67B Solid solution

No. 67B-1 $NaKC_4H_4O_6 \cdot 4H_2O$–$NaNH_4C_4H_4O_6 \cdot 4H_2O$

1a This solid solution was first studied by Kurtschatow et al. in 1932. 32Kur

b Phase diagram: Fig. 67B-1-001.
$NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ is classified into four regions as follows (see Fig. 67B-1-001). 58Mak

region	x	state (region) $T\rightarrow$		
I	0...0.03	P	F(I)	P
II	0.03...0.18	P		P
III	0.18...0.88	F(III)		P
III–IV bounary	0.88...0.93	(F)(IV)	F(III)	P
IV	0.93...1.00	(F)(IV)		P

In upper half temperature of the region II, broad dielectric peak appears, however they do not correspond to the phase transition.
In (F)(IV) phase, P_s can be reversed by mechanical stress.
Direction of P_s:

state	F(I)	F(III)	(F)(IV)
direction	[100]	[100]	[010]

Density ρ_X: see the tables in subsection 3a.

2a Crystal growth:
Cooling or evaporation method from aqueous solution: Fig. 67B-1-002.
Relation of molar fraction x in the mixed crystals and the aqueous solutions: Fig. 67B-1-003.

3a Unit cell parameters: Fig. 67B-1-004.

T = 294 K: 96Suz1

x	0.9	0.94	1
a [Å]	12.165(9)	12.172(4)	12.206(7)
b [Å]	14.420(7)	14.421(4)	14.451(6)
c [Å]	6.239(4)	6.239(2)	6.250(4)
V [Å^3]	1094(1)	1095.2(6)	1102(1)
ρ_X [$\cdot 10^3$ kg m^{-3}]	1.598	1.592	1.573

x = 0.28: 96Suz2

T [K]	213	253	293
a [Å]	11.899(2)	11.929(1)	11.955(1)
b [Å]	14.273(2)	14.297(1)	14.317(1)
c [Å]	6.211(1)	6.221(1)	6.231(1)
V [Å^3]	1054.9(2)	1061.0(2)	1066.5(1)
ρ_X [$\cdot 10^3$ kg m^{-3}]	1.740	1.730	1.721

b Z = 4 in III and IV regions. 96Suz1, 96Suz2

Crystal structures: Table 67B-1-001, Table 67B-1-002; Fig. 67B-1-005.
Occupancies of NH_4 ions in the two crystallographically independent sites: Fig. 67B-1-006.

For x = 0.28: (96Suz2)

T [K]	213	253	293
α [site (1)]	0.384(3)	0.393(4)	0.387(3)
β [site (2)]	0.159(2)	0.173(3)	0.187(2)

Interatomic distances and angles: Table 67B-1-003, Table 67B-1-004, Table 67B-1-005, Table 67B-1-006, Table 67B-1-007.
Bond lengths and bond angles of tartrate ion: Table 67B-1-008.

4	Thermal expansion: see Tables in subsection 3a.	
5a	Dielectric constants in low frequency region: Fig. 67B-1-007, Fig. 67B-1-008, Fig. 67B-1-009, Fig. 67B-1-010, Fig. 67B-1-011, Fig. 67B-1-012. Dielectric dispersion: Fig. 67B-1-013, Fig. 67B-1-014, Fig. 67B-1-015, Fig. 67B-1-016, Fig. 67B-1-017, Fig. 67B-1-018. Dielectric relaxation time, dielectric strength, dispersion parameter: Fig. 67B-1-019, Fig. 67B-1-020, Fig. 67B-1-021, Fig. 67B-1-022, Fig. 67B-1-023, Fig. 67B-1-024. Phase diagram in regard to p: Fig. 67B-1-025, Fig. 67B-1-026.	
c	Spontaneous polarization: Fig. 67B-1-027, Fig. 67B-1-028.	
d	Pyroelectricity: Fig. 67B-1-029, Fig. 67B-1-030.	
6a	Heat capacity: Fig. 67B-1-031, Fig. 67B-1-032, Fig. 67B-1-033.	
7a	Piezoelectricity: Fig. 67B-1-034.	
8a	Elastic properties: Fig. 67B-1-035, Fig. 67B-1-036.	
9a	Refractive indices: Fig. 67B-1-037.	
10a	Raman scattering at RT for partially deuterated crystals: see	85Lat
13a	NMR: Table 67B-1-009; Fig. 67B-1-038.	
14a	Relation between Bragg reflection intensity and spontaneous polarization: see Fig. 67B-1-028 in subsection 5c.	

Table 67B-1-001. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 1.0, 0.94, 0.9). *x*, *y*, *z* and U_{eq} [Å^2] [96Suz1]. $U_{eq} = (1/3)\Sigma_i\Sigma_j U_{ij}\, a_i^* a_j^*\, a_i\, a_j$. U_{ij} are defined by eq. (d) in Introduction. *T* = RT.

		x	*y*	*z*	U_{eq}
ARS	Na	0.2699(1)	0.4917(1)	0.4827(1)	0.0268(4)
	N(1)	0.0	0.0	0.0480(6)	0.0541(19)
	N(2)	0.0	0.5	0.1382(5)	0.0292(12)
	C(1)	0.1568(1)	0.1877(1)	0.2825(3)	0.0197(8)
	C(2)	0.1311(1)	0.2729(1)	0.4194(3)	0.0192(8)
	C(3)	0.1803(1)	0.2628(1)	0.6435(3)	0.0198(8)
	C(4)	0.1540(2)	0.3494(1)	0.7781(3)	0.0231(8)
	O(1)	0.1220(1)	0.1104(1)	0.3508(2)	0.0246(6)
	O(2)	0.2097(1)	0.2010(1)	0.1146(2)	0.0310(6)
	O(3)	0.2312(1)	0.4039(1)	0.8161(2)	0.0337(7)
	O(4)	0.0556(1)	0.3591(1)	0.8360(2)	0.0341(7)
	O(5)	0.1719(1)	0.3553(1)	0.3235(2)	0.0264(7)
	O(6)	0.2954(1)	0.2469(1)	0.6299(4)	0.0271(7)
	O(7)	0.3908(1)	0.0831(1)	0.4800(3)	0.0350(8)
	O(8)	0.2420(1)	0.0402(1)	0.8782(2)	0.0408(8)
	O(9)	0.4333(2)	0.2969(2)	0.0478(3)	0.0544(11)
	O(10)	0.4251(2)	0.3970(1)	0.4291(4)	0.0602(12)
$RS_{0.06}ARS_{0.94}$	Na	0.2698(1)	0.4917(1)	0.4821(1)	0.0265(4)
	N(1)*	0.0	0.0	0.0495(39)	0.0518(128)
	N(2)†	0.0	0.5	0.1430(16)	0.0282(58)
	C(1)	0.1561(2)	0.1876(1)	0.2827(3)	0.0188(10)
	C(2)	0.1305(2)	0.2731(1)	0.4196(3)	0.0181(10)
	C(3)	0.1805(2)	0.2628(1)	0.6434(3)	0.0190(10)
	C(4)	0.1540(2)	0.3495(2)	0.7782(3)	0.0222(11)
	O(1)	0.1219(1)	0.1103(1)	0.3508(2)	0.0243(8)
	O(2)	0.2096(1)	0.2011(1)	0.1145(2)	0.0298(8)
	O(3)	0.2315(1)	0.4039(1)	0.8154(2)	0.0335(8)
	O(4)	0.0556(1)	0.3592(1)	0.8372(3)	0.0334(9)
	O(5)	0.1718(1)	0.3554(1)	0.3233(2)	0.0259(9)
	O(6)	0.2953(1)	0.2469(1)	0.6294(3)	0.0276(9)
	O(7)	0.3910(2)	0.0831(1)	0.4795(3)	0.0340(10)
	O(8)	0.2427(2)	0.0400(1)	0.8785(3)	0.0409(10)
	O(9)	0.4331(2)	0.2971(2)	0.0473(4)	0.0549(14)
	O(10)	0.4250(2)	0.3974(2)	0.4279(4)	0.0603(15)
$RS_{0.1}ARS_{0.9}$	Na	0.2699(1)	0.4918(1)	0.4823(2)	0.0272(6)
	N(1)‡	0.0	0.0	0.0480(30)	0.0688(127)
	N(2)§	0.0	0.5	0.1443(24)	0.0269(83)
	C(1)	0.1567(2)	0.1873(2)	0.2824(4)	0.0202(14)
	C(2)	0.1299(2)	0.2732(2)	0.4195(4)	0.0186(13)
	C(3)	0.1801(2)	0.2629(2)	0.6435(5)	0.0206(14)
	C(4)	0.1538(3)	0.3493(2)	0.7784(4)	0.0239(15)
	O(1)	0.1218(1)	0.1104(1)	0.3507(3)	0.0250(10)
	O(2)	0.2097(2)	0.2010(1)	0.1146(3)	0.0305(10)
	O(3)	0.2317(1)	0.4040(1)	0.8153(3)	0.0340(11)
	O(4)	0.0552(2)	0.3592(1)	0.8374(3)	0.0331(11)
	O(5)	0.1712(2)	0.3556(1)	0.3227(3)	0.0269(11)
	O(6)	0.2957(2)	0.2469(1)	0.6298(4)	0.0269(11)
	O(7)	0.3913(2)	0.0831(1)	0.4795(4)	0.0341(13)
	O(8)	0.2428(2)	0.0401(1)	0.8792(3)	0.0421(13)
	O(9)	0.4336(3)	0.2973(2)	0.0462(5)	0.0556(18)
	O(10)	0.4253(2)	0.3976(2)	0.4269(5)	0.0613(19)

*: α = 0.94, †: β = 0.92, ‡: α = 0.90, §: β = 0.93. α, β: occupancies of N atoms.

