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에티렌디암모늄 비스(파라-메틸벤젠슬폰네이트) 수화물의 결정 및 분자구조

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The Crystal and Molecular Structure of Ethylenediammonium *bis*(*P*-Methylebenzenesulfonate) Monohydrate

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요 약. 에티렌디암모늄 비스(파라-메틸벤젠슬폰네이트) 수화물, $C_2H_{10}N_2^{2+} \cdot 2C_7O_3H_7S^- \cdot H_2O$의 결정구조를 X-선 회절법에 의하여 결정하였다. 결정은 단사축계이며 공간군은 $P2_1$, 단위세포 내에는 2개의 분자가 들어 있으며 그의 단위세포상수는 $a=12.649(2)$, $b=7.727(1)$, $c=11.295(2)$Å, $\beta=111.8(1)°$이다. 분자구조는 Mo-Kα 선을 이용하여 측정한 1134개의 회절반점에 대하여 직접법에 의하여 풀었으며 신뢰도인자는 6.0%이다. *A* 및 *B*로 된 2개의 파라-메틸벤젠슬폰네이트는 에틸렌디암모늄이온과 수소결합에 의하여 짝을 이루고 있다. *B*쪽에 있는 슬폰네이트기는 그의 위치가 두곳으로 퍼져있다. 모두 6개의 수소결합이 있으며 이들중 4개는 에틸렌디암모늄이온과 슬폰네이트기 사이의 결합이며 나머지 2개는 물분자와 연결되어 있다.

ABSTRACT. The crystal structure of ethylenediammonium bis (*p*-methylbenzenesulfonate) monohydrate, $C_2H_{10}N_2^{2+}$. $2(C_7O_3H_7S^-)$. H_2O has been determined by X-ray diffraction techniques. The space group is $P2_1$, in 2 unit cell with $a=12.649(2)$ Å, $b=7.727(1)$ Å, $c=11.295(2)$ Å, $\beta=111.8(1)°$, and $z=2$. The structure was solved by direct methods and refined to $R=0.060$ for 1134 reflections measured with Mo-Kα radiation. Two *p*-methylbenzenesulfonates, fragment *A* and *B*, form a pair through the hydrogen bonds to the ethylenediammonium ion. The sulfonategroup in the fragment *B* are disordered. There are six unique hydrogen bonds, of which four are between the ethylenediammonium ion and the sulfonate groups and remaining two involve the water molecule.

INTRODUCTION

Aromatic sulfonic acids easily react with weak bases such as ammonia ($K_b=1.8\times10^{-5}$) and ethylenediamine ($K_b=8.5\times10^{-5}$) to form salts. It has been known that the salts of aromatic sulfonic acids with long alkyl side chains have some useful detergent properties since they do not form the hard scum. Actually the first synthetic detergents that we have used are a series of the salts of alkylbenzenesulfonates. They have a large non-polar hydrocarbone tail

and a polar end, like soap, R-  -$SO_3^-Na^+$, R=branched alkyl.

In an effort to elucidate the association modes between the alkylbenzenesulfonate and various bases other than alkali metal ions, the crystal structure analysis of the title compound has been done.

EXPERIMENTAL

Needle, colorless crystals of ethylenediammonium *bis*(*p*-methylbenzenesulfonate) monohydrate (EMBS · H_2O) were obtained by slow evaporation from a 2 : 1 mixture of an aqueous solution of *p*-methylbenzenesulfonic acid and ethylenediamine. The density was measured by the flotation method in a carbontetrachloride-benzene mixture. Preliminary oscillation and Weissenberg photographs showed that the crystal belonged to a monoclinic system with the space group $P2_1$.

The unit cell parameters were determined by the least-squares fit of the 2θ angles for 25 centered reflections as measured ($24° \leqslant 2\theta \leqslant 49°$) with Mo-K$\alpha$ radiation on the automated Rigaku-Denki AFC diffractometer. The crystal data are shown in *Tabel* 1.

A crystal (0.3×0.3×0.5mm) was used to measure the intenity. 1962 independent reflections were measured with the graphite-monochromated Mo-Kα radiation, using 2θ-ω scan technique over a scan range of ($1.2° + 0.5 \tan\theta$) in ω at a scan rate of 8°/min. Background counts were also measured for 10 seconds at each end of the scan ranges. Three standard reflections 103, 402, $2\bar{1}\bar{1}$ that were obtained at every 50 reflections showed no apparent change in their intensities during the data collection. The intensities were modified for Lorentz and polarization effects and then were converted to structure amplitudes. The 1134 reflections were treated as observed by the critetion on $|F_0| \geqslant 6\sigma$ (F), and were used for the subsequent calculations. No correction for the absorption and extinction effects was made.

