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SYNTHESIS AND CRYSTAL STRUCTURE OF Na-A WITH OCCLUDED AgI AND Ag_4I_4 NANOCLUSTERS

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Ag_4I_4 nanoclusters, which may have photostimulated luminescence properties as other silver halides have shown, have been synthesized within the sodalite-units of Na-A. The Ag-A crystal, prepared by an ion-exchange of Na-A with 0.05 M $AgNO_3$ aqueous solution, was thoroughly washed with DI water and then reacted with 0.05 M NaI aqueous solution by flow method at 294 K. The crystal structure of resulting Na-A·(AgI) has been determined by single-crystal diffraction technique in the space group of $Pm\bar{3}m$ at 294 K (a = 12.259(2) Å). It was refined with all measured reflections to the final error indice $R_1 = 0.0915$ for 93 reflections with $F_o > 4\sigma(F_o)$.
Twelve Na^+ ions per unit cell are found and distributed at three crystallographically distinct positions: 7.5 Na^+ ions at Na(1) and 0.5 Na^+ ions at Na(4) opposite 6-rings on the 3-fold axes equipoints in the large cavity (Na(1)-O(3) = 2.36(4) Å and Na(4)-O(3) = 2.92(79) Å), 2 Na^+ ions at Na(2) near the 8-rings (Na(2)-O(1) = 2.71(36) Å) and 2 Na^+ ions opposite 4-rings (Na(3)-O(1) = 3.16(17) Å). AgI moieties are found both in the large-cavity (1.0 per α–cage) and the sodalite-unit (0.5 per β-cage). The approach distances between Ag^+ and I^- are 3.09(17) Å and 3.47(16) Å, respectively, for those in the large cavity and sodalite unit. Considering the limited space of the sodalite-unit, the arrangement of four sets of AgI pairs in the same sodalite-unit (Ag_4I_4 with T_d symmetry in 12.5% of all sodalite-units), having arrangement of two interpenetrating cubes, becomes most plausible. The resulting slightly distorted cubic structure of four AgI ionic pairs is well stabilized by the interactions of four tetrahedrally arranged Ag^+ ions from the four tetrahedron corners of the cube to the oxygens atoms of 6-ring windows (Ag(2)-O(3) = 2.28(5) Å) and of those four I^- anions from the other four tetrahedron corners of the cube to Na^+ ions at Na(1) on the other sides of 6-rings in the large cavity (I(2)-Na(1) = 2.94(28) Å). Each of these Ag^+ and I^- ions bonds to three I^- and Ag^+ ions, respectively (Ag(2)-I(2) = 3.47(16) Å).

INTRODUCTION

Zeolites, with the periodic nature of unique-sized pores and channels, are known to be ideal hosts for the preparation of nano-sized clusters and are very useful for the stabilization of unusual chemical species and finely dispersed bulk materials.[1,2] Various nano-compounds for the utilization of their electric and chemical properties have been synthesized in these zeolites with uniformity in size and regularity in orientation.[3-9]

Silver and silver halide clusters have been studied extensively due to their photosensitive properties.[10-12] They are very sensitive to light stimulation and may find applications as photocatalysts for solar energy conversion and as a medium for optical information or image storage.[13-18] Silver microclusters on silver halides grains may work as latent images and reduction sensitization centers which play a dominant role in the photographic processes.

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Rosseti *et al.* explained the particle size dependence of the excited state electronic properties of silver bromide and silver iodide as a consequence of electron and hole localization in the small crystallites.[19] Many previous studies to prepare such silver halide nanoparticles with a high degree of mono-dispersity have been reported.[20,21] Water-in-oil micro-emulsion technology has also been applied.[20] Formation of silver halide nanoparticles in supercritical fluids offers significant advantages over conventional liquid-phase systems, including rapid separation of solvent and the possibility of depositing the particles *in situ* in porous materials by the use of unique properties of the supercritical fluid phase.[21]

The incorporation of silver halides into zeolites, which can, due to their regular pore and channel systems, stabilize unusual silver halide species, should yield regular arrays of nano-sized clusters with quantum-size effects.[15,22-24] Additionally, silver halides in various modified zeolites may have applications in some fields such as semiconductor, photoconductor, information storage and photoelectric conversion materials. For preparation of nano-compounds into zeolites, ion-exchange or impregnation methods are commonly used with the porous materials in the solutions of corresponding materials.[25,26]

Recently, both Ag and AgI nanoparticles encapsulated within zeolite-Y were shown to exhibit strong photostimulated luminescence (PSL).[27] Due to the effect of quantum-size confinement, the luminescence efficiency in nanophase materials may be enhanced relative to bulk materials. In addition, the luminescence wavelength is tunable with sizes of the clusters. Light scattering is significantly reduced in nanoparticles compared with micron-sized particles, since the intensity of light scattering is inversely proportional to the size of the particles. Therefore, nanophase materials may represent an efficient PSL phosphor for X-ray storage. A number of studies on PSL phenomenon with nanoparticles has recently been reported. [28]

Takushi Hirono *et al*. proposed an optical recording medium, consisting of a silver halide dispersed in a large-pore zeolite.[29] They claimed that the material would darken when exposed to light and may fade back to its original color on heating. G. A. Ozin *et al*. have extensively studied the synthesis, structure, and spectroscopic properties of dispersed semiconductor-component (silver halides) clusters in various halosodalites, claiming that they show thermochromic, photochromic, and barochromic properties.[10,15,30]

T. Kodaira *et al*. discussed the electronic states and structures of the AgI clusters, prepared by direct absorption of bulk AgI into the Na-A zeolite, on the basis of their optical absorption spectra.[12] X-ray powder diffraction patterns showed significant and systematic changes, depending on the loading density of AgI in the range from 0.2 to 4.0 molecules per α–cage. They suggested that $(AgI)_n$ clusters, where n = 2 ~ 4, form in α–cages with increasing loading density, and that the cluster occupies a position of tetrahedral symmetry in the α–cage.

Nanoscale silver iodide guests were introduced into zeolites NaZSM-5 and NaY by Qing-Zhou Zhai *et. al*. who studied their products using X-ray diffraction (XRD), diffusion thermal anlysis (DTA), X-ray photoelectric spectroscopy (XPS), and adsorption techniques (to check the locations of silver iodide in the host-guest materials).[31] They prepared those host-guest nanocomposite materials, such as NaY-AgI and NaZSM-5-AgI, by a heat diffusion method and investigated their quantum confinement effect.

In this work, fully Ag^+-exchanged zeolite A (Ag_{12}-A) has been treated with aqueous NaI solution in an attempt to synthesize nanoclusters of AgI in the zeolite. The crystal structure of the resulting product was determined to verify that nanoclusters had formed and to learn their positions, their size and geometry, and to observe their interactions with the zeolite framework.

EXPERIMENTAL SECTION

Colorless single crystals of zeolite 4A, $Na_{12}Si_{12}Al_{12}O_{48}\cdot 27H_2O$ (Na_{12}-A·$27H_2O$ or Na-A·$27H_2O$), were synthesized by Kokotailo and Charnell,[32] having sizes of 80 ~ 100 μm on an edge. A crystal of hydrated Ag_{12}-A (or Ag-A) was prepared by the dynamic (flow) ion-exchange of Na-A with *ca.* 10 mL of aqueous 0.05 M $AgNO_3$ (Aldrich 99.998 %).[33,34] The resulting Ag-A crystal was thoroughly washed with DI water and reacted with *ca*. 10 mL of aqueous 0.05 M NaI (Aldrich 99.99 %) by the flow method at 294 K for 3 days. The crystal was then isolated in its capillary by sealing both ends with a small torch. No attempts were made either to dry or to remove solvent, neither by evacuation nor heating. The color of the crystal after ion-exchange with Ag^+ was pale reddish brown. Upon reaction with NaI, it became pale yellow.

The structure of resulting crystal was determined by single-crystal X-ray diffraction techniques at 294(1) K and 1 atm. The cubic space group $Pm\bar{3}m$ (no systematic absences) was used in this work for reasons discussed previously.[35,36] The cell constant, a is 12.259(2) Å at 294(1) K. Background intensity was counted at each end of a scan range for a time equal to half the scan time. The intensities of three reflections in diverse regions were recorded every 3 h to monitor crystal and instrumental stability. Only small random fluctuations of these check reflections were observed during the course of data collection. Absorption corrections (μ = 5.13 mm^{-1})[37] was judged to be negligible for this crystal since semi-empirical ψ-scans showed only negligible fluctuations for several reflections. A summary of the experimental and crystallographic data is presented in Table 1.