Table 67B-1-002. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). *x*, *y*, *z* and U_{eq} [$\cdot 10^{-4}$ Å^2] [96Suz2]. $U_{eq} = (1/3)\Sigma_i\Sigma_j U_{ij}\, a_i^* a_j^*\, a_i\, a_j$. U_{ij} are defined by eq. (d) in Introduction. α, β: occupancies of N atoms. T = 213 K (1st row), 253 K (2nd row), 293 K (3rd row).

	x	*y*	*z*	U_{eq}	α/β
Na	0.26861(7)	0.49267(6)	0.47909(12)	194(4)	
	0.26889(8)	0.49286(7)	0.47729(15)	228(5)	
	0.26904(8)	0.49290(7)	0.47620(12)	266(5)	
KN(1)	0.0	0.0	0.0449(2)	425(8)	0.384(3)
	0.0	0.0	0.0462(3)	507(11)	0.393(4)
	0.0	0.0	0.0474(3)	588(12)	0.387(3)
KN(2)	0.0	0.5	0.1620(1)	218(4)	0.159(2)
	0.0	0.5	0.1608(2)	255(5)	0.173(3)
	0.0	0.5	0.1599(2)	289(5)	0.187(2)
C(1)	0.1550(2)	0.1883(1)	0.2835(3)	145(9)	
	0.1544(2)	0.1881(2)	0.2849(4)	167(11)	
	0.1543(2)	0.1882(2)	0.2846(4)	185(11)	
C(2)	0.1254(2)	0.2737(1)	0.4230(3)	145(9)	
	0.1255(2)	0.2735(2)	0.4235(4)	163(11)	
	0.1255(2)	0.2736(2)	0.4240(4)	176(10)	
C(3)	0.1787(2)	0.2638(1)	0.6459(3)	143(8)	
	0.1788(2)	0.2639(2)	0.6453(4)	171(10)	
	0.1789(2)	0.2641(2)	0.6452(4)	194(11)	
C(4)	0.1529(2)	0.3516(2)	0.7811(3)	184(10)	
	0.1536(3)	0.3517(2)	0.7805(4)	215(12)	
	0.1540(3)	0.3524(2)	0.7787(4)	242(12)	
O(1)	0.1203(1)	0.1088(1)	0.3508(3)	181(7)	
	0.1203(2)	0.1091(1)	0.3521(3)	216(9)	
	0.1201(2)	0.1093(1)	0.3527(3)	253(9)	
O(2)	0.2095(1)	0.2032(1)	0.1171(2)	218(8)	
	0.2087(2)	0.2027(1)	0.1178(3)	256(9)	
	0.2085(2)	0.2025(1)	0.1183(3)	303(10)	
O(3)	0.2333(2)	0.4060(1)	0.8149(2)	267(8)	
	0.2341(2)	0.4059(1)	0.8134(3)	308(10)	
	0.2346(2)	0.4059(1)	0.8119(3)	347(10)	
O(4)	0.0538(2)	0.3620(1)	0.8445(3)	270(8)	
	0.0546(2)	0.3626(1)	0.8435(3)	308(10)	
	0.0554(2)	0.3630(1)	0.8424(3)	355(10)	
O(5)	0.1650(1)	0.3575(1)	0.3246(3)	193(7)	
	0.1645(2)	0.3571(1)	0.3242(3)	221(9)	
	0.1641(2)	0.3570(1)	0.3240(3)	258(9)	
O(6)	0.2963(2)	0.2478(1)	0.6291(3)	218(8)	
	0.2958(2)	0.2478(2)	0.6278(4)	249(10)	
	0.2954(2)	0.2479(1)	0.6272(3)	281(10)	
O(7)	0.3951(2)	0.0825(2)	0.4797(4)	245(9)	
	0.3949(2)	0.0824(2)	0.4800(4)	289(12)	
	0.3952(2)	0.0827(2)	0.4814(4)	337(12)	
O(8)	0.2466(2)	0.0410(1)	0.8825(3)	410(11)	
	0.2476(3)	0.0414(2)	0.8844(3)	444(13)	
	0.2482(3)	0.0414(2)	0.8855(3)	511(14)	
O(9)	0.4375(2)	0.3003(2)	0.0383(4)	448(13)	
	0.4365(3)	0.2989(3)	0.0397(5)	523(18)	
	0.4357(3)	0.2976(3)	0.0397(5)	600(19)	
O(10)	0.4238(2)	0.3957(2)	0.4222(5)	481(14)	
	0.4250(2)	0.3977(2)	0.4181(5)	562(18)	
	0.4253(2)	0.3986(2)	0.4149(5)	637(18)	

Table 67B-1-003. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). Interatomic distances [Å] around Na atoms [96Suz2]. T = 213 K, 253 K, 293 K.

	213 K	253 K	293 K
Na–O(1^i)	2.369(2)	2.374(2)	2.382(2)
Na–O(3)	2.461(2)	2.468(2)	2.469(2)
Na–O(5)	2.483(2)	2.494(2)	2.502(2)
Na–O(7^i)	2.346(2)	2.351(3)	2.362(3)
Na–O(8^i)	2.356(2)	2.363(2)	2.367(2)
Na–O(10)	2.335(3)	2.335(3)	2.336(3)
Mean	2.392	2.398	2.403

Symmetry code: (i) 1/2 – x, 1/2 + y, 1 – z.

Table 67B-1-004. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 1.0, 0.94, 0.90). Interatomic distances [Å] around Na atoms [96Suz1]. T = RT.

	ARS	$RS_{0.06}ARS_{0.94}$	$RS_{0.10}ARS_{0.90}$
Na–O(1^i)	2.401(2)	2.398(2)	2.397(2)
Na–O(3)	2.485(2)	2.479(2)	2.477(2)
Na–O(5)	2.512(2)	2.504(2)	2.508(2)
Na–O(7^i)	2.376(2)	2.371(3)	2.374(3)
Na–O(8^i)	2.367(2)	2.360(2)	2.366(3)
Na–O(10)	2.361(3)	2.352(3)	2.353(3)
Mean	2.417	2.411	2.413

Symmetry code: (i) 1/2 – x, 1/2 + y, 1 – z.

Table 67B-1-005. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). Interatomic distances [Å] around K/N(1) and K/N(2) [96Suz2]. T = 213 K, 253 K, 293 K.

	213 K	253 K	293 K
KN(1)–O(1)	2.841(2)	2.849(2)	2.851(2)
KN(1^i)–O(9)	2.990(3)	3.021(4)	3.047(4)
KN(1)–O(8^{ii})	3.157(3)	3.176(3)	3.190(3)
KN(1)–O(10^{iii})	3.384(3)	3.359(3)	3.347(3)
KN(2)–O(4^{ii})	2.860(2)	2.860(2)	2.864(2)
KN(2^{iv})–O(7)	2.810(2)	2.820(3)	2.823(3)
KN(2)–O(5)	3.003(2)	3.010(2)	3.015(2)
KN(2)–O(8^v)	3.085(3)	3.081(3)	3.081(3)

Symmetry codes: (i) 1/2 + x, 1/2 – y, – z;
(ii) x, y, 1 + z;
(iii) –1/2 + x, 1/2 – y, – z;
(iv) 1/2 + x, 1/2 – y, 1 – z;
(v) 1/2 – x, 1/2 + y, 1 – z.

Table 67B-1-006. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 1.0, 0.94, 0.90). Interatomic distances [Å] and geometry of hydrogen bonds around the two kinds of K/NH_4 ions sites [96Suz1]. *T* = RT.

	N…O [Å]	N–H [Å]	O…H [Å]	N–H...O [°]
ARS				
N(1)–H(N1a)...O(1)	2.889(3)	0.90(3)	2.00(3)	171(3)
N(1)–H(N1b^{i})...O(9^{i})	3.104(3)	0.74(5)	2.50(4)	141(4)
N(2)–H(N2a)...O(4^{ii})	2.859(3)	1.00(3)	1.86(3)	174(2)
N(2)–H(N2b^{iii})...O(7^{iii})	2.985(3)	0.78(3)	2.31(3)	145(2)
N(1)–O(8^{ii})	3.192(2)			
N(1)–O(10^{iv})	3.456(4)			
N(2)–O(5)	3.181(2)			
N(2)–O(8^{v})	3.204(2)			
$RS_{0.06}ARS_{0.94}$				
KN(1)–H(N1a)...O(1)	2.875(16)	0.77(4)	2.10(4)	175(5)
KN(1)–H(N1b^{i})...O(9^{i})	3.097(6)	0.54(6)	2.80(7)	119(8)
KN(2)–H(N2a)...O(4^{ii})	2.867(7)	0.89(3)	1.98(3)	172(3)
KN(2)–H(N2b^{iii})...O(7^{iii})	2.957(8)	0.77(4)	2.27(3)	151(3)
KN(1)–O(8^{ii})	3.193(9)			
KN(1)–O(10^{iv})	3.449(21)			
KN(2)–O(5)	3.160(4)			
KN(2)–O(8^{v})	3.187(3)			
$RS_{0.10}ARS_{0.90}$				
KN(1)–H(N1a)...O(1)	2.880(12)	1.00(5)	1.89(5)	169(4)
KN(1)–H(N1b^{i})...O(9^{i})	3.089(5)	0.87(9)	2.49(9)	127(7)
KN(2)–H(N2a)...O(4^{ii})	2.870(10)	0.88(4)	2.00(3)	177(3)
KN(2)–H(N2b^{iii})...O(7^{iii})	2.948(12)	0.78(4)	2.28(4)	144(3)
KN(1)–O(8^{ii})	3.189(7)			
KN(1)–O(10^{iv})	3.433(16)			
KN(2)–O(5)	3.148(6)			
KN(2)–O(8^{v})	3.185(3)			

Symmetry codes: (i) $-1/2 + x$, $1/2 - y$, $-z$;
(ii) x, y, $-1 + z$;
(iii) $-1/2 + x$, $1/2 - y$, $1 - z$;
(iv) $-1/2 + x$, $1/2 - y$, $-z$;
(v) $1/2 - x$, $1/2 + y$, $1 - z$.