Table 1. Crystal data of EMBS · H_2O

Chemical formula	: $C_2H_{10}N_2^{2+} \cdot 2C_7O_3H_7S^- \cdot H_2O$
Molecular weight	: 422.56
Crystal system	: Monoclinic
Space group	: $P2_1$
Unit cell parameter	: a=12.649(2)Å
	b=7.727(1)Å
	c=11.295(2)Å
	β=111.8(1)°
	V=1025.01Å^3
	z=2
Density	: D_m=1.375g/cm^3
	D_c=1.369
F(000)	: 428.0

STRUCTURE-DETERMINATION AND REFINEMENT

An overall temperature factors (B=3882Å^2) and a scale multiplier(K=0.061) were obtained through the calculation of normalized structure factors. The positons of two *p*-methylbenzenesulfonates(MBS) were obtained by the direct methods(MULTAN 77)[1] that used 245 E values with $E \geqslant 1.20$ for phase generation. All other remaining non-hydrogen atom positions were obtained by the subsequent difference Fourier synthesis(SHELX 76)[2]. An isotropic full-matrix least-squares refinement with all non-hydrogen atoms gave R=0.126, and a few cycles of anisotropic refinement lowered the R value to 0.093.

It was found at this stage that the oxygen atoms in the sulfonate group of the fragment *B* were disordered. The occupancy factors of the major sites for the oxygen atoms O_{11}, O_{12} and O_{13} were refined to 0.71, 0.54 and 0.67, respectively. The effect of high thermal motion (probably due to positional disorders of oxygen atoms having comparably large occupancy factors

for the minor sites) was seen in the large anisotropic temperature factors and the large e.s.d.'s in bonds and angles for the disordered atoms. The average value of angles of the major site–S_{11}–minor site was 53.3°. Although most of hydrogen atoms were found in a differece Fourier map, the hydrogen atoms that belonged to the methyl group in the fragment *B* and the methylene groups of ethylenediamine were geometrically calculated with the assumption of C–H=1.0Å ∠and H—C—H=109°.

The final refinement, anisotropically for non-hydrogen atoms and isotropically for hydrogen atoms, gave $R=0.060$ for 1134 reflections, and the weighted *R* factor ($R_w=\sum\sqrt{w}(|F_0|-k|F_0|)/\sum\sqrt{w}|F_0|$) was 0.652. The function $\sum w(|F_c|-k|F_c|)^2$ was minimized, where $w=1.00/(\sigma^2|F|+0.0174|F|^2)$. The atomic scattering factors were taken from the International Tables for X-Ray Crystallography[3]. The final atomic coordinates and the temperature factors are listed in *Table* 2.