Table 1
Summary of experimental and crystallographic data

Crystal cross-section (mm)	0.08
Ion exchange for Ag^+ (days, mL)	3, 10.0
Reaction of Ag-A with NaI (days, mL)	3, 10.0
Temperature for data collection (K)	294
Radiation (Mo Kα : □) λ_1	0.70930
λ_2	0.71359
Space group, No.	$Pm\bar{3}m$, 221
Scan technique	θ-2θ
Scan width (deg)	0.80 + 0.35 tanθ
Unit cell constant, a_0 (□)	12.259(2)
2θ range for a_0 (deg)	20-30
No. of reflection for a_0	15
2θ range in data collection (deg)	2-70
No. of unique reflections, m	874
No. of reflections (F_o>4σ(F_o))	93
No. of variables, s	42
Data/parameter ratio, m/s	20.8
Weighting parameters : a/b	0.0037/254.5320
Final error indices	0.0915
R_1[a] (F_o>4σ(F_o))	0.5301
wR_2[b] (intensities)	0.940
Goodness of fit[c]	

R_1 = (|F_o - |F_c||/(F_o, R_1 is calculated using only the 93 reflections for which F_o > 4((F_o). [b]R_2 = ((w|F_o - F_c|)^2/(wF_o^2)$^{1/2}$, R_2 is calculated using all 874 unique reflections measured. [c]Goodness-of-fit = (w(F_o - |F_c|)^2/(m-s)|$^{1/2}$.

STRUCTURE DETERMINATION

Full-matrix least-squares refinement (SHELXL97)[38] was initiated with the atomic parameters of the framework atoms [(Si,Al), O(1), O(2), and O(3)] in Na_{12}-A.[39] The refinement using isotropic thermal parameters for [(Si,Al), O(1), O(2), and O(3)] and the Na(1) position at a peak (0.2189, 0.2189, 0.2189), opposite a six-ring in the large cavity from the initial difference Fourier function, led to convergence with R_1 = 0.2131 and wR_2 = 0.7273 and an occupancy of 6.8(14) for Na(1). A subsequent refinement including Na(2), a peak found near an eight-ring at (0.0493, 0.4763, 0.5), led to R_1 = 0.1527 and wR_2 = 0.6863, with occupancies of 6.4(13) and 2.5(11) for Na(1) and Na(2), respectively. Inclusion of another peak at Na(3),

opposite a four-ring at (0.2543, 0.2543, 0.5) in the large cavity, led to R_1 = 0.1341 and wR_2 = 0.5877, with occupancies of 6.4(12), 3.0(13), and 3.3(14) for isotropically refined Na(1), Na(2), and Na(3), respectively. Refinement including the O(4) position at a peak (0.3181, 0.3181, 0.3181), opposite a six-ring in the large cavity from the difference Fourier function, led to R_1 = 0.1200 and wR_2 = 0.4717, with resulting occupancies of 7.5(11), 4.1(13), 4.7(16), and 4.0(13) for Na(1), Na(2), Na(3), and O(4), respectively. The next difference Fourier function (based on this model) revealed a peak at (0.2491, 0.5, 0.5) in the large cavity. Refinement including this peak as O(5) converged to R_1 = 0.1171 and wR_2 = 0.5974, with occupancies of 7.5(12), 4.3(16), 4.3(20), 3.8(13), and 1.2(11) for Na(1), Na(2), Na(3), O(4), and O(5), respectively. Refinement using fixed occupancies of 8, 2, 2, 4, and 2 for Na(1), Na(2), Na(3), O(4), and O(5), respectively, led to R_1 = 0.1195 and wR_2 = 0.5974. The addition of two other peaks at Ag(1) opposite a four-ring at (0.2032, 0.3399, 0.5) and I(1) at (0.2800, 0.2800, 0.4166) in the large cavity led to R_1 = 0.1079 and wR_2 = 0.6124, with occupancies 0.7(3) and 0.5(2) for Ag(1) and I(1), respectively. The cycles of refinement with occupancies fixed at 8, 2, 2, 4, 2, 1, and 1 for Na(1), Na(2), Na(3), O(4), O(5), Ag(1), and I(1), respectively, converged to R_1 = 0.1216 and wR_2 = 0.6117. A subsequent refinement including Ag(2), a peak found opposite a six-ring inside the sodalite unit at (0.1852, 0.1852, 0.1852), led to R_1 = 0.1047 and wR_2 = 0.5865, with occupancy of 0.4(3) for isotropically refined Ag(2). The addition of another peak at I(2) (0.0952, 0.0952, 0.0952, inside the sodalite unit) reduced the error indices to R_1 = 0.0928 and wR_2 = 0.5278, with occupancies 0.3(2) and 0.6(2) for Ag(2) and I(2), respectively. The cycles of refinement with occupancies fixed at 0.5 and 0.5 for Ag(2) and I(2), respectively, converged to R_1 = 0.0946 and wR_2 = 0.5281.

Considering the observed content of each sodalite unit, 0.5 Ag^+ and 0.5 I^- ions on the four threefold axes opposite 6-rings (out of eight 6-rings per unit cell), there must be at least three different kinds of 6-rings per unit cell: 0.5 6-rings with Ag^+ ions at Ag(2) and 0.5 with I^- anions at I(2), and the rest three with nothing in the sodalite unit. This naturally suggests that there should also be at least three kinds of corresponding O(3) oxygens on the 6-rings as well as the same numbers of Na^+ ions at the other sides, namely the large–cavity sides (although eight Na^+ ions at Na(1) are refined as one kind at this moment). This suggestion agrees very well with the observed and unusually elongated thermal parameters for Na^+ ions at Na(1) and another peak was found at (0.2663, 0.2663, 0.2663) near the Na(1) (0.2189, 0.2189, 0.2189) on the same threefold axis in a subsequent difference Fourier function. A refinement with this peak included as Na(4) with varied occupancy for the Na^+ ions at Na(1) converged to R_1 = 0.0923 and wR_2 = 0.5047, resulting in the occupancies of 7.5(12) and 0.5(fixed) for Na(1) and Na(4), respectively. When this model was refined with anisotropic thermal parameters for all of the framework atoms [(Si,Al), O(1), O(2), and O(3)] and with fixed occupancies of 7.5 and 0.5 for Na(1) and Na(4), respectively, converged to R_1 = 0.0923 and wR_2 = 0.5047.

The final cycles of refinement with occupancies fixed as shown in Table 2 converged to R_1 = 0.0915 and R_2 = 0.5301 with anisotropic thermal parameters for the framework atoms [(Si,Al), O(1), O(2), and O(3)]. Extensive, but unsuccessful efforts were made to locate solvent (maybe H_2O) molecules. A final difference Fourier function was featureless; the notation Na-A(AgI) will be used for this crystal. All shifts in the final cycles of refinement were less than 0.1 % for the corresponding estimated standard deviations. The final structural parameters are given in Table 2. Selected interatomic distances and angles are given in Table 3.

Fixed weights were used initially; the final weights were assigned using the formula $w = 1/[\sigma^2(F_o^2) + (aP)^2 + bP]$ where $P = [\mathrm{Max}(F_o^2,0) + 2F_c^2]/3$, with a = 0.004 and b = 254.53 as refined parameters (see Table 1). Atomic scattering factors for Ag^+, I^-, Na^+, O^-, and $(Si,Al)^{1.75+}$ were used.[40,41] The function describing $(Si,Al)^{1.75+}$ is the mean of the Si^{4+}, Si^0, Al^{3+}, and Al^0 functions. All scattering factors were modified to account for anomalous dispersion.[42,43]

Table 2

Positional, thermal, and occupancy parameters[a]

Atoms	Wyckoff position	x	y	z	U_{11} or U_{iso} [b]	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}	Occupancy[c] fixed	Occupancy[c] varied
(Si,Al)	24(k)	0	1840(13)	3724(13)	1(101)	1(102)	52(96)	75(95)	0	0	24[d]	
O(1)	12(h)	0	2248(46)	5000[e]	180(294)	332(365)	362(353)	0	0	0	12	
O(2)	12(i)	0	2912(29)	2912(29)	436(397)	183(241)	183(241)	-121(255)	0	0	12	
O(3)	24(m)	1111(24)	1111(24)	3368(30)	394(170)	394(170)	401(300)	69(155)	69(155)	-47(289)	24	
Na(1)	8(g)	2186(41)	2186(41)	2186(41)	748(174)						7.5	7.5(12)
Na(2)	24(I)	366(272)	4428(264)	5000[e]	834[f]						2	4.3(16)
Na(3)	12(j)	2560(127)	2560(127)	5000[e]	390[f]						2	4.3(20)
O(4)	8(g)	3232(90)	3232(90)	3232(90)	341(393)						4	3.8(13)
O(5)	6(f)	2448(258)	5000[e]	5000[e]	509(909)						2	1.2(11)
Ag(1)	24(l)	1921(133)	3400(155)	5000[e]	876(632)						1	0.7(3)
I(1)	24(m)	2702(185)	2702(185)	4093(224)	2179(1336)						1	0.5(2)
Ag(2)	8(g)	1714(81)	1714(81)	1714(81)	299(386)						0.5	0.3(2)
I(2)	8(g)	802(128)	802(128)	802(128)	1660(1214)						0.5	0.6(6)
Na(4)	8(g)	2732(593)	2732(593)	2732(593)	748[g]						0.5	