Table 67B-1-007. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 1.0, 0.94, 0.90). Interatomic distances [Å] and angles [°] of oxygen and hydrogen atoms creating hydrogen bonds [96Suz1]. T = RT.

	O–H	O...O	O...H	O–H...O
ARS				
O(6)–H(6)...O(10)	0.69(3)	2.964(3)	2.33(3)	153(3)
O(7)–H(7a)...O(6)	0.81(3)	2.799(2)	2.00(3)	171(2)
O(7)–H(7b)...$O(4^{i})$	0.80(3)	2.940(2)	2.16(3)	163(2)
O(8)–H(8a)...$O(3^{ii})$	0.73(3)	2.763(2)	2.04(3)	171(3)
O(8)–H(8b)...$O(2^{iii})$	0.85(3)	2.782(2)	1.94(3)	175(2)
O(9)–H(9a)...O(2)	0.69(4)	3.089(3)	2.51(4)	143(5)
O(9)–H(9b)...$O(4^{i})$	0.88(3)	2.800(3)	1.92(3)	171(2)
O(10)–H(10a)...O(9)	0.83(3)	2.790(3)	1.96(3)	172(2)
O(10)–H(10b)...$O(1^{i})$	0.71(3)	2.771(3)	2.07(2)	170(3)
	Angle [°]			
H(7a)–O(7)–H(7b)	105(3)			
H(8a)–O(8)–H(8b)	109(3)			
H(9a)–O(9)–H(9b)	95(4)			
H(10a)–O(10)–H(10b)	101(3)			
$RS_{0.06}ARS_{0.94}$				
O(6)–H(6)...O(10)	0.60(3)	2.964(3)	2.44(3)	145(5)
O(7)–H(7a)...O(6)	0.74(4)	2.795(3)	2.06(3)	173(4)
O(7)–H(7b)...$O(4^{i})$	0.91(3)	2.934(3)	2.05(4)	165(3)
O(8)–H(8a)...$O(3^{ii})$	0.79(3)	2.756(2)	1.98(3)	168(4)
O(8)–H(8b)...$O(2^{iii})$	0.80(3)	2.780(2)	1.98(3)	178(3)
O(9)–H(9a)...O(2)	0.76(4)	3.081(3)	2.48(4)	137(4)
O(9)–H(9b)...$O(4^{i})$	0.84(3)	2.797(3)	1.96(3)	173(3)
O(10)–H(10a)...O(9)	0.92(3)	2.782(4)	1.88(3)	168(2)
O(10)–H(10b)...$O(1^{i})$	0.70(4)	2.768(3)	2.07(4)	172(3)
	Angle [°]			
H(7a)–O(7)–H(7b)	101(4)			
H(8a)–O(8)–H(8b)	103(3)			
H(9a)–O(9)–H(9b)	110(4)			
H(10a)–O(10)–H(10b)	98(3)			
$RS_{0.10}ARS_{0.90}$				
O(6)–H(6)...O(10)	0.75(3)	2.968(4)	2.29(4)	151(4)
O(7)–H(7a)...O(6)	0.87(3)	2.795(3)	1.94(3)	167(3)
O(7)–H(7b)...$O(4^{i})$	0.85(4)	2.928(3)	2.13(4)	158(3)
O(8)–H(8a)...$O(3^{ii})$	0.76(3)	2.753(3)	2.00(3)	174(4)
O(8)–H(8b)...$O(2^{iii})$	0.88(4)	2.775(3)	1.89(3)	177(3)
O(9)–H(9a)...O(2)	0.73(4)	3.087(5)	2.45(4)	147(5)
O(9)–H(9b)...$O(4^{i})$	0.76(3)	2.794(4)	2.05(4)	168(4)
O(10)–H(10a)...O(9)	0.88(3)	2.783(5)	1.92(5)	169(4)
O(10)–H(10b)...$O(1^{i})$	0.71(4)	2.766(4)	2.07(4)	168(4)
	Angle [°]			
H(7a)–O(7)–H(7b)	96(3)			
H(8a)–O(8)–H(8b)	108(4)			
H(9a)–O(9)–H(9b)	103(4)			
H(10a)–O(10)–H(10b)	96(4)			

Symmetry codes: (i) $1/2 + x, 1/2 - y, 1 - z$;
(ii) $1/2 - x, -1/2 + y, 2 - z$;
(iii) $x, y, 1 + z$.

Table 67B-1-008. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). Bond lengths [Å] and angles [°] of tartrate molecule [96Suz2]. *T* = 213 K, 253 K, 293 K.

	213 K	253 K	293 K
C(1)–C(2)	1.536(3)	1.535(3)	1.538(3)
C(2)–C(3)	1.529(3)	1.526(3)	1.525(3)
C(3)–C(4)	1.539(3)	1.541(3)	1.542(3)
C(1)–O(1)	1.278(2)	1.270(3)	1.274(3)
C(1)–O(2)	1.239(2)	1.242(3)	1.239(3)
C(2)–O(5)	1.423(2)	1.424(3)	1.424(3)
C(2)–H(2)	1.07(2)	1.06(3)	1.07(2)
C(3)–O(6)	1.421(3)	1.419(3)	1.417(3)
C(3)–H(3)	1.08(2)	1.03(3)	1.05(2)
C(4)–O(3)	1.250(3)	1.251(3)	1.249(3)
C(4)–O(4)	1.252(3)	1.254(4)	1.253(4)
O(2)–O(5)	2.605(2)	2.607(2)	2.610(2)
O(5)–H(5)	0.78(3)	0.80(4)	0.78(3)
H(5)...O(2)	2.11(3)	2.06(3)	2.12(3)
O(3)–O(6)	2.645(2)	2.643(3)	2.641(3)
O(6)–H(6)	0.69(4)	0.65(4)	0.65(4)
H(6)...O(3)	2.46(4)	2.38(5)	2.36(4)
O(1)–C(1)–O(2)	126.5(2)	126.3(2)	126.4(2)
O(1)–C(1)–C(2)	116.5(2)	116.8(2)	116.4(2)
O(2)–C(1)–C(2)	117.0(2)	116.9(2)	117.2(2)
C(1)–C(2)–O(5)	110.4(2)	110.5(2)	110.3(2)
C(1)–C(2)–C(3)	110.0(2)	110.0(2)	110.2(2)
O(5)–C(2)–C(3)	109.2(2)	109.4(2)	109.6(2)
C(2)–O(5)–H(5)	108(2)	105(2)	109(2)
C(2)–C(3)–O(6)	110.9(2)	110.8(2)	110.8(2)
C(2)–C(3)–C(4)	109.6(2)	109.8(2)	109.5(2)
O(6)–C(3)–C(4)	111.6(2)	111.5(2)	111.5(2)
C(3)–O(6)–H(6)	121(3)	116(4)	115(4)
C(3)–C(4)–O(4)	117.1(2)	117.1(2)	116.8(2)
C(3)–C(4)–O(3)	116.4(2)	116.3(2)	116.3(2)
O(3)–C(4)–O(4)	126.5(2)	126.6(2)	126.8(2)
O(5)–H(5)...O(2)	121(5)	125(3)	121(3)
O(6)–H(6)...O(3)	98(4)	107(4)	109(4)

Table 67B-1-009. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Minimum proton spin-lattice relaxation time $(T_1)_{min}$ [74Mor]. Parameter: x.

x	Rotational axis	$(T_1)_{min}$ [10^{-3} s]
0.10	Threefold	260
	Twofold	280
0.20	Threefold	160
	Twofold	180
0.40	Threefold	75
	Twofold	74
0.60	Threefold	46
	Twofold	47
0.87	Threefold	24
	Twofold	60, 60
0.91	Threefold	23
	Twofold	54, 55
1.00	Threefold	18
	Twofold	40, 40

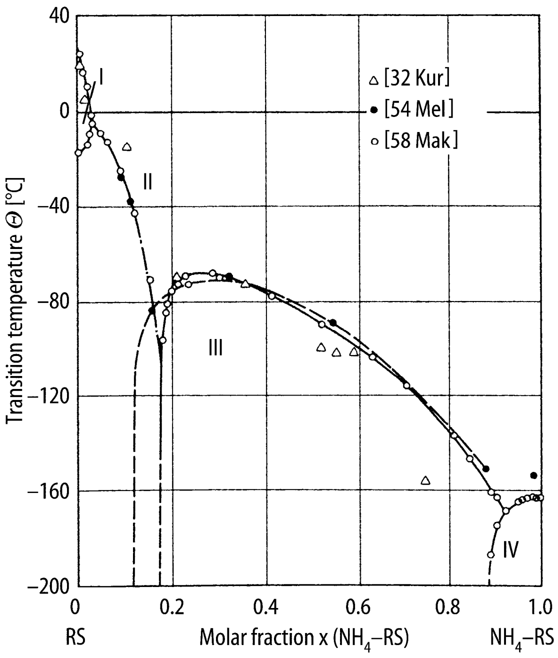

Fig. 67B-1-001. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Θ vs. x [58Mak]. The dashed-dotted line shows the temperature where the broad dielectric peak appears.