Table 2. Fractional atomic coordinates and thermal parameters for EMBS · H_2O

A. Non-hydrogen atoms[a]

ATOM	*X*	*Y*	*Z*	*U*11	*U*22	*U*33	*U*23	*U*13	*U*12	s.o.f.
S1	1981(1)	472(0)	3949(2)	37(1)	44(1)	47(1)	−2(1)	14(1)	−5(1)	
O1	1779(6)	−1144(9)	3311(8)	54(4)	45(4)	79(5)	−22(4)	18(3)	−21(3)	
O2	1971(6)	1908(11)	3088(7)	61(4)	75(5)	62(4)	29(4)	−6(3)	−11(4)	
O3	1224(5)	839(10)	4597(6)	37(3)	74(6)	75(4)	−18(4)	24(3)	0(3)	
O1	3391(6)	447(13)	5116(7)	36(3)	23(4)	47(4)	−6(5)	22(3)	−3(5)	
O2	3634(7)	1359(13)	6241(8)	43(5)	43(5)	50(5)	−10(5)	23(4)	1(4)	
O3	4723(8)	1332(13)	7096(8)	57(5)	46(5)	40(4)	−4(5)	16(4)	−5(5)	
O4	5618(6)	573(14)	6870(7)	47(4)	26(4)	46(4)	16(5)	13(3)	2(4)	
O5	5322(7)	−306(14)	5696(8)	44(5)	65(6)	45(5)	7(5)	6(4)	15(5)	
O6	4228(7)	−354(13)	4821(8)	51(5)	51(5)	42(5)	−1(5)	21(4)	1(5)	
O7	6806(9)	658(19)	7791(11)	53(5)	57(3)	47(6)	−2(7)	4(5)	0(7)	
S11	8013(2)	368(5)	2357(2)	36(1)	48(1)	55(1)	9(2)	16(1)	−6(1)	
O11	8393(8)	−645(27)	3441(17)	22(5)	136(15)	142(13)	102(12)	−4(7)	−10(7)	0.71
O11A	8294(33)	−1208(49)	2275(80)	42(24)	54(23)	469(97)	−40(38)	6(41)	29(19)	0.29
O12	8320(20)	−391(43)	1329(30)	64(14)	154(31)	150(25)	−79(23)	82(17)	−22(17)	0.54
O12A	8336(15)	1048(46)	1372(17)	35(10)	205(23)	57(10)	35(15)	−10(8)	−35(15)	0.46
O13	8417(10)	2087(16)	2639(19)	65(3)	61(7)	153(14)	−52(9)	60(9)	−27(6)	0.67
O13A	8310(23)	1693(70)	3421(28)	66(18)	250(58)	67(18)	82(27)	44(15)	−22(22)	0.33
O11	6509(7)	394(15)	1785(6)	53(5)	39(4)	30(4)	6(6)	23(3)	3(6)	
O12	5942(8)	1179(14)	2460(10)	50(5)	52(6)	59(5)	11(5)	23(5)	2(5)	
O13	4797(9)	1145(14)	2001(10)	59(6)	48(5)	64(6)	−4(5)	38(5)	20(5)	
O14	4152(7)	395(18)	894(8)	56(5)	57(5)	50(5)	9(7)	33(4)	1(7)	
O15	4722(9)	−403(15)	213(9)	65(7)	53(6)	46(6)	0(5)	17(5)	−11(6)	
O16	5905(7)	−348(13)	841(8)	52(5)	45(5)	42(5)	1(4)	33(4)	−9(5)	
O17	2858(10)	273(22)	430(19)	51(6)	59(9)	135(11)	7(10)	30(7)	−1(7)	
N111	−24(8)	5692(11)	1683(8)	36(5)	46(5)	64(5)	−23(4)	20(4)	−11(4)	
O111	40(7)	5466(15)	2249(8)	64(5)	51(5)	61(5)	−28(6)	21(4)	−8(6)	
O112	163(11)	5198(12)	5395(10)	125(9)	27(6)	66(5)	3(5)	45(6)	1(6)	
N112	30(10)	16989(13)	4205(12)	91(9)	43(5)	57(6)	−14(5)	42(6)	−16(5)	
O	162(7)	−463(13)	710(7)	67(5)	101(7)	71(5)	−18(5)	44(4)	−8(5)	

Table 2(continued)

B. Hydrogen atoms[b]

Atom	X	Y	Z	U11
HC2	298(8)	206(14)	644(9)	5(3)
HC3	490(8)	194(16)	801(10)	8(4)
HC5	598(8)	−96(14)	548(9)	8(3)
HC6	403(8)	−103(14)	393(9)	8(3)
HM1	684(9)	−58(11)	736(11)	7(4)
HM2	738(8)	149(14)	758(11)	9(3)
HM3	705(8)	22(13)	855(12)	9(4)
HC12	642(1)	182(1)	335(1)	4(2)
HC13	438(1)	170(1)	265(1)	3(2)
HC15	424(1)	−108(2)	−66(1)	4(2)
HC16	633(1)	−90(1)	6(1)	4(2)
HM11*	198(9)	−13(2)	−5(10)	4(3)
HM12	−730(1)	126(2)	−18(2)	4(3)
HM13*	276(12)	107(20)	84(13)	8(4)
HN11	−39(1)	364(1)	73(1)	5(2)
HN21	52(6)	348(10)	187(6)	6(2)
HN31	−44(5)	312(8)	182(5)	7(3)
H1C1*	−67(1)	614(2)	178(1)	5(2)
H2C1*	71(1)	610(2)	221(1)	5(3)
H1C2*	−52(1)	460(1)	364(1)	3(2)
H2C2*	85(1)	448(1)	405(1)	4(2)
HN12	−38(7)	−300(10)	398(7)	9(2)
HN22	64(11)	783(18)	401(13)	8(6)
HN32	26(7)	660(11)	467(11)	8(3)
HW1	−36(9)	−41(15)	86(10)	6(4)
HW2	41(7)	39(14)	115(8)	6(3)