Table 3

Selected interatomic distances (Å) and angles (deg)[a]

Distances		Angles	
(Si,Al)-O(1)	1.64(2)	O(1)-(Si,Al)-O(2)	109.4(27)
(Si,Al)-O(2)	1.65(2)	O(1)-(Si,Al)-O(3)	114.1(17)
(Si,Al)-O(3)	1.69(2)	O(2)-(Si,Al)-O(3)	105.4(19)
		O(3)-(Si,Al)-O(3)	107.7(28)
Na(1)-O(3)			
Na(1)-O(2)	2.36(4)	(Si,Al)-O(1)-(Si,Al)	144.6(38)
Na(2)-O(1)	2.96(3)	(Si,Al)-O(2)-(Si,Al)	164.3(37)
Na(3)-O(1)	2.71(36)	(Si,Al)-O(3)-(Si,Al)	142.1(30)
Na(4)-O(3)	3.16(17)		
Ag(1)-O(1)	2.92(79)	O(3)-Na(1)-O(3)	120.5(32)
Ag(2)-O(3)		Na(1)-O(4)-Na(3)	97.0(61)
	2.75(17)	O(4)-Na(3)-O(4)	123.5(100)
Na(1)-O(4)	2.28(5)		
Na(3)-O(4)		Ag(2)-I(2)-Ag(2)	118.0(22)
Na(2)-O(5)	2.22(18)	I(2)-Ag(2)-I(2)	47.3(76)
	2.46(11)		
Ag(1)-I(1)	2.65(42)	Na(1)-I(1)-Ag(1)	84.0(62)
Na(1)-I(1)		O(1)-Ag(1)-I(1)	114.0(64)
Na(1)-I(2)	3.09(17)		
Ag(2)-I(2)	2.50(29)		
	2.94(28)		
	3.47(16)		

[a] The numbers in parentheses are the estimated standard deviations in the units of the least significant digit given for the corresponding parameter.

RESULTS AND DISCUSSION

Crystal Structure of Na-A·(AgI)

AgI clusters are found, probably in the form of Ag_4I_4 nanoclusters in the sodalite units and AgI molecules in the large cavities (*vide infra*). The structural parameters of the framework atoms and Na^+ ions are somewhat different from those in hydrated and dehydrated Na_{12}-A. This suggests that this crystal contains some kind of occluded materials in its cavities, together with water molecules.

Framework and Na^+ Ions

In dehydrated Na_{12}-A,[39] eight equivalent Na^+ ions at Na(1) per unit cell lie on 3-fold axes opposite 6-rings in the large cavity, another three at Na(2) lie near the centers of the 8-rings, and one additional Na^+ ion at Na(3) lie along the remaining 2-fold axes opposite 4-rings. In Na-A·(AgI), however, the twelve Na^+ ions per unit cell are distributed over four crystallographically distinct positions as in the hydrated Na_{12}-A: 7.5 Na^+ at Na(1) and 0.5 Na^+ at Na(4) on the 3-fold axes opposite 6-ring in the large cavity, two Na^+ at Na(2) near 8-ring planes, the remaining two Na^+ at Na(3) on 2-fold axes opposite 4-rings. Therefore, each of the eight 6-rings per unit cell contains a Na^+ ion (7.5 out of eight 6-rings at Na(1) and the other 0.5 at Na(4)); each Na^+ ion lies on a 3-fold axis and is 0.69 or 1.85 Å away from the (111) planes of averagely positioned O(3)s, respectively (see Table 4). Considering the sum of ionic radii of Na^+ and O^{2-}, 2.29 Å = 0.97 + 1.32 Å, the Na(1)-O(3) approach distance, 2.36(4) Å, is close to the calculated bonding distance, and Na(4)-O(3), 2.92(79) Å, is somewhat longer (see Table 3). This longer distance can be explained by the intercationic repulsion of Na^+ ion at Na(4) with Ag^+ ion at Ag(2) opposite a six-ring inside the sodalite unit and by the same occupancies of Na(4) and Ag(2) (0.5).

Table 4

Deviations of atoms (Å) from the (111) plane at O(3)[a]

Atom	Charge	Distance
Ag(2)	+1	-0.32
Na(1)	+1	0.69
Na(4)	+1	1.85
I(2)	-1	-2.25
O(4)	0	2.91

[a]A negative deviation indicates that the atom lies on the same side of the plane as the origin, *i.e.*, inside the sodalite unit.

Two Na^+ ions per unit cell at Na(2) occupy the two 8-rings and lie *ca.* 0.8 Å away from the 8-ring plane. Each Na(2) ion is 2.71(36) from one O(1) oxygen of an 8-ring. The Na(2)-O(1) approach distances, 2.71(36) Å, are similar to the corresponding ones in dehydrated Na_{12}-A,[39] 2.64(3) Å and the Na(2)-O(2) approach distances, *ca.* 3.2 Å, are much longer than the corresponding distance, 2.40(6) Å. This indicates that the O(1) and O(2) oxygens contribute unequally to the coordination spheres of the 8-ring Na^+ ions: *i.e*, Na(2) coordinates primarily to the O(1) oxygens. Similar unequal coordination of 8-ring oxygens to Na^+ ions, with a lesser degree of inequality, was seen in the crystal structures of hydrated and dehydrated Na_{12}-A.[39]

The Na(3) position, occupied by two Na^+ ions per unit cell, opposite 4-rings in the large cavity, is also found in dehydrated Na_{12}-A. Each of these Na^+ ions is 3.16(17) Å and 3.21(17) Å from O(1) and O(3), respectively. Perhaps, they are only virtually longer than other Na-O distances probably because this low occupancy position at Na(3) has averaging effect with oxygen atoms of other 4-rings, having no Na^+ ions on them, and because the Na^+ ion at Na(3) makes reasonable approaches to two oxygen atoms of H_2O molecules opposite a six-ring in the large cavity at O(4) (see Figs. 1 and 2). The bond distance of Na(3)-O(4) is 2.46(11) Å, and that is a reasonable one for Na^+ ion to O atom, considering the esd and the symmetric structures.

Six O atoms, which are probable those of H_2O molecules were found per unit cell. Four O atoms per unit cell at O(4) lie on the 3-fold axes opposite 6-ring in the large cavity with approach distances of 2.22(18) Å and 2.46(11) Å to Na^+ ions at Na(1) and Na(3), respectively, and two O atoms per unit cell at O(5) have a reasonable approach distances of 2.65(42) Å to Na^+ ions at Na(2) near eight-rings in large-cavity (see Figs. 1 and 2).

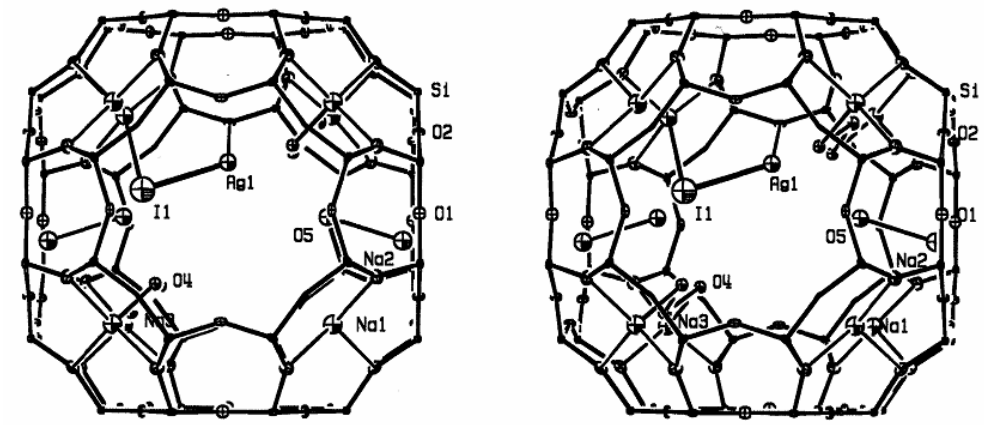

Fig. 1 – A stereoview of the large cavity of Na-A·(AgI) with one AgI ionic pair and two water molecules near Na^+ ions. The zeolite A framework is drawn with light bonds between oxygens and tetrahedrally coordinated (Si,Al) atoms. Ellipsoids of 20% probability are shown.