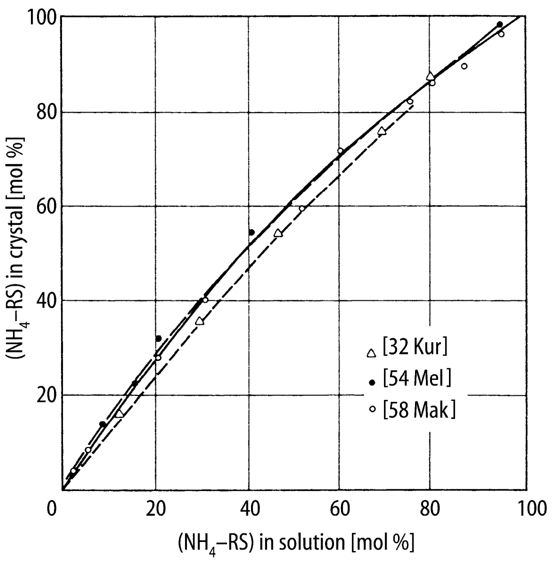

Fig. 67B-1-002. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Relation between the compositions of the crystals and the aqueous solutions [58Mak].

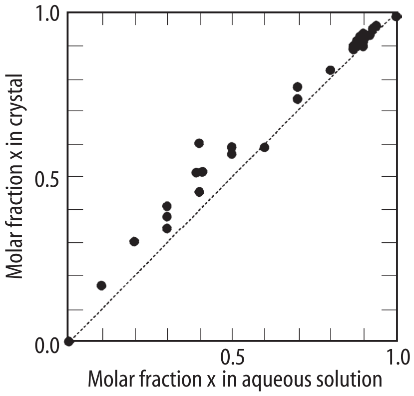

Fig. 67B-1-003. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Relation between the compositions of the crystal and the aqueous solution [94Shi].

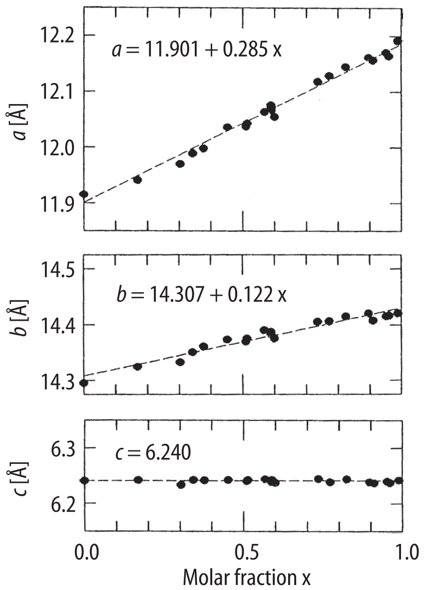

Fig. 67B-1-004. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. *a*, *b*, *c* vs. x [94Shi]. *T* = RT.

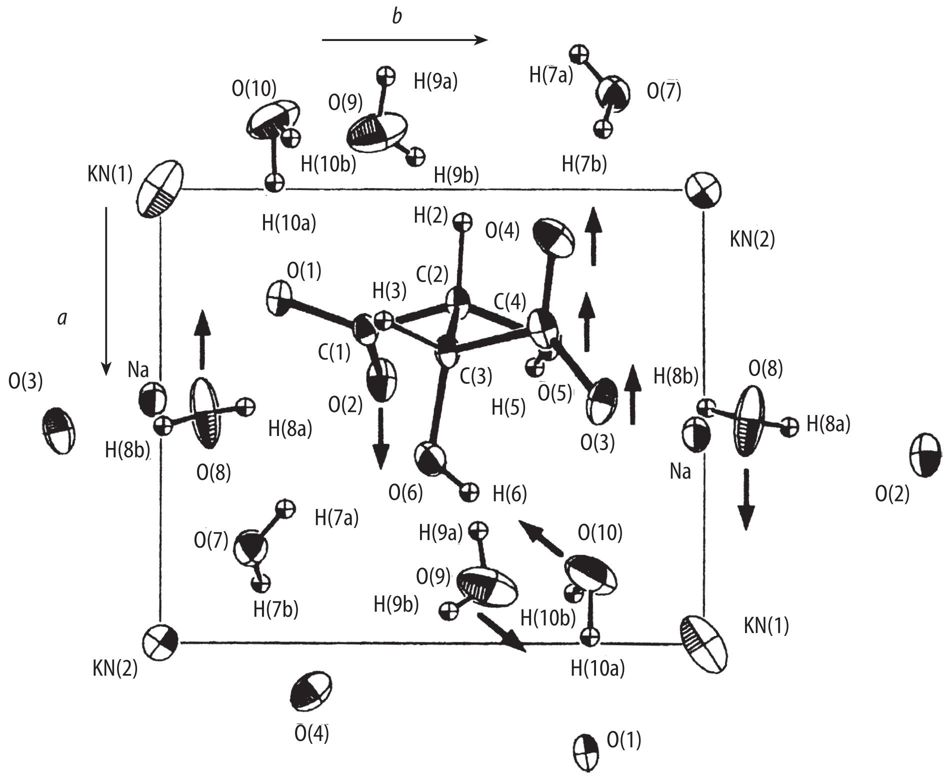

Fig. 67B-1-005. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). Structure of the paraelectric phase viewed along *c* [96Suz2]. *T* = 213 K. Arrows indicate the directions of the atomic displacements as the temperature lowers.

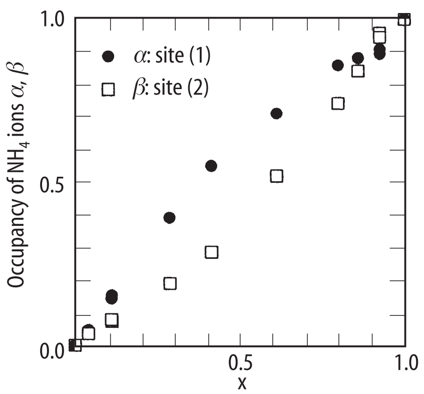

Fig. 67B-1-006. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. α, β vs. x [94Shi]. α, β: occupancies of NH_4 ions for the two independent crystallographic sites (1) and (2) [94Shi]. x = $(\alpha + \beta)/2$.

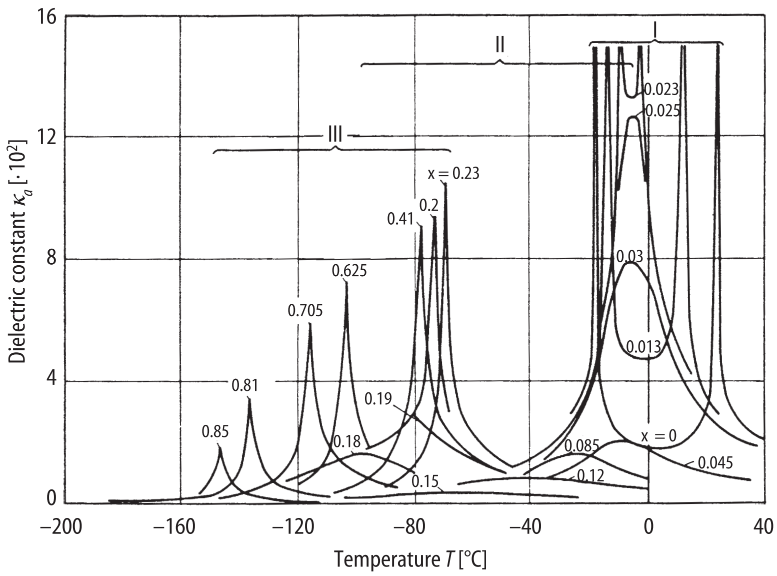

Fig. 67B-1-007. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. κ_a vs. T [58Mak]. Parameter: x. For regions I, II, III, see 1b and Fig. 67B-1-001.

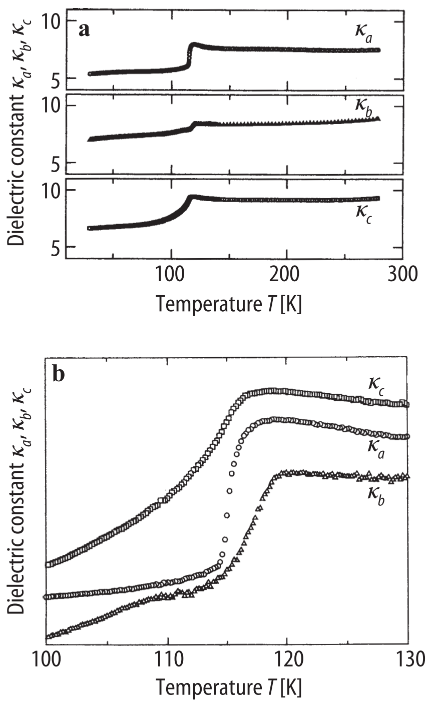

Fig. 67B-1-008. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.96). κ_a, κ_b, κ_c vs. T [95Nod]. (**a**) $20 \le T \le 270$ K, (**b**) $100 \le T \le 130$ K.

