated injector seal was used. The front of the capillary column was inserted 2 mm past the injection port seal. The linear velocity of air injected at 80 °C with a column back pressure of 30 psi (helium) was measured at 34 cm s^{-1} (1 psi = 6894.76 Pa). Supelco Thermogreen LB-2 septa were used in the injection port. The GC injection port and GC–MS transfer line temperature were maintained at 280 and 300 °C, respectively.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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Research data are not shared.

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