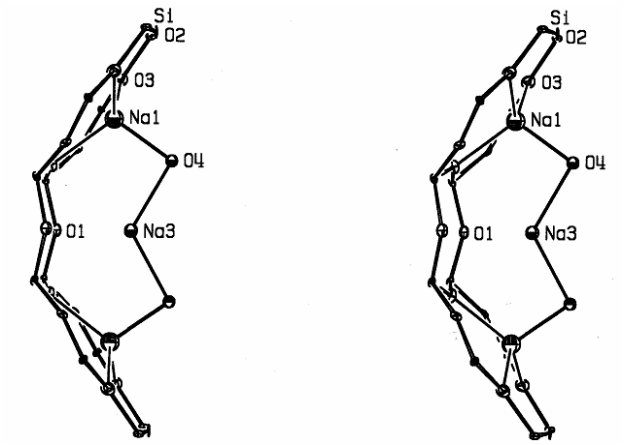

Fig. 2 – A stereoview of water molecules coordinated to Na^+ ions in the large cavity of Na-A·(AgI). See the caption to Fig. 1 for other details.

AgI Nanocluster in the Sodalite Unit

A half of Ag^+ ions and a half of I^- anions per unit cell are found at two (nonequivalent) threefold-axis positions in the same sodalite unit. 0.5 Ag^+ ions at Ag(2) lie opposite 6-rings in the sodalite unit and the 0.5 I^- ions at I(2) occupy similar positions, recessed more deeply into the sodalite unit. It is impossible for both a Ag^+ and an I^- ion to approach the same 6-ring because of an inevitable short approach distance (1.94(31) Å).

The Ag^+ ions at Ag(2) are each 2.28(5) Å from three 6-ring oxygens at O(3) (see Table 3) and extend 0.32 Å into the sodalite unit from the (111) planes at O(3) (see Table 4). Considering the ionic radii of the framework oxygens to be 1.32 Å, the ionic radii of these Ag^+ ions must be 2.28 – 1.32 = 0.96 Å. This is very close to those commonly found in variously Ag^+-exchanged zeolites A, *ca.* 0.99 Å.[33,34]. Each Ag^+ ion at Ag(2) can bond to up to three I^- ions at I(2) opposite the three adjacent 6-rings with approach distances of 3.47(16) Å. It is somewhat longer than the calculated distance, because the sum of the Ag^+ and I^- radii is known to be in the range from 2.78 to 3.33 Å (γ_{Ag+} = 1.13 Å, γ_{I-} = from 1.65 to 2.20 Å).[44,45]

Considering the possible arrangements of I^- ions at I(2) and Ag^+ ions at Ag(2) within the limited space of the sodalite unit, an interpenetrating tetrahedral (puckered cubic) arrangement of four Ag^+ and four I^- ions in 12.5% of all sodalite units seems most plausible (see Figs. 3 and 4). It gives the greatest number of bonds per atom and justifies the symmetry of the Ag(2) and I(2) positions. In this arrangement, each Ag^+ ion would bond to three I^- ions (in addition to three framework oxygens), and each I^- ions would bond to three adjacent Ag^+ ions (Ag(2)-I(2) = 3.47(16) Å, Ag(2)-I(2)-Ag(2) = 118.0(22)°, and I(2)-Ag(2)-I(2) = 47.3(76)°). The resulting neutral Ag_4I_4 nanocluster, with T_d symmetry, would be well stabilized by the interactions of its four tetrahedrally arranged Ag^+ ions with 12 6-ring oxygens (Ag(2)-O(3) = 2.28(5) Å) and of those four I^- anions from the other four tetrahedron corners of the cube to Na^+ ions at Na(1) on the other sides of 6-rings in the large cavity (I(2)-Na(1) = 2.94(28) Å). The stereoview of such a sodalite unit with a Ag_4I_4 nanocluster at its center is shown in Fig. 3.

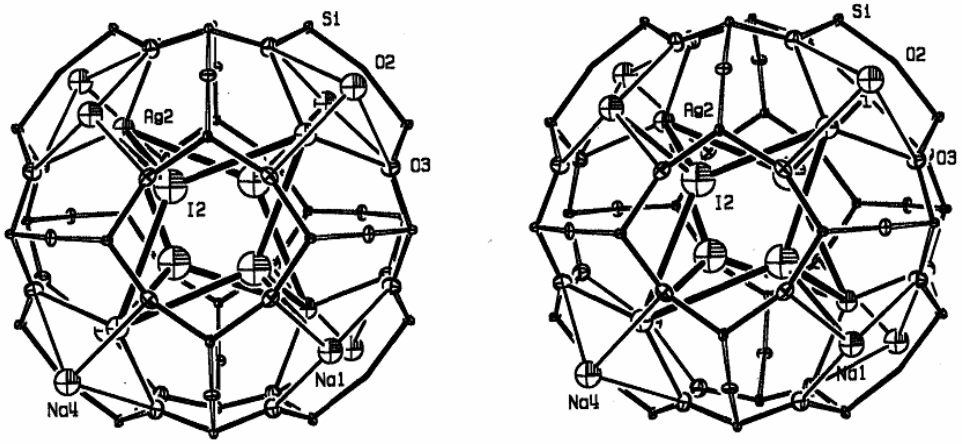

Fig. 3 – A stereoview of a sodalite unit of Na-A·(AgI) showing four AgI units as Ag_4I_4 nanocluster in two interpenetrating cubes. See the caption to Fig. 1 for other details.

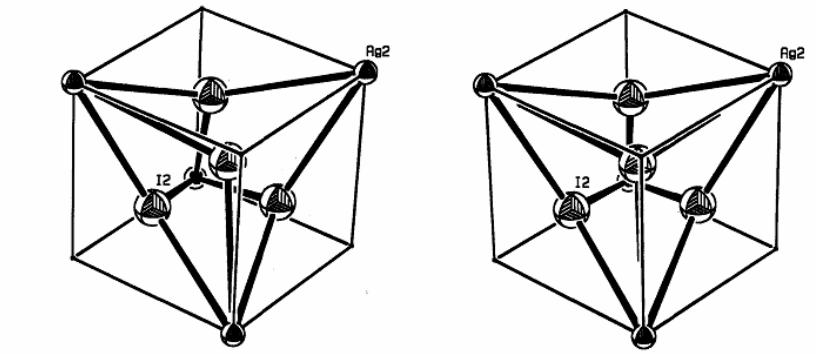

Fig. 4 – Ag_4I_4 nanocluster found in sodalite units of Na-A·(AgI). See the caption to Fig. 1 for other details.

The Ag_4I_4 cluster has a diameter of *ca.* 8.0 Å. It is a neutral ionic nanocluster which, like others reported earlier,[10-13] may have interesting optical properties. This Ag_4I_4 nanocluster is different from silver halide clusters of Ag_nX, X = Cl, Br, and I,[10] that were reported by Ozin *et al.*; each has a halide anion at its center. Syntheses and properties of $(AgI)_n$ nanoclusters have been extensively studied by various spectroscopic methods.[12,13]

Compared with previous studies on AgI in fully K^+-exchanged zeolite A (K-A·(AgI)), the occupancy (0.125) of Ag_4I_4 nanocluster in Na-A·(AgI) is far less than that (0.5) found in K-A·(AgI).[46] That is probably because those 7.5 Na^+ ions on the 3-fold axes opposite 6-ring in Na-A make strong bonds with O(3) and extend only 0.69 Å into the large cavity from the (111) planes at O(3) (see Table 4), leaving smaller space for the cluster in the sodalite unit, and because the inclusion and stabilization of the Ag^+ ions (precursor of Ag_4I_4 clusters) in the sodalite unit were definitely disturbed by the intercationic repulsive force with those Na^+ ions on the 3-fold axes. Ag^+ ions of Ag_4I_4 clusters in K-A·(AgI), however, were more stable in the sodalite unit, because those K^+ ions on the 3-fold axes opposite 6-ring extend 1.48 Å into the large cavity from the (111) planes at O(3), making larger space for the clusters in the sodalite unit, and because each of those K^+ ions can lie on same 3-fold axis with Ag^+ ion of Ag_4I_4 at the other side of 6-ring inside sodalite unit.

AgI Molecule in the Large Cavity

A AgI molecule is found near opposite a 4-ring in the large cavity and well stabilized by interaction with framework oxygens and Na^+ ions (see Table 2 and Figs. 1 and 5). An I^- ion at I(1) is bonded to a Ag^+ ions at Ag(1) opposite a 4-ring in the large cavity (I(1)-Ag(1) = 3.09(17) Å) and is also bonded to a Na^+ ion at Na(1) on a 3-fold axis opposite 6-ring in the large cavity (I(1)-Na(1) = 2.50(29) Å, Na(1)-I(1)-Ag(1) = 84.0(62)°). Because the Ag^+ ion at Ag(1) makes a strong ionic bond with the I^- anion, this Ag^+ ion is somewhat further away from framework oxygen (Ag(1)-O(1) = 2.75(17) Å, O(1)-Ag(1)-I(1) = 114.0(64)°).

SUMMARY

Ag_4I_4 nanoclusters with T_d symmetry have been synthesized and stabilized in about 12.5% of the sodalite cavities of Na-A by treating Ag-A with NaI in H_2O. One AgI molecule per unit cell is retained deep in large cavity and stabilized by the interactions with Na^+ ions and framework oxygens.