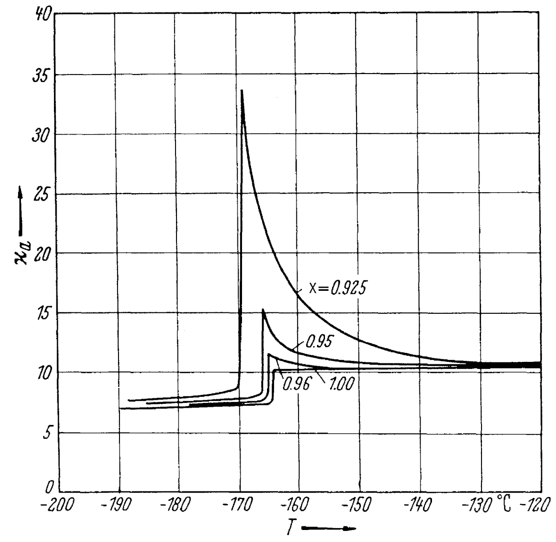

Fig. 67B-1-009. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. κ_a vs. T [58Mak]. Parameter: x. x ≥ 0.925.

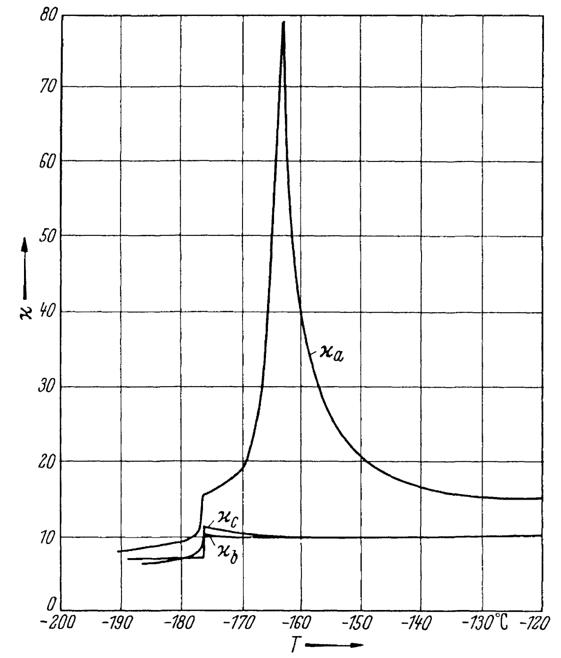

Fig. 67B-1-010. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.905). κ_a, κ_b, κ_c vs. T [58Mak].

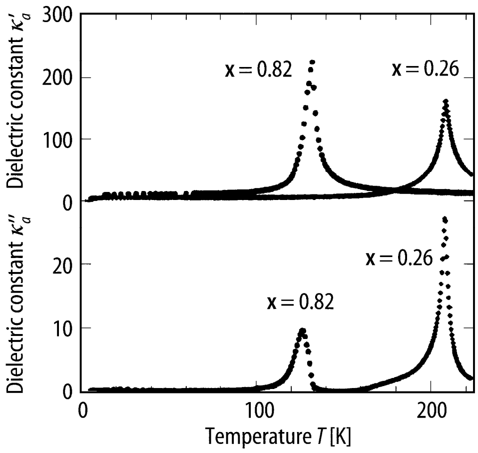

Fig. 67B-1-011. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.26, 0.82). κ_a', κ_a'' vs. T [98Noz]. f = 10 kHz.

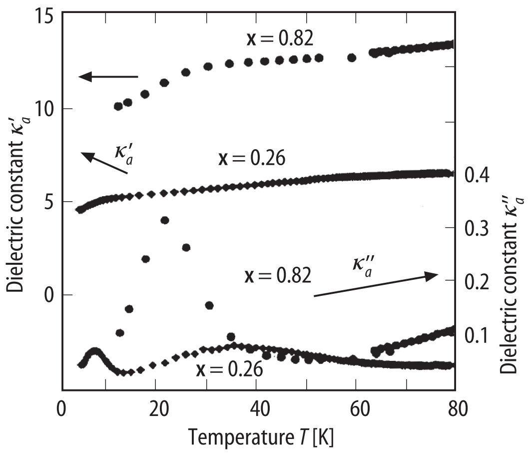

Fig. 67B-1-012. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.26, 0.82). κ_a', κ_a'' vs. T in low temperature region [98Noz]. f = 10 kHz.

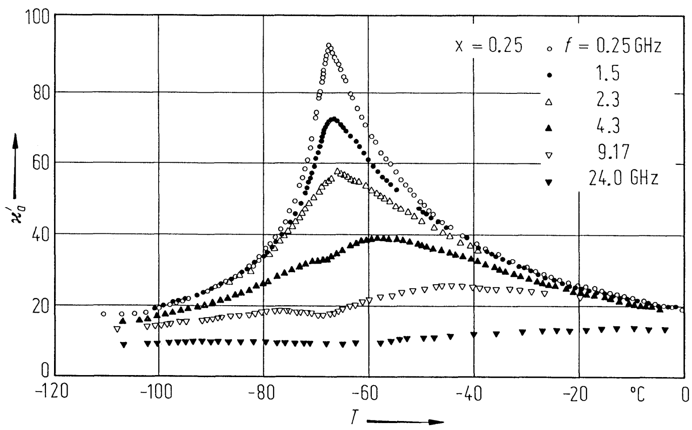

Fig. 67B-1-013. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.25). κ_a' vs. T [80Hor]. Parameter: f.

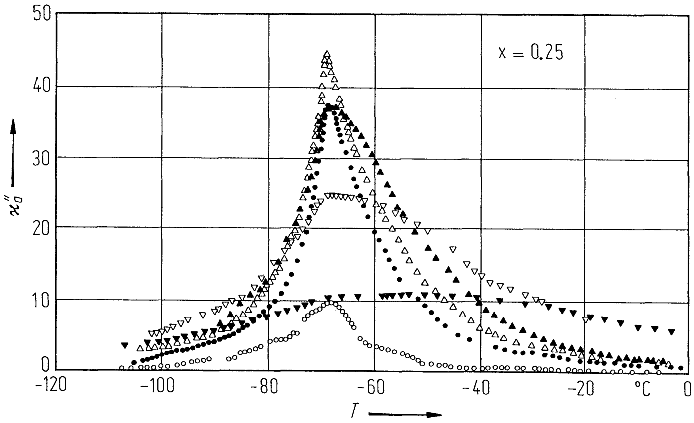

Fig. 67B-1-014. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.25). κ_a'' vs. *T* [80Hor]. Parameter: *f*. See Fig. 67B-1-013 for the symbols.

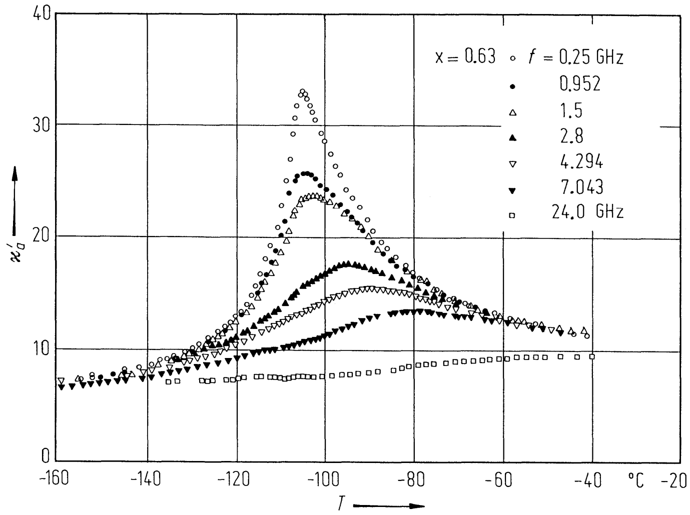

Fig. 67B-1-015. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.63). κ_a' vs. *T* [80Hor]. Parameter: *f*.

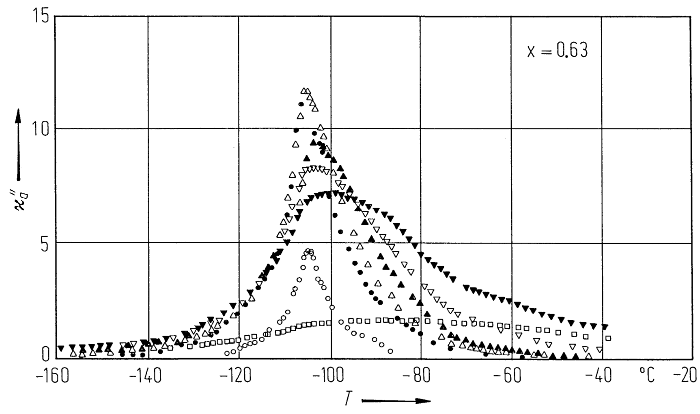

Fig. 67B-1-016. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.63). κ_a'' vs. *T* [80Hor]. Parameter: *f*. See Fig. 67B-1-015 for the symbols.

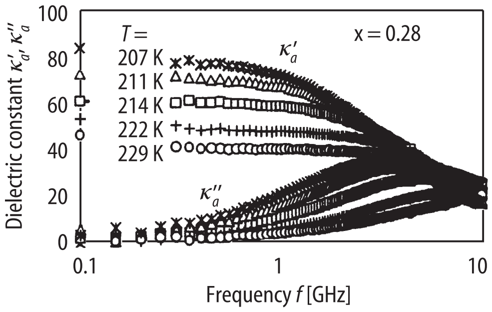

Fig. 67B-1-017. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). κ'_a, κ''_a vs. f [96Noz]. Parameter: T. Θ_{II-I} is about 206 K.