[a] Positional parameters×10^4; thermal parameters×10^3, the expression used for the anisotropic temprature factor is $\exp(-2\pi(U11\ h^2a^{*2}+U22\ k^2b^{*2}+U33\ l^2c^{*2}+2U23\ klb^*c^*+2U13\ hla^*c^*+2U12\ hka^*b^*)$. E. s. d.'s in parentheses are for the least significant figure. [b] Positional parameters ×10^3; thermal parameters×10^2, which are the isotropic temperature factor for the expression $\exp(-8\pi U\ \sin^2\theta/\lambda^2)$.

*Hydrogen atoms which are not located in a difference Fourier map. s. o. f.: site occupancy factor.

RESULTS AND DISCUSSIONS

EMBS · H_2O is a salt-like compound composed of two deprotonated *p*-methylbenzenesulfonate anions and a doubly protonated ethylenediammonium cation. Its molecular structure and atomic numberings are shown in *Fig.* 1. Its

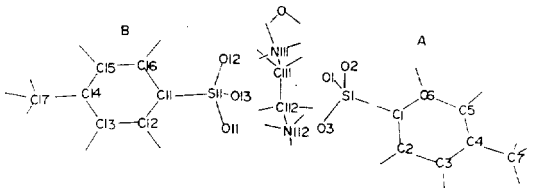

Fig. 1. The atomic numbering scheme of EMBS·H_2O.

Table 3. (a) Bond distances(Å) and (b) angles(°) with e. s. d.'s in parentheses

(a) Bond distances

Fragment A		Fragment B		Ethylenediamine	
S_1–O_1	1.417(7)	S_{11}–O_{11}	1.380(16)	N_{111}–C_{111}	1.502(12)
S_1–O_2	1.472(7)	S_{11}–O_{11A}	1.281(53)	C_{111}–C_{112}	1.484(13)
S_1–O_3	1.434(6)	S_{11}–O_{12}	1.476(28)	C_{112}–N_{112}	1.527(14)
S_1–C_1	1.781(8)	S_{11}–O_{12A}	1.421(24)		
C_1–C_2	1.384(11)	S_{11}–O_{13}	1.417(14)		
C_1–C_6	1.371(11)	S_{11}–O_{13A}	1.516(37)		
C_2–C_3	1.357(12)	S_{11}–C_{11}	1.767(9)		
C_3–C_4	1.380(12)	C_{11}–C_{12}	1.372(13)		
C_4–C_5	1.412(12)	C_{11}–C_{16}	1.360(12)		
C_4–C_7	1.478(14)	C_{12}–C_{13}	1.347(13)		
C_5–C_6	1.370(12)	C_{13}–C_{14}	1.392(14)		
		C_{14}–C_{15}	1.380(14)		
		C_{14}–C_{17}	1.524(18)		
		C_{15}–C_{16}	1.391(13)		

(b) Angles

A. Fragment A

S_1–C_1–C_2	119.7(6)	C_1–C_2–C_3	117.9(8)
S_1–C_1–C_6	118.5(6)	C_1–C_6–C_5	118.5(8)
O_1–S_1–O_2	111.9(4)	C_2–C_1–C_6	121.7(7)
O_1–S_1–O_3	114.0(4)	C_2–C_3–C_4	124.1(8)
O_2–S_1–O_3	110.2(4)	C_3–C_4–C_5	115.4(7)
O_1–S_1–C_1	108.0(4)	C_3–C_4–C_7	122.8(8)
O_2–S_1–C_1	105.0(4)	C_4–C_5–C_6	122.4(8)
O_3–S_1–C_1	107.4(4)	C_5–C_4–C_7	121.9(8)

B. Fragment B

S_{11}–C_{11}–C_{12}	121.1(7)	C_{11}–C_{12}–C_{13}	121.4(9)
S_{11}–O_{11}–C_{16}	119.5(7)	C_{11}–C_{16}–C_{15}	119.8(8)
O_{11}–S_{11}–O_{12}	111.5(13)	C_{12}–C_{11}–C_{16}	119.6(8)
O_{11}–S_{11}–O_{13}	110.8(9)	C_{12}–C_{13}–C_{14}	120.6(9)
O_{12}–S_{11}–O_{13}	111.8(12)	C_{13}–C_{14}–C_{15}	118.0(9)
O_{11}–S_{11}–C_{11}	107.7(7)	C_{13}–C_{14}–C_{17}	122.0(10)
O_{12}–S_{11}–C_{11}	106.3(11)	C_{14}–C_{15}–C_{16}	120.7(9)
O_{13}–S_{11}–C_{11}	108.8(6)	C_{15}–C_{14}–C_{17}	119.9(10)