ACKNOWLEDGMENT. We gratefully acknowledge the support of the Central Laboratory of Kyungpook National University for the diffractometer and computing facilities. This work was supported by Korea Research Foundation Grant (KRF-2002-037-C00017).

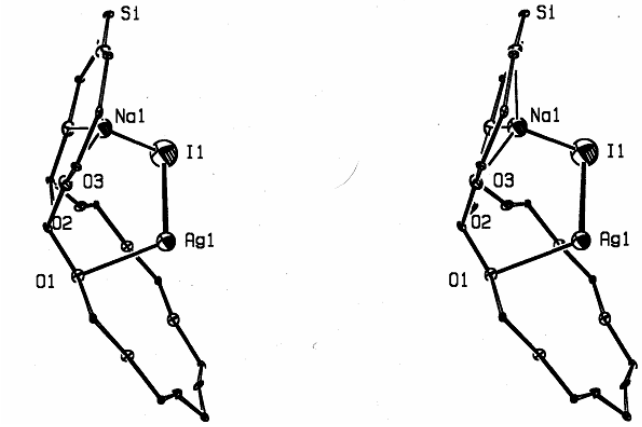

Fig. 5 – A stereo- and close-view of AgI ionic pair in the large cavity of Na-A·(AgI). See the caption to Fig. 1 for other details.

Supplementary material available: observed and calculated structure factors for Na-A·(AgI). To be found at the end of the article.

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Supplementary Material

Table
for observed and calculated structure factors squared
for

Synthesis and Crystal Structure of Na-A with Occluded AgI and Ag_4I_4 Nanoclusters