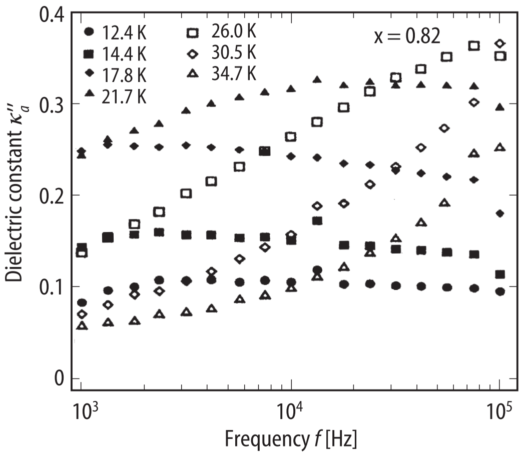

Fig. 67B-1-018. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.82). κ''_a vs. f [98Noz]. Parameter: T.

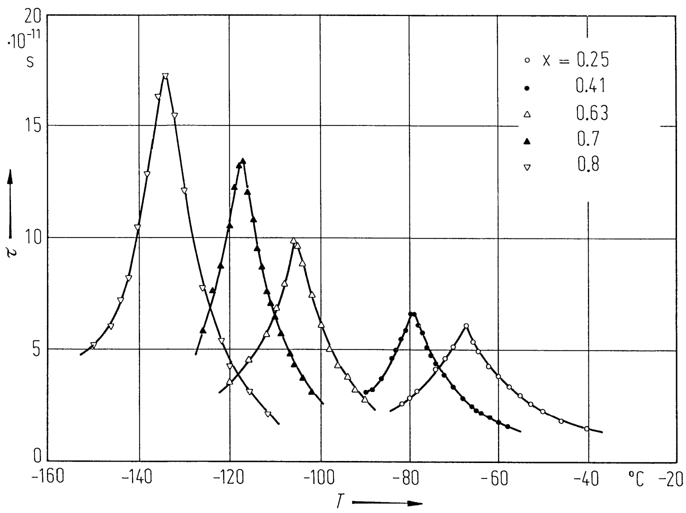

Fig. 67B-1-019. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. τ vs. T [80Hor]. Parameter: x. τ: dielectric relaxation time.

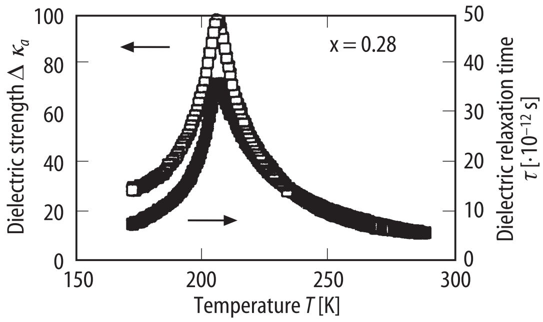

Fig. 67B-1-020. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.28). $\Delta\kappa_a$, τ vs. T [96Noz]. $\Delta\kappa_a = \kappa_s - \kappa_\infty$: dielectric strength. τ: dielectric relaxation time.

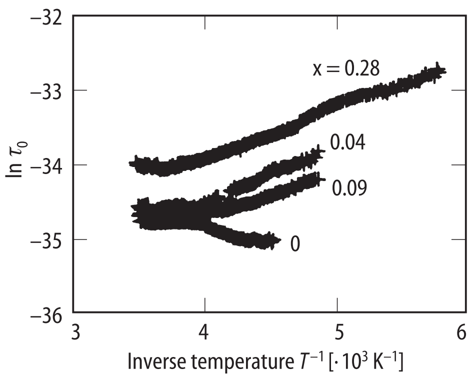

Fig. 67B-1-021. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0, 0.04, 0.09, 0.28). ln τ_0 vs. $1/T$ [96Noz]. Parameter: x. τ_0: relaxation time of the elementary process.

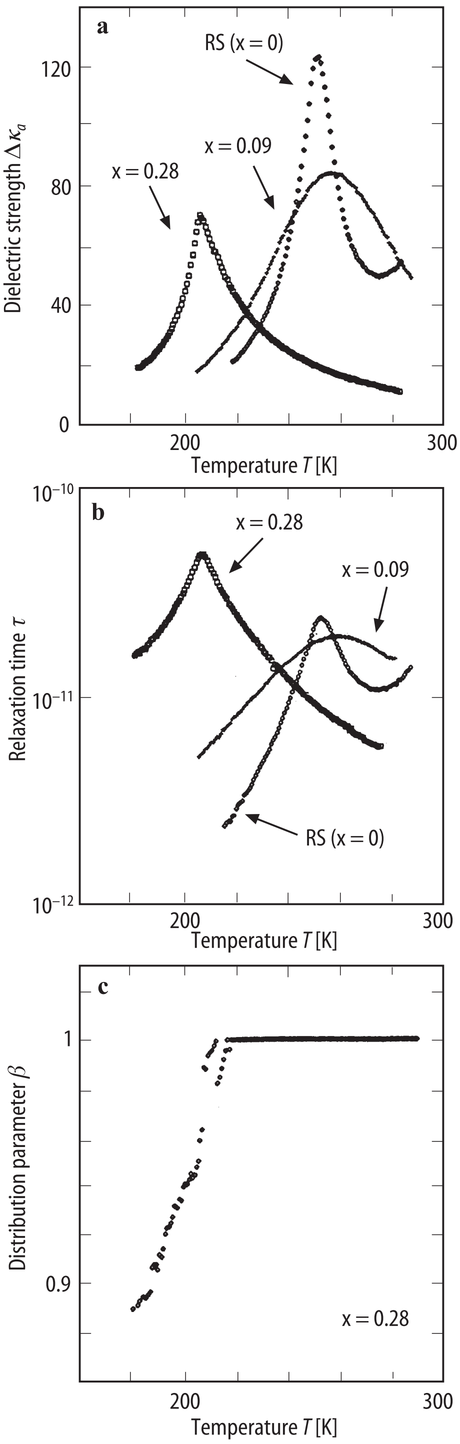

Fig. 67B-1-022. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0, 0.09, 0.28). $\Delta\kappa_a$, τ, β vs. T [96Oka]. (**a**) $\Delta\kappa_a$: dielectric strength. (**b**) τ: relaxation time. (**c**) β: distribution parameter of τ.

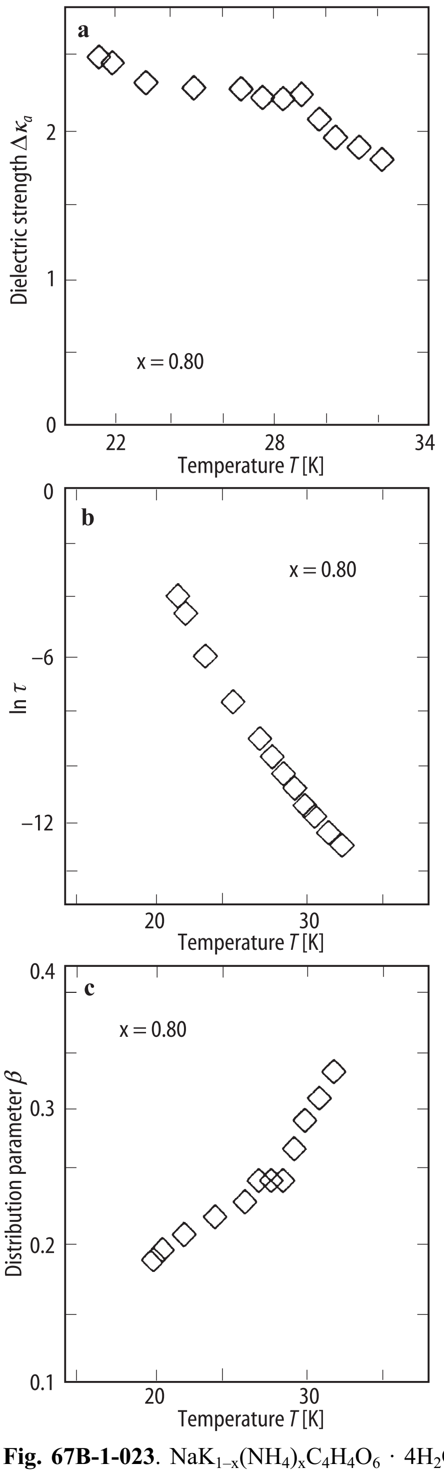

Fig. 67B-1-023. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.80). $\Delta\kappa_a$, ln τ, β vs. T [96Oka]. (**a**) $\Delta\kappa_a$: dielectric strength. (**b**) ln τ, τ: relaxation time. (**c**) β: distribution parameter of τ.