O_{11}-S_{11}-O_{11A}	60.7(25)	O_{11A}-S_{11}-O_{12A}	97.3(26)
O_{11}-S_{11}-O_{12A}	142.9(12)	O_{11A}-S_{11}-O_{13A}	134.9(28)
O_{11}-S_{11}-O_{13A}	77.2(16)	O_{12A}-S_{11}-O_{13A}	108.3(17)
O_{11A}-S_{11}-O_{12}	53.5(26)	O_{12}-S_{11}-O_{12A}	45.2(14)
O_{11A}-S_{11}-O_{13}	145.2(25)	O_{12}-S_{11}-O_{13A}	146.3(18)
O_{12A}-S_{11}-O_{13}	69.2(11)		
O_{13A}-S_{11}-O_{13}	39.3(15)		
O_{11A}-S_{11}-C_{11}	106.0(24)		
O_{12A}-S_{11}-C_{11}	107.1(10)		
O_{13A}-S_{11}-C_{11}	101.5(14)		
C. Ethylenediamine			
N_{111}-C_{111}-C_{112}	106.1(7)		
C_{111}-C_{112}-N_{112}	106.7(8)		

bond distances and angles are given in *Table* 3. Corresponding average distances and angles of MBS in the fragment A are in a good agreement with those of 1-[(2S, 3S)-2-(N, N-dibenzylamino)-3-methyl-1-pentyl] pyridium *p*-toluenesulfonate[4] and 1-[2S)-2-2-(N, N-dibenzylamino) -1-propyl} pyridium *p*-toluenesulfonate[4], and other similar compounds.[5~9]. The average bond distances and angles[10] of the methylbenzene group in the fragment *B* also show a good agreement with those of the fragment A except for the exocyclic C—C bonds of the methyl group. These are 1.524(18) Å for the C_{14}—C_{17} bond and 1.478(14) Å for the C_4—C_7 in the fragments *B* and *A* respectively.

In the ethylenediammonium ion, the C—C length of 1.484 (13) Å is shorter than that of 1.55(2) Å in tris(ethylenediamine) zinc (11) chloride dihydrate[11], but average C—N length of 1.515 (13) Å is longer than that of 1.47(2) Å. A single ion forms a zig-zag shape with the N—C—C—N torsion angle of 176.7°.

The least squares planes in the molecules are given in *Table* 4. The MBS molecule is essentially planar. The selected torsion angles are given in *Table* 5.

The packing drawings of the structure are shown in *Fig.* 2 and *Fig.* 3, which represent two projections of the crystal structure along the *b*- and *c*-axis respectively. The crystal packing consists of alternating hydrophobic and hydrophilic layers perpendicular to the direction of the a-axis. In the hydrophilic layers, the water molecules and the ethylenediammonium cations are hydrogen-bonded[12] to the juxtaposed sulfonate ends of the molecules which are related by the 2-fold screw axis at $a=0.0$. There are six hydrogen bonds, of which four are between

Table 4. Least-squares planes

A. Fragment A

$0.3446X - 0.8390Y - 0.4212Z = -1.2170$

Displacement of the atoms from the above plane:

C_1 : −0.015Å C_2 : 0.023Å C_3 : −0.021Å

C_4 : 0.009 C_5 : −0.001 C_6 : 0.004

S_1* : 0.072 C_7* : 0.043

B. Fragment B

$0.1594X + 0.8653Y - 0.4753Z = 0.5798$

Displacement of the atoms from the above plane:

C_{11} : −0.013Å C_{12} : 0.013Å C_{13} : −0.014Å

C_{14} : 0.016 C_{15} : −0.017 C_{16} : 0.016

S_{11}* : −0.051 C_{17}* : −0.064

C. Ethylenediamine

$0.9608X - 0.0102Y + 0.2769Z = -0.2479$

Displacement of the atoms from the above plane:

N_{11} : 0.000Å C_{111} : 0.000Å C_{112} : 0.000Å

N_{112}* : 0.084

*Atoms which are not used to define the planes.