by

Nam Ho Heo,* Jeong Seok Noh, and Seok Han Kim

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
0	0	1	15146.28	15353.31	242.36	o	0	3	5	1752.07	1726.48	353.52	o
0	1	1	6142.53	6430.53	254.96	o	1	3	5	822.55	1674.44	318.55	o
1	1	1	9504.56	6315.79	302.61	o	2	3	5	234.16	162.69	325.38	o
0	0	2	1293.69	-91.65	241.63	o	3	3	5	7.56	-81.22	349.25	o
0	1	2	2714.94	1302.37	280.9	o	0	4	5	2649.39	2890.64	371.65	o
1	1	2	135.59	-179.08	268.62	o	1	4	5	2509.91	2244.09	363.01	o
0	2	2	2148.03	2711.37	318	o	2	4	5	1079.97	1422.54	367.9	o
1	2	2	6907.37	5052.61	371.65	o	3	4	5	255.15	292.82	349.76	o
2	2	2	1152.4	707.86	337.47	o	4	4	5	6297.69	6668	461.89	o
0	0	3	5167.67	6668.27	372.87	o	0	5	5	29606.15	31237.82	763.28	o
0	1	3	46.05	162.53	316.94	o	1	5	5	478.9	700.41	350.2	o
1	1	3	12864.23	12034.51	447.1	o	2	5	5	323.43	301.05	358.01	o
0	2	3	1497.69	3501.34	355.09	o	3	5	5	454.01	-24.42	390.8	o
1	2	3	6425.47	5922.19	398.86	o	4	5	5	1516.01	1972.49	433.78	o
2	2	3	8519.67	8893.19	412.62	o	5	5	5	6823.45	9189.93	545.02	o
0	3	3	794.84	1571.24	319.16	o	0	0	6	30565.6	29687.74	706.16	o
1	3	3	20.45	-259.95	300.56	o	0	1	6	132.04	617.8	341.42	o
2	3	3	16082.67	21162.11	523.77	o	1	1	6	272.33	0	333.6	o
3	3	3	2681.62	2404.54	339.95	o	0	2	6	133.23	65.03	325.15	o
0	0	4	1185.38	715.07	320.55	o	1	2	6	44.67	-316.84	333.09	o
0	1	4	9901.51	10950.38	426.29	o	2	2	6	471.43	635.35	350.26	o
1	1	4	3064.25	2783.04	317.58	o	0	3	6	563.94	187.45	350.44	o
0	2	4	4206.44	3792.47	362.39	o	1	3	6	4.68	-138.07	333	o
1	2	4	1257.57	673.4	295.64	o	2	3	6	988.26	441.13	367.61	o
2	2	4	2712.65	2590.71	332.14	o	3	3	6	832.2	383.51	375.35	o
0	3	4	450.35	-220.09	309.76	o	0	4	6	504.85	350.25	374.68	o
1	3	4	185.25	520.56	284.68	o	1	4	6	4034.26	3795.83	431.91	o
2	3	4	492.48	-16.3	317.93	o	2	4	6	412.61	936.13	358.17	o
3	3	4	1812.25	937.62	345.44	o	3	4	6	1747.75	1647.58	393.45	o
0	4	4	13087.12	14448.95	496.1	o	4	4	6	316.62	520.62	431.14	o
1	4	4	158.46	-8.13	325.24	o	0	5	6	6417.53	6883.4	478.48	o
2	4	4	785.3	334.78	342.94	o	1	5	6	161.41	24.39	373.92	o
3	4	4	3124.21	3387.88	405.88	o	2	5	6	288.4	-48.8	422.96	o
4	4	4	1788.65	1928.86	369.36	o	3	5	6	1391.33	940.46	457.96	o
0	0	5	8724.9	8143.68	416.73	o	4	5	6	484.3	797.77	431.45	o
0	1	5	262.65	529.03	301.14	o	5	5	6	4395.03	5225.86	530.04	o
1	1	5	987.38	843.02	294.65	o	0	6	6	3046.67	4615.85	511.04	o
0	2	5	501.47	-105.98	301.65	o	1	6	6	1296.81	1626.44	449.52	o
1	2	5	507.71	163.05	317.95	o	2	6	6	849.69	1622.81	456.67	o
2	2	5	4268.63	3429.95	368.09	o	3	6	6	916.03	1370.26	456.75	o
4	6	6	815.1	-154.88	489.08	o	0	1	8	3.49	1031.48	381.73	o
5	6	6	2612.41	3120.64	525.58	o	1	1	8	550.1	350.22	407.23	o
6	6	6	532.95	-211.62	512.78	o	0	2	8	1869.32	2377.45	450.9	o
0	0	7	194.21	-276.46	357.77	o	1	2	8	915.53	1542.05	424.27	o
0	1	7	5688.91	5032.51	419.38	o	2	2	8	1519.7	1661	441.84	o
1	1	7	1458.07	1834.57	376.74	o	0	3	8	2370.59	2596.04	443.63	o
0	2	7	755.75	1084.83	375.2	o	1	3	8	787.37	1663.16	407.64	o
1	2	7	483.98	651.51	366.47	o	2	3	8	5176.65	5041.8	549.11	o
2	2	7	339.54	-24.41	358.02	o	3	3	8	19769.52	19903	750.89	o
0	3	7	352.4	675.4	390.59	o	0	4	8	22.86	-292.41	454.87	o
1	3	7	4361.64	3980.95	440.48	o	1	4	8	1568.81	1079.88	466.31	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
2	3	7	130.63	-154.42	398.24	o	2	4	8	505.87	1041.99	464.01	o
3	3	7	165.27	300.76	406.43	o	3	4	8	306.36	-105.73	463.57	o
0	4	7	1997.57	2568.19	410.25	o	4	4	8	4696.36	5128.05	613.05	o
1	4	7	37.62	-194.96	398.04	o	0	5	8	385.17	968.15	455.6	o
2	4	7	1962.9	2132.36	442.87	o	1	5	8	1446.2	948.15	514.94	o
3	4	7	2875.48	1564.54	494.06	o	2	5	8	190.45	284.5	471.45	o
4	4	7	37.23	-414.28	463.02	o	3	5	8	1631.08	1177.64	531.58	o
0	5	7	1265.75	1642.48	457.61	o	4	5	8	128.17	154.4	503.82	o
1	5	7	1218.19	1682.89	424.81	o	5	5	8	2057.14	1883.26	507.66	o
2	5	7	0.41	-259.9	438.58	o	0	6	8	1526.12	621.22	523.14	o
3	5	7	83.45	-454.99	454.99	o	1	6	8	1686.61	1897.58	531.65	o
4	5	7	4137.5	3555.88	529.25	o	2	6	8	3.87	-276.14	495.44	o
5	5	7	1954.16	1924.39	524.09	o	3	6	8	566.51	1025.7	520.99	o
0	6	7	31.74	0	454.88	o	4	6	8	23.76	-16.25	487.35	o
1	6	7	352.7	73.21	479.95	o	5	6	8	686.39	928.31	496.73	o
2	6	7	699.77	578.44	464.38	o	6	6	8	77.51	121.86	487.45	o
3	6	7	178.74	0	471.43	o	0	7	8	317.18	-73.19	496.06	o
4	6	7	799.93	48.89	529.69	o	1	7	8	314.05	276.49	520.44	o
5	6	7	528.04	406.96	520.9	o	2	7	8	0.06	519.79	503.55	o
6	6	7	913.7	1043.27	472.73	o	3	7	8	23.34	-113.71	487.35	o
0	7	7	103.53	-16.25	503.77	o	4	7	8	856.38	529.62	488.88	o
1	7	7	156.7	-48.76	520.14	o	5	7	8	3.16	40.61	487.31	o
2	7	7	398.8	642.69	488.12	o	6	7	8	44.25	129.97	528	o
3	7	7	3483.18	3912.54	576.59	o	7	7	8	280.75	-528.42	528.42	o
4	7	7	1308.02	1624.63	506.17	o	0	8	8	340.54	56.93	487.93	o
5	7	7	210.45	113.8	487.7	o	1	8	8	1.83	-487.31	487.31	o
6	7	7	62.62	-284.33	471.17	o	2	8	8	3725.74	5187.42	584.61	o
7	7	7	926.12	627.44	521.51	o	3	8	8	8645.59	11424.61	711.94	o
0	0	8	11638.14	10809.15	567.55	o	4	8	8	0.07	-519.79	519.79	o
5	8	8	203.71	-398.25	512.04	o	0	8	9	76.08	576.8	519.93	o
6	8	8	0.2	276.14	487.31	o	1	8	9	615.94	406.98	512.8	o
7	8	8	460.43	317.23	569.39	o	2	8	9	15.68	-544.19	544.19	o
8	8	8	5953.21	6710.61	670.23	o	3	8	9	582.17	268.56	520.85	o
0	0	9	3082.88	3418.58	502.49	o	4	8	9	235.65	755.93	528.34	o
0	1	9	814.41	130.44	464.68	o	5	8	9	32.15	-576.71	576.71	o
1	1	9	292.66	284.64	439.17	o	6	8	9	207.85	544.53	520.15	o
0	2	9	2424.32	3087.31	492.66	o	7	8	9	106.71	-389.98	593.09	o
1	2	9	858.23	856.09	464.74	o	8	8	9	412.37	-24.4	585.51	o
2	2	9	2697.99	3451.93	517.79	o	0	9	9	12.57	-341.13	552.3	o
0	3	9	1408.33	1904.16	506.69	o	1	9	9	429.63	187.07	528.69	o
1	3	9	1148.45	995.86	481.61	o	2	9	9	581.8	73.24	569.64	o
2	3	9	133.94	-260.05	487.59	o	3	9	9	33.74	97.47	527.97	o
3	3	9	1508.71	1602.03	514.94	o	4	9	9	141.87	901.94	511.91	o
0	4	9	3798.09	3235.15	544.69	o	5	9	9	771.95	2312.31	553.65	o
1	4	9	0.07	-113.7	503.55	o	6	9	9	0.29	259.9	552.28	o
2	4	9	1418.58	522.89	539.23	o	7	9	9	197.87	-56.89	585.12	o
3	4	9	217.13	48.77	512.13	o	8	9	9	318.03	317.05	617.84	o
4	4	9	3361.32	2798.29	559.66	o	9	9	9	1414.32	1231.42	693.18	o
0	5	9	1973.22	2742.81	524	o	0	0	10	13886.35	13719.33	695.41	o
1	5	9	874.31	578.71	513.5	o	0	1	10	14.62	706.64	454.85	o
2	5	9	27.43	552.34	487.36	o	1	1	10	2549.24	2446	525.32	o
3	5	9	1041.39	1353.77	497.47	o	0	2	10	428.48	1082.12	512.58	o
4	5	9	5.99	-113.71	471.07	o	1	2	10	92.45	503.74	528.12	o
5	5	9	108.28	633.75	471.25	o	2	2	10	59.59	227.46	503.67	o
0	6	9	384.82	-187.08	504.31	o	0	3	10	1373.91	1061.71	514.52	o
1	6	9	83.66	-259.98	495.59	o	1	3	10	1483.22	768.02	522.91	o
2	6	9	175.92	56.89	503.89	o	2	3	10	5.52	-89.34	495.44	o
3	6	9	5.05	641.63	462.95	o	3	3	10	4.23	300.51	495.44	o
4	6	9	131.25	-487.54	487.54	o	0	4	10	41.02	276.18	471.14	o