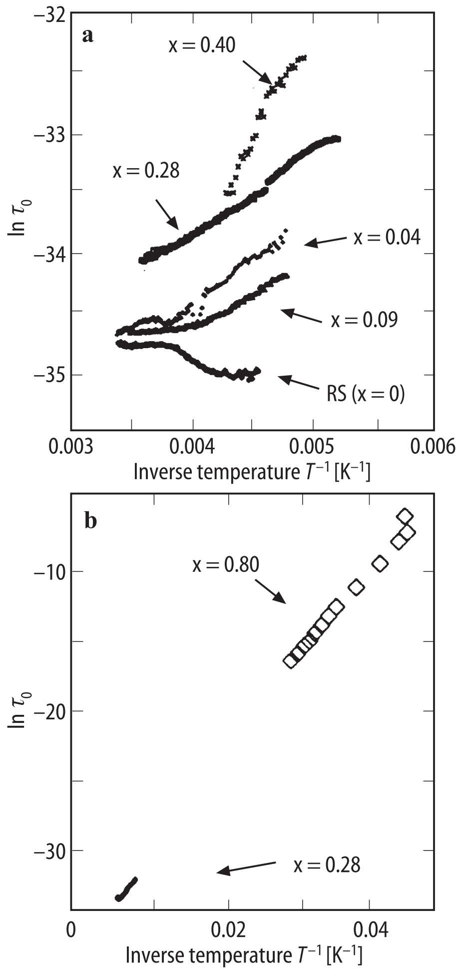

Fig. 67B-1-024. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. ln τ_0 vs. $1/T$ [96Oka]. Parameter: x. (**a**) x = 0, 0.04, 0.09, 0.28, 0.40, (**b**) x = 0.28, 0.80. τ_0: relaxation time of the elementary process.

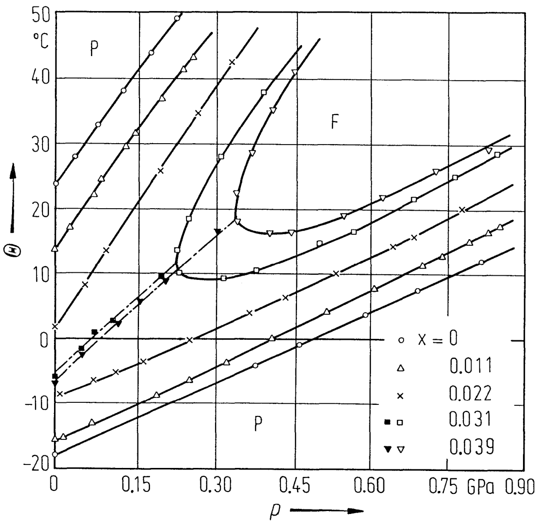

Fig. 67B-1-025. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Θ vs. p [80Ges]. Parameter: x. $0 \le x \le 0.039$. Solid squares and triangles show the peak positions of broad dielectric constant maxima.

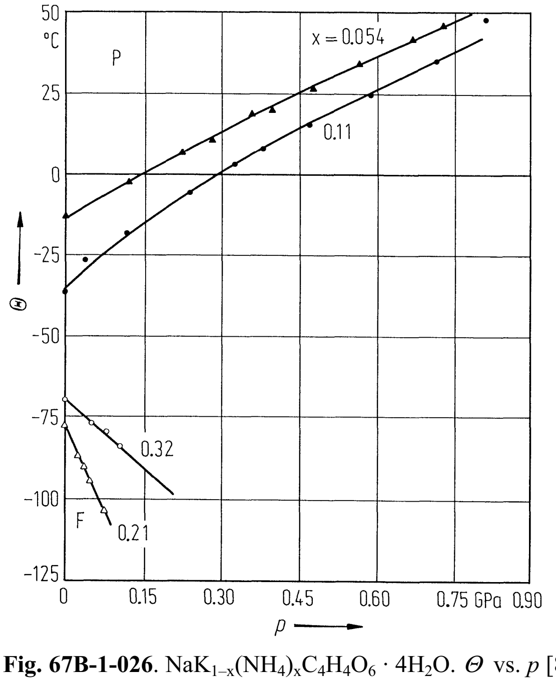

Fig. 67B-1-026. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Θ vs. p [80Ges]. Parameter: x. $0.054 \le x \le 0.32$. Solid symbols show the peak positions of broad dielectric constant maxima.

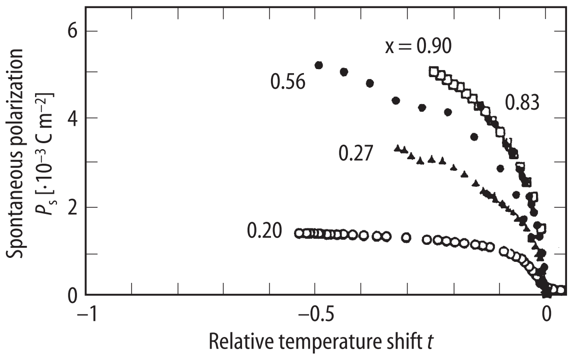

Fig. 67B-1-027. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. P_s vs. t [96Shi]. Parameter: x. $t = (T - \Theta)/\Theta$. Open circle: [58Mak], open square: [80Iva], full triangle, full circle, full square: [96Shi].

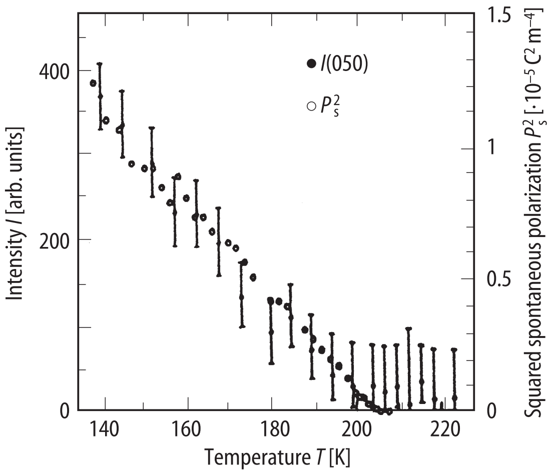

Fig. 67B-1-028. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.27, 0.28). P_s^2, $I(050)$ vs. T [96Shi]. I for x = 0.28, and P_s^2 for x = 0.27.

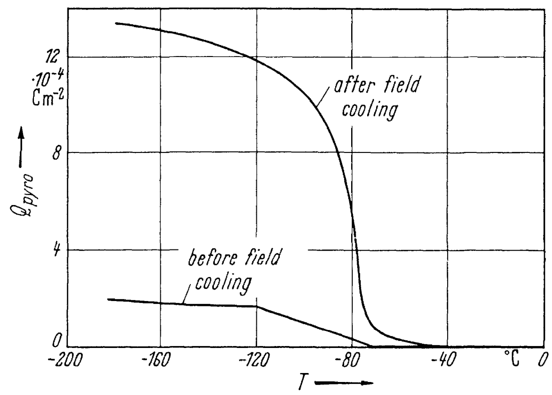

Fig. 67B-1-029. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.20). Q_{pyro} vs. T [58Mak]. Q_{pyro}: pyroelectric charge.

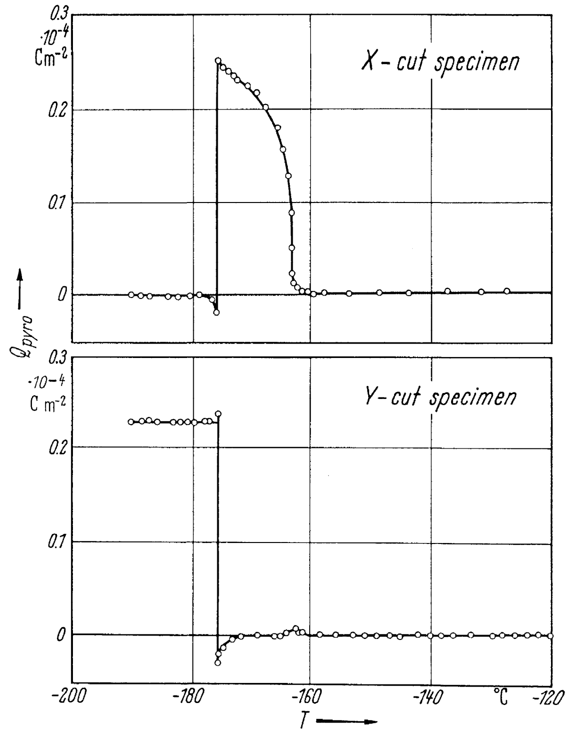

Fig. 67B-1-030. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.905). Q_{pyro} vs. T [58Mak]. Q_{pyro}: pyroelectric charge.

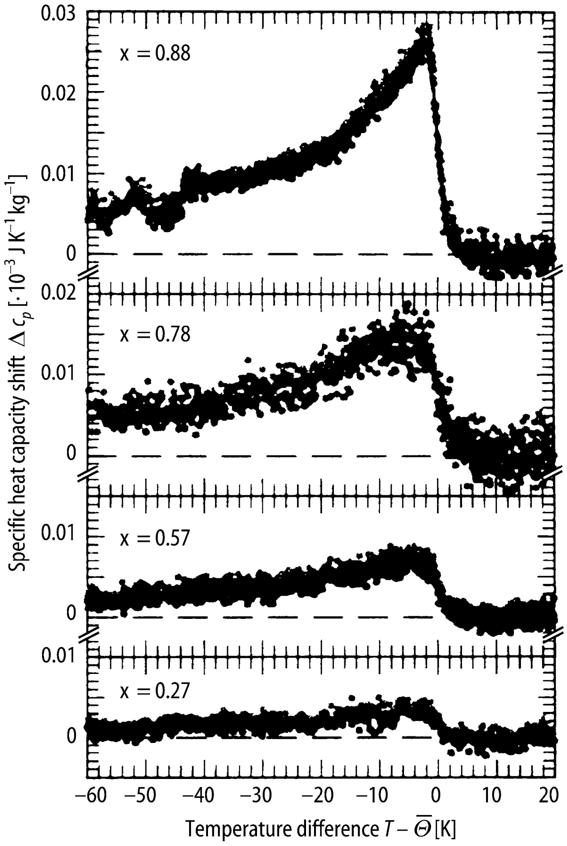

Fig. 67B-1-031. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. Δc_p vs. $T - \overline{\Theta}$ in the region III [99Nod]. Parameter: x. $\overline{\Theta}$: estimated mean value of Θ. $\overline{\Theta}$ = 206.8 K for x = 0.27, 178.6 K for x = 0.57, 141.9 K for x = 0.78 and 119.9 K for x = 0.88.