Table 5. Selected torsion angles(°)

A. Fragment A			
O_1-S_1-C_1-C_2	−147.8	O_1-S_1-C_1-C_6	38.0
O_2-S_1-C_1-C_2	92.7	O_2-S_1-C_1-C_6	−81.5
O_3-S_1-C_1-C_2	−24.6	O_3-S_1-C_1-C_6	161.3
B. Fragment B			
O_{11} -S_{11}-C_{11}-C_{12}	66.9	O_{11} -S_{11}-C_{11}-C_{16}	−114.4
O_{11A}-S_{11}-C_{11}-C_{12}	130.5	O_{11A}-S_{11}-C_{11}-C_{16}	−50.7
O_{12} -S_{11}-C_{11}-C_{12}	−173.6	O_{12} -S_{11}-C_{11}-C_{16}	5.1
O_{12A}-S_{11}-C_{11}-C_{12}	−126.4	O_{12A}-S_{11}-$C_{11}C_{16}$	52.3
O_{13} -S_{11}-C_{11}-C_{12}	−53.2	O_{13} -S_{11}-C_{11}-C_{16}	125.6
O_{13A}-S_{11}-C_{11}-C_{12}	−13.1	O_{13A}-S_{11}-C_{11}-C_{16}	165.6
C. Ethylenediamine			
N_{111}-C_{111}-C_{112}-N_{112}	176.7		

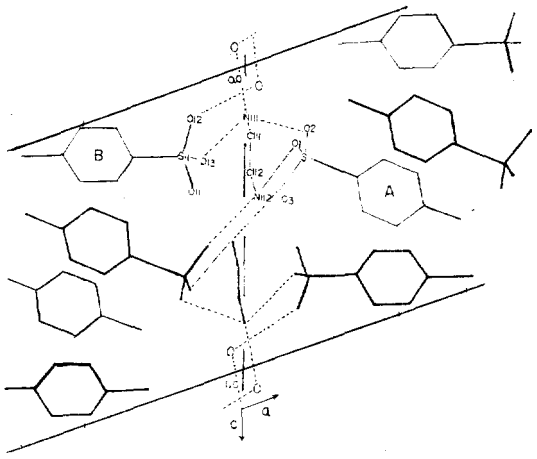

Fig. 2. Projection of the structure along the *b*-axis.

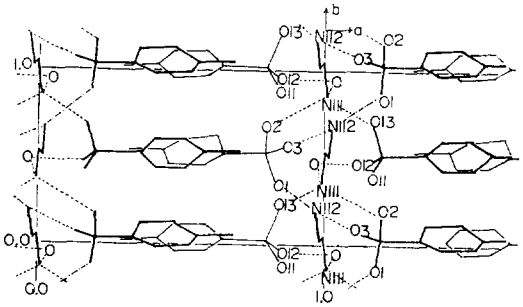

Fig. 3. Projection of the structure along the *c*-axis.

Table 6. Hydrogen bonds

a	*b*	*c*	*a*–*c*(Å)	*b*–*c*(Å)	$\angle abc$ (°)	position of *c*
N_{111}	$-HN_{21}\cdots O_2$		2.80(1)	2.20	150	x, y, z
N_{111}	$-HN_{31}\cdots O_{13}$		2.86(2)	2.14	162	$x-1, y, z$
	$-HN_{31}\cdots O_{12A}$*		2.84(3)	2.15	153	$x-1, y, z$
N_{112}	$-HN_{22}\cdots O_1$		2.83(1)	2.05	153	$x, y+1, z$
N_{112}	$-HN_{32}\cdots O_3$		2.88(1)	2.39	137	$\bar{x}, \frac{1}{2}+y, \bar{z}+1$
	$-HN_{32}\cdots O_{13A}$*		2.61(4)	2.24	120	$\bar{x}+1, \frac{1}{2}+y, \bar{z}+1$
O	$-HW_1\cdots O_{12}$		2.67(3)	1.93	176	$x-1, y, z$
	$-HW_1\cdots O_{12A}$*		2.92(3)	2.24	153	$x-1, y, z$
N_{111}	$-HN_{11}\cdots O$		2.72(1)	1.89	139	$\bar{x}, \frac{1}{2}+y, \bar{z}$

* Shows alternatives.

the ethylenediammonium ions and the sulfonate groups and the remaining two involve the water molecules, as listed in *Table* 6. The benzene rings and the methyl groups form the hydrophobic layers.

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