5	6	9	287.85	821.15	463.42	o	1	4	10	263.48	650.42	479.68	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
6	6	9	387.45	-544.9	544.9	o	2	4	10	2.48	885.28	462.95	o
0	7	9	2738.23	3478.9	533.32	o	3	4	10	137.21	576.95	463.18	o
1	7	9	7.74	576.66	479.2	o	4	4	10	8.01	-357.37	495.44	o
2	7	9	990.48	847.76	489.09	o	0	5	10	1025.74	1899.78	529.98	o
3	7	9	1046.41	1182.15	513.62	o	1	5	10	164.13	487.61	479.48	o
4	7	9	291.62	-276.43	512.2	o	2	5	10	19.31	-357.38	487.34	o
5	7	9	766.62	936.49	521.18	o	3	5	10	23.89	-259.92	495.47	o
6	7	9	46.14	398.03	479.26	o	4	5	10	376.71	146.39	496.1	o
7	7	9	671.95	740.71	561.64	o	5	5	10	468.29	479.98	512.52	o
0	6	10	1015.58	1434.77	529.89	o	6	10	10	40.84	-657.94	657.94	o
1	6	10	66.79	487.43	479.3	o	7	10	10	94.31	-706.79	706.79	o
2	6	10	4.26	-333	519.8	o	8	10	10	57.33	1007.26	682.34	o
3	6	10	95.69	146.24	503.72	o	9	10	10	41.32	-170.58	706.67	o
4	6	10	675.04	0	504.74	o	10	10	10	34.67	-251.8	714.78	o
5	6	10	5.9	-300.51	511.68	o	0	0	11	12176.48	12303.48	705.24	o
6	6	10	78.57	406.19	544.3	o	0	1	11	249.94	723.54	479.65	o
0	7	10	233.76	479.58	544.61	o	1	1	11	617.61	748.98	504.75	o
1	7	10	350.83	-65.05	544.83	o	0	2	11	64.29	-503.67	503.67	o
2	7	10	0.01	284.26	519.79	o	1	2	11	18.7	333.02	479.22	o
3	7	10	876.08	1490.78	537.66	o	2	2	11	1112.66	1459.89	448.57	o
4	7	10	0.02	625.38	527.91	o	0	3	11	851.5	1352.52	505.16	o
5	7	10	281.43	625.96	552.8	o	1	3	11	227.02	796.61	503.98	o
6	7	10	159.36	276.28	536.31	o	2	3	11	290.41	284.57	520.35	o
7	7	10	40.61	-601.09	601.09	o	3	3	11	224.93	251.98	495.83	o
0	8	10	268.96	-24.39	536.53	o	0	4	11	92.86	-162.49	511.85	o
1	8	10	119.73	-300.63	560.63	o	1	4	11	1267.44	954.65	522.2	o
2	8	10	76.88	495.56	536.18	o	2	4	11	217.2	211.33	520.2	o
3	8	10	3.7	454.82	536.04	o	3	4	11	17.53	552.31	487.34	o
4	8	10	411.63	618.09	561.16	o	4	4	11	1188.83	1149.93	538.26	o
5	8	10	360.62	430.95	585.44	o	0	5	11	4160.21	4945	626.37	o
6	8	10	448.62	1358.24	569.32	o	1	5	11	110.79	430.62	528.12	o
7	8	10	39.9	-446.75	625.45	o	2	5	11	264.64	-528.41	528.41	o
8	8	10	189.86	1056.43	576.97	o	3	5	11	387.08	447.3	520.49	o
0	9	10	5.98	-121.83	552.29	o	4	5	11	329.09	325.24	536.64	o
1	9	10	175.48	-300.68	576.97	o	5	5	11	3376.71	3465.86	599.55	o
2	9	10	0.23	-154.31	568.52	o	0	6	11	3677.34	4136.98	625.07	o
3	9	10	104.82	259.98	584.96	o	1	6	11	45.14	560.49	528	o
4	9	10	5.42	0	601.02	o	2	6	11	23.61	755.39	495.47	o
5	9	10	341.05	187	560.99	o	3	6	11	233.29	634	544.59	o
6	9	10	27.84	-170.57	552.33	o	4	6	11	326.59	1081.37	512.23	o
7	9	10	42.92	617.33	617.33	o	5	6	11	1742.65	1625.38	612.58	o
8	9	10	8.97	341.12	625.39	o	6	6	11	250.25	-130.05	609.62	o
9	9	10	28.06	292.41	674.16	o	0	7	11	0.08	162.44	536.04	o
0	10	10	407.39	609.91	585.51	o	1	7	11	273.18	-170.71	552.78	o
1	10	10	417.74	-292.77	618.06	o	2	7	11	288.33	-268.27	585.33	o
2	10	10	65.12	617.38	601.13	o	3	7	11	15.61	-316.77	560.43	o
3	10	10	597.19	772.99	618.39	o	4	7	11	600.01	406.87	602.17	o
4	10	10	53.07	787.94	576.74	o	5	7	11	21.67	48.73	560.44	o
5	10	10	3.85	982.74	609.14	o	6	7	11	115.08	-349.36	601.22	o
7	7	11	137.77	536.26	568.76	o	9	11	11	12.54	-787.84	787.84	o
0	8	11	476.23	-618.2	618.2	o	10	11	11	312.24	276.36	772.19	o
1	8	11	118.48	503.74	536.24	o	11	11	11	1674.05	2071.71	831.95	o
2	8	11	241.84	0	560.84	o	0	0	12	94.75	-536.22	536.22	o
3	8	11	517.81	602	561.32	o	0	1	12	787.01	749.31	504.97	o
4	8	11	0.79	211.17	536.04	o	1	1	12	245.38	24.39	520.25	o
5	8	11	610.37	1188	585.86	o	0	2	12	169.88	-536.36	536.36	o
6	8	11	365.73	1829.41	561.02	o	1	2	12	449.11	496.22	488.08	o
7	8	11	17.42	-666.02	666.02	o	2	2	12	462.38	1431.75	479.96	o
8	8	11	379.4	-32.52	642.32	o	0	3	12	100.14	-178.74	503.73	o
0	9	11	166.54	1235.15	560.69	o	1	3	12	184.89	1129.65	503.87	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
1	9	11	346.42	780.53	585.4	o	2	3	12	224.76	804.68	487.69	o
2	9	11	106.47	-625.58	625.58	o	3	3	12	344.73	227.68	528.54	o
3	9	11	71.55	-625.51	625.51	o	0	4	12	863.55	1140.42	545.77	o
4	9	11	518.63	-170.83	601.96	o	1	4	12	674.84	846.62	545.42	o
5	9	11	124.54	698.73	609.36	o	2	4	12	257.31	276.38	520.25	o
6	9	11	81.1	-544.29	625.52	o	3	4	12	815.2	260.61	570.08	o
7	9	11	260.48	406.39	658.36	o	4	4	12	8.24	-73.1	560.42	o
8	9	11	97.08	-292.46	666.17	o	0	5	12	5.1	-576.66	576.66	o
9	9	11	190.85	-552.57	747.6	o	1	5	12	488.36	1260.93	561.32	o
0	10	11	1150.64	1092.09	643.84	o	2	5	12	71.09	194.97	552.41	o
1	10	11	86.8	1177.96	625.54	o	3	5	12	171.28	73.14	568.84	o
2	10	11	73.61	-146.22	617.39	o	4	5	12	632.42	-626.65	626.65	o
3	10	11	258.19	146.3	625.86	o	5	5	12	155.55	1121.36	528.18	o
4	10	11	237.31	780.23	666.45	o	0	6	12	222.91	-243.83	560.81	o
5	10	11	314.39	1544.54	642.2	o	1	6	12	894.14	1392.87	562.04	o
6	10	11	167.52	-97.51	650.05	o	2	6	12	102.99	552.46	552.46	o
7	10	11	63.31	-398.04	714.84	o	3	6	12	80.32	381.82	560.55	o
8	10	11	84	0	723	o	4	6	12	475.76	1781.34	544.98	o
9	10	11	105.18	-32.5	723.04	o	5	6	12	7.86	-568.54	601.03	o
10	10	11	1.2	-739.08	739.08	o	6	6	12	327.89	812.99	601.61	o
0	11	11	3827.58	7103.25	780.13	o	0	7	12	720.52	1221.05	586.11	o
1	11	11	85.73	1137.33	649.9	o	1	7	12	57.13	568.63	576.75	o
2	11	11	13.79	714.74	641.65	o	2	7	12	103.7	422.47	560.59	o
3	11	11	550.03	-284.71	691.44	o	3	7	12	161.59	430.67	568.81	o
4	11	11	38	-129.96	649.81	o	4	7	12	26.59	-633.55	633.55	o
5	11	11	2528.96	2282.03	719.78	o	5	7	12	412.08	-56.92	618.02	o
6	11	11	1487.46	1940.93	742.12	o	6	7	12	628.1	1114.76	634.68	o
7	11	11	3.69	-560.41	690.36	o	7	7	12	22.35	1315.81	617.29	o
8	11	11	249.76	219.43	763.96	o	0	8	12	24.57	-276.16	609.18	o
1	8	12	916.03	1514.91	635.28	o	11	11	12	3.52	1120.81	795.94	o
2	8	12	456.83	780.78	618.12	o	0	12	12	292.52	105.67	747.8	o
3	8	12	0.18	397.97	592.89	o	1	12	12	289.18	804.69	755.92	o
4	8	12	756.36	1041.94	659.35	o	2	12	12	59.12	-243.69	755.45	o
5	8	12	231.47	-642.06	642.06	o	3	12	12	367.18	463.4	739.82	o
6	8	12	53.65	-154.34	649.84	o	4	12	12	51.39	503.62	763.55	o
7	8	12	377.28	-512.22	699.22	o	5	12	12	79.09	-771.73	771.73	o
8	8	12	154.85	-747.53	747.53	o	6	12	12	258.22	0	739.58	o
0	9	12	670.84	-634.77	634.77	o	7	12	12	103.53	308.71	804.27	o
1	9	12	34.39	-259.92	633.56	o	8	12	12	272.02	-162.55	731.47	o
2	9	12	934.31	496.8	684.12	o	9	12	12	20.39	292.4	747.24	o
3	9	12	308.95	-479.62	674.72	o	10	12	12	240.84	-885.81	885.81	o
4	9	12	114.76	333.1	690.58	o	0	0	13	143.38	-422.54	568.8	o
5	9	12	93.84	316.83	641.79	o	0	1	13	13.56	48.73	536.06	o
6	9	12	146.19	97.5	658.14	o	1	1	13	229.32	1235.45	512.06	o
7	9	12	105.74	-203.1	698.68	o	0	2	13	399.15	935.24	544.88	o
8	9	12	437.54	-455.36	739.96	o	1	2	13	303.11	943.07	536.58	o
9	9	12	298.22	-438.93	723.42	o	2	2	13	26.5	121.84	568.57	o
0	10	12	60.22	341.17	674.22	o	0	3	13	506.46	244.06	561.33	o
1	10	12	229.45	707.06	707.06	o	1	3	13	7.38	-576.66	576.66	o