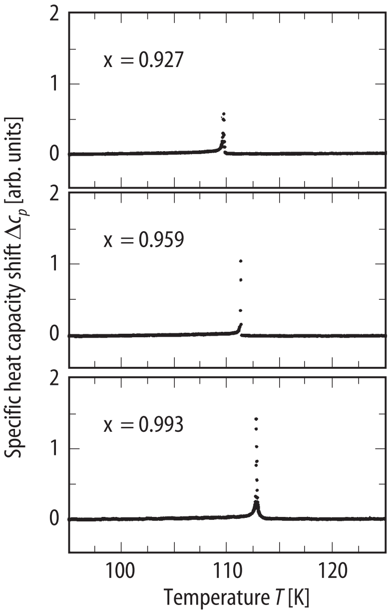

Fig. 67B-1-032. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.927, 0.959, 0.993). Δc_p vs. T in the region IV [95Nod].

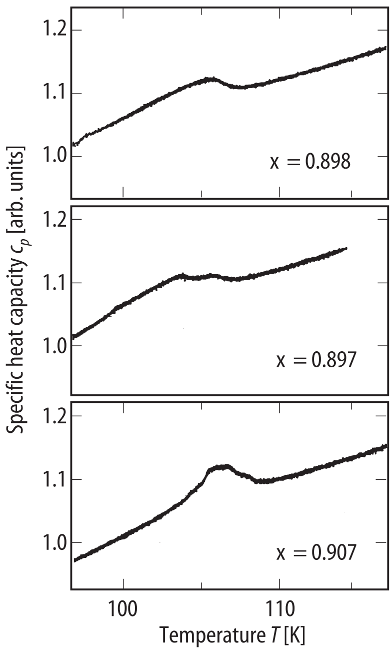

Fig. 67B-1-033. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$ (x = 0.898, 0.897, 0.907). c_p vs. T in the region of III–IV boundary [95Nod].

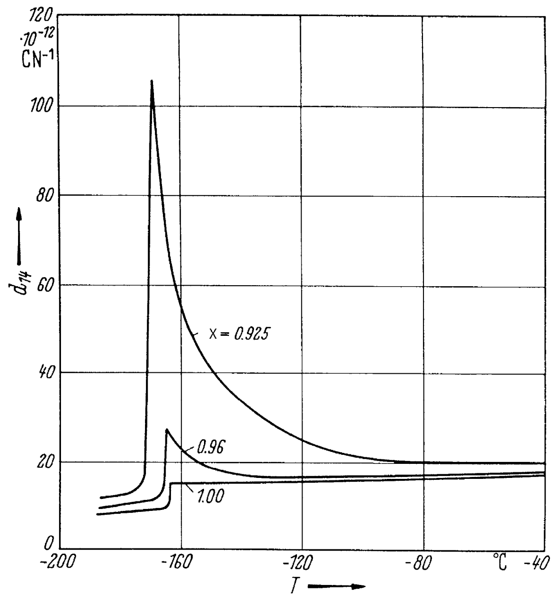

Fig. 67B-1-034. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. d_{14} vs. T [59Mak]. Parameter: x.

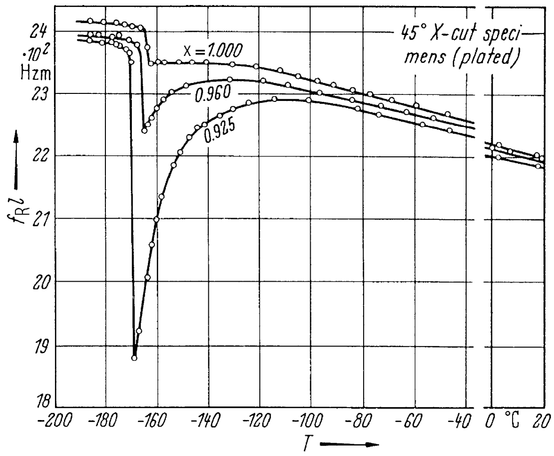

Fig. 67B-1-035. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. $f_R l$ vs. T [59Mak]. Parameter: x. $f_R l$: frequency constant of the 45° X-cut bar (see the caption of Fig. 67A-1-067 in No. 67A-1).

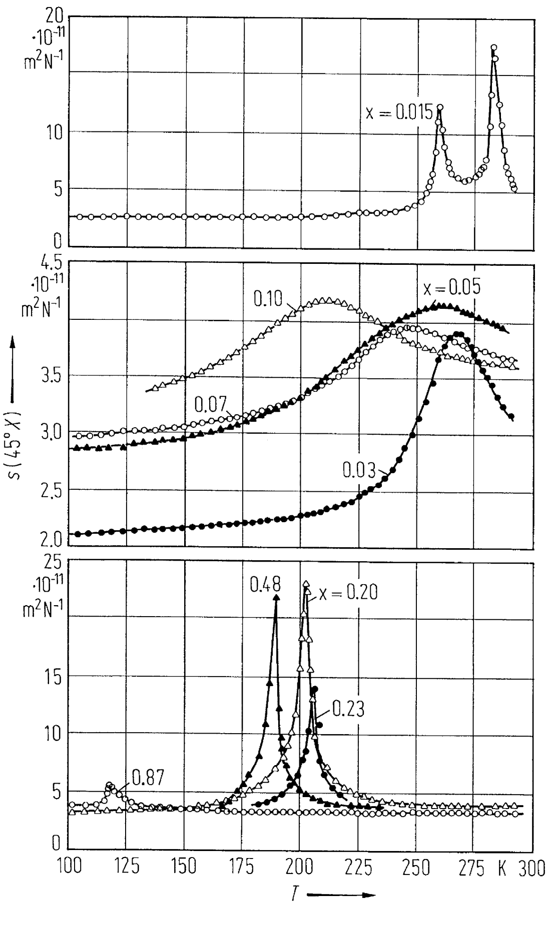

Fig. 67B-1-036. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. $s(45°\ X)$ vs. T [78Mae]. Parameter: x. $s(45°\ X)=(1/4)(s_{22}+s_{33}+2s_{23}+s^E_{44})$.

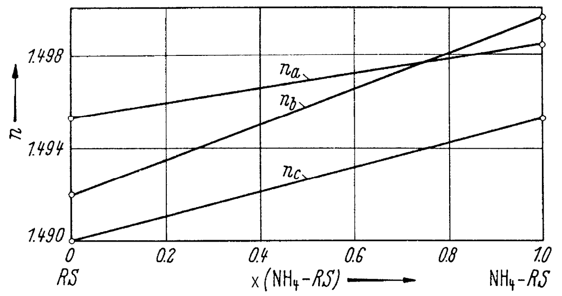

Fig. 67B-1-037. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. n_a, n_b, n_c vs. x [58Mak].

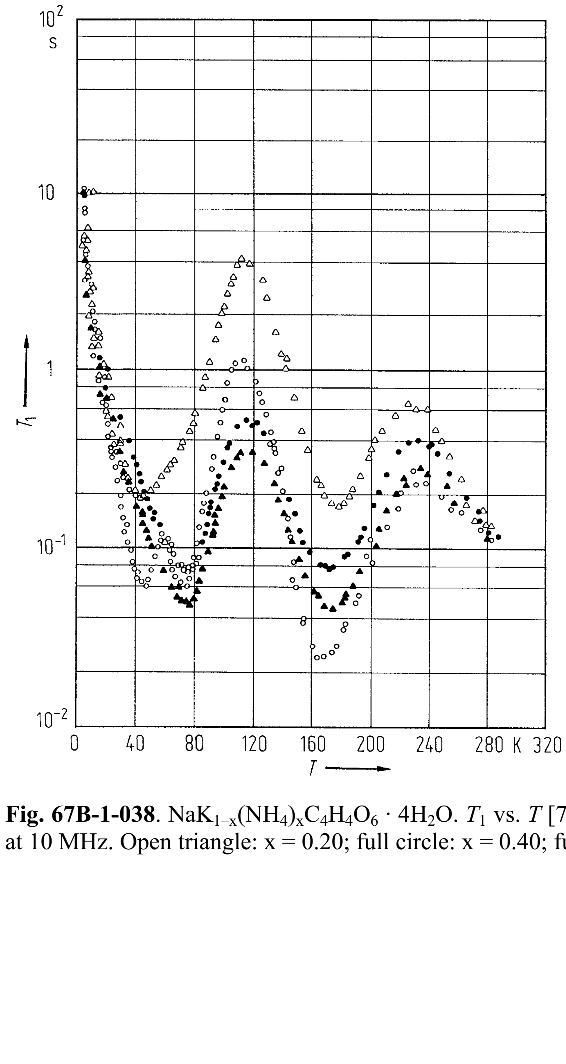

Fig. 67B-1-038. $NaK_{1-x}(NH_4)_xC_4H_4O_6 \cdot 4H_2O$. T_1 vs. T [74Mor]. Parameter: x. T_1: proton spin-lattice relaxation time at 10 MHz. Open triangle: x = 0.20; full circle: x = 0.40; full triangle: x = 0.60; open circle: x = 0.87.

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