2	10	12	0.05	308.63	706.59	o	2	3	13	818.12	1099.36	570.04	o
3	10	12	42.68	-698.56	698.56	o	3	3	13	1259.53	709.46	603.45	o
4	10	12	394.89	1902.62	682.99	o	0	4	13	140.9	97.51	576.91	o
5	10	12	270.06	-227.58	699	o	1	4	13	277.56	959.23	536.52	o
6	10	12	411.41	609.82	723.66	o	2	4	13	139.56	-203.14	560.65	o
7	10	12	96.06	422.44	698.65	o	3	4	13	103.05	1169.92	560.59	o
8	10	12	230.68	-32.51	723.28	o	4	4	13	126.15	24.37	601.25	o
9	10	12	0.03	-268.02	771.57	o	0	5	13	707.84	1123.32	553.52	o
10	10	12	91.98	511.79	779.87	o	1	5	13	0.12	-568.52	568.52	o
0	11	12	24.29	357.38	657.91	o	2	5	13	164.13	576.94	552.57	o
1	11	12	315.21	-780.38	780.38	o	3	5	13	253.38	211.33	617.74	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
2	11	12	78.9	658.01	723	o	4	5	13	220.11	81.27	609.54	o
3	11	12	7.83	-666	747.22	o	5	5	13	4.45	308.63	552.29	o
4	11	12	560.4	748.36	732.09	o	0	6	13	227.76	625.82	568.93	o
5	11	12	3.84	1535.03	674.11	o	1	6	13	3.96	-186.8	601.02	o
6	11	12	58.72	787.94	706.71	o	2	6	13	9.74	-162.44	617.27	o
7	11	12	378.59	-113.82	764.21	o	3	6	13	19.1	641.66	584.8	o
8	11	12	25.06	-381.75	812.23	o	4	6	13	60.23	203.08	617.37	o
9	11	12	106.3	0	739.28	o	5	6	13	176.28	268.16	617.58	o
10	11	12	93.53	1779.09	755.5	o	6	6	13	229.54	-520.14	690.81	o
0	7	13	57.3	-227.45	625.48	o	2	11	13	95.21	568.67	723.02	o
1	7	13	343.61	227.65	609.77	o	3	11	13	109.17	-609.31	755.55	o
2	7	13	143.17	455.01	625.65	o	4	11	13	46.83	-787.91	787.91	o
3	7	13	23.66	-235.55	674.15	o	5	11	13	87.49	-406.18	747.38	o
4	7	13	291.36	268.25	666.56	o	6	11	13	79.05	-763.6	763.6	o
5	7	13	92.29	-682.41	682.41	o	7	11	13	6.87	-812.19	812.19	o
6	7	13	1.98	-714.72	714.72	o	8	11	13	108.56	-812.4	812.4	o
7	7	13	627.12	593.93	707.84	o	9	11	13	93.16	211.22	836.74	o
0	8	13	733.16	683.7	675.57	o	10	11	13	18.82	609.16	812.21	o
1	8	13	69.99	389.93	641.75	o	0	12	13	0.01	0	787.81	o
2	8	13	518.62	959.83	658.86	o	1	12	13	55.93	-259.94	747.31	o
3	8	13	2707.3	1473.41	720.33	o	2	12	13	124.48	73.12	779.95	o
4	8	13	60.35	657.98	625.48	o	3	12	13	124.18	641.83	771.82	o
5	8	13	78.64	-446.8	698.63	o	4	12	13	100.03	40.62	779.89	o
6	8	13	36.59	-560.46	747.28	o	5	12	13	7.27	1137.07	722.85	o
7	8	13	0.28	-779.69	779.69	o	6	12	13	45.77	-787.9	787.9	o
8	8	13	2026.44	2074.23	751.29	o	7	12	13	91.98	1023.58	796.12	o
0	9	13	877.14	692.09	732.8	o	8	12	13	40.2	-592.95	836.63	o
1	9	13	26.7	186.81	682.28	o	0	13	13	0.14	1372.58	795.93	o
2	9	13	16.56	601.04	674.14	o	1	13	13	41.9	-747.28	771.65	o
3	9	13	195.58	967.03	674.48	o	2	13	13	146.65	0	763.74	o
4	9	13	598.38	496.26	691.51	o	3	13	13	453.32	-813.13	813.13	o
5	9	13	29.69	268.04	641.67	o	4	13	13	0.43	-779.69	779.69	o
6	9	13	12.49	722.86	706.62	o	5	13	13	180.59	-658.17	828.8	o
7	9	13	124.17	-389.98	739.33	o	6	13	13	6.25	-129.95	763.46	o
8	9	13	216.39	-837.02	837.02	o	7	13	13	12.03	-730.98	828.44	o
9	9	13	11.97	-755.35	755.35	o	0	0	14	207.13	81.27	593.28	o
0	10	13	79.57	-333.07	714.87	o	0	1	14	12.25	48.73	584.79	o
1	10	13	109.51	-333.09	731.18	o	1	1	14	186.51	211.29	593.24	o
2	10	13	11.31	430.47	682.25	o	0	2	14	501.68	1439.81	593.82	o
3	10	13	8.51	-471.07	722.85	o	1	2	14	225.14	162.55	593.31	o
4	10	13	94.34	-714.9	714.9	o	2	2	14	37.42	268.05	601.08	o
5	10	13	9.29	-178.68	755.34	o	0	3	14	464.49	325.34	577.48	o
6	10	13	29.09	722.89	731.02	o	1	3	14	15.68	-609.16	609.16	o
7	10	13	103.97	-779.9	779.9	o	2	3	14	651.49	-162.76	642.91	o
8	10	13	0.69	852.79	763.45	o	3	3	14	986.31	1148.61	643.55	o
9	10	13	0.54	-121.83	722.84	o	0	4	14	167.73	-650.08	650.08	o
10	10	13	80.91	1332.24	771.73	o	1	4	14	305.33	-585.31	617.83	o
0	11	13	190.33	276.28	715.09	o	2	4	14	142.38	1040.04	609.4	o
1	11	13	7.21	0	763.46	o	3	4	14	285.25	-260.12	634.04	o
4	4	14	706.41	496.47	651.11	o	9	9	14	29.9	1104.64	714.77	o
0	5	14	303.88	682.85	609.69	o	0	10	14	24.13	389.87	739.13	o
1	5	14	23.96	-32.49	617.3	o	1	10	14	107.31	32.5	723.05	o
2	5	14	247.9	349.5	601.46	o	2	10	14	10.83	-755.35	763.47	o
3	5	14	180.24	585.08	585.08	o	3	10	14	5.97	2574.64	722.85	o
4	5	14	92.57	1023.62	584.93	o	4	10	14	147.37	552.5	723.12	o
5	5	14	10.6	154.32	625.4	o	5	10	14	2.63	-601.01	787.82	o
0	6	14	38.52	-121.84	617.32	o	6	10	14	2.87	-820.3	820.3	o
1	6	14	8.4	235.54	617.27	o	7	10	14	83.16	-276.2	771.73	o
2	6	14	42.9	1259.03	649.82	o	8	10	14	2.06	-81.22	706.6	o
3	6	14	18.33	365.5	657.9	o	9	10	14	14.89	503.57	787.84	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
4	6	14	43.54	560.47	625.45	o	0	11	14	27.61	146.2	755.38	o
5	6	14	31.39	414.25	649.8	o	1	11	14	1.02	-227.41	804.06	o
6	6	14	67.3	-690.48	690.48	o	2	11	14	144.54	0	747.49	o
0	7	14	42.28	1112.82	674.19	o	3	11	14	154.14	1941.89	771.88	o
1	7	14	266.95	-32.51	674.63	o	4	11	14	23.26	154.32	739.13	o
2	7	14	112.1	1064.3	658.08	o	5	11	14	10.61	-674.13	820.32	o
3	7	14	57.29	381.79	690.46	o	6	11	14	0.87	-649.74	779.69	o
4	7	14	324	-32.52	707.24	o	7	11	14	26.2	0	844.72	o
5	7	14	177.17	-32.5	715.07	o	8	11	14	136.51	-511.85	861.2	o
6	7	14	2.35	-308.63	698.48	o	0	12	14	54.34	666.08	747.31	o
7	7	14	492.8	317.17	691.27	o	1	12	14	15.25	81.22	812.21	o
0	8	14	325.72	-455.24	723.51	o	2	12	14	74.87	-820.46	820.46	o
1	8	14	104.35	-292.47	714.93	o	3	12	14	110.19	-162.48	804.28	o
2	8	14	514.72	292.81	715.75	o	4	12	14	204.05	-771.97	771.97	o
3	8	14	2199.56	1993.9	825.34	o	5	12	14	0.21	787.81	755.32	o
4	8	14	65.44	-568.63	731.09	o	6	12	14	3.37	422.34	820.31	o
5	8	14	175.61	1031.96	698.81	o	7	12	14	68.67	-129.97	812.32	o
6	8	14	60.65	-779.82	779.82	o	0	13	14	33.45	576.69	869.1	o
7	8	14	1.03	1388.83	698.47	o	1	13	14	23.1	-162.44	804.1	o
8	8	14	1401.5	611.38	774.41	o	2	13	14	121.49	836.8	779.93	o
0	9	14	446.09	-65.05	740	o	3	13	14	322.18	-755.94	820.97	o
1	9	14	128.5	-739.34	739.34	o	4	13	14	30.37	641.67	795.99	o
2	9	14	49.76	105.6	722.94	o	5	13	14	147.38	-40.62	836.85	o
3	9	14	96.78	-788.02	788.02	o	0	0	15	8.65	609.15	617.27	o
4	9	14	711.65	1407.79	699.82	o	0	1	15	570.59	406.78	675.26	o
5	9	14	11.32	609.15	698.49	o	1	1	15	111.79	113.74	593.09	o
6	9	14	0.46	-592.89	763.45	o	0	2	15	150.73	528.15	617.53	o
7	9	14	229.35	-804.54	804.54	o	1	2	15	325.32	268.28	642.24	o
8	9	14	56.57	2168.83	706.7	o	2	2	15	327.38	-178.85	666.63	o
0	3	15	119.65	235.61	633.72	o	1	9	15	27.97	251.79	747.26	o
1	3	15	195.92	-674.5	674.5	o	2	9	15	384.89	512.2	780.49	o
2	3	15	106	633.69	641.82	o	3	9	15	97.39	-779.89	779.89	o
3	3	15	11.38	982.77	641.64	o	4	9	15	99.81	-113.73	796.14	o
0	4	15	542.4	-333.52	667.04	o	5	9	15	45.07	40.61	747.29	o
1	4	15	159.93	-455.03	682.55	o	6	9	15	27.43	828.48	787.87	o
2	4	15	179.87	495.69	641.95	o	7	9	15	117.34	446.83	796.17	o
3	4	15	345.97	97.56	699.17	o	8	9	15	200.83	-544.44	804.46	o
4	4	15	4.43	1210.16	625.38	o	9	9	15	148.02	-471.24	844.98	o
0	5	15	9.7	406.1	657.88	o	0	10	15	146	-381.87	755.61	o
1	5	15	281.74	186.95	682.78	o	1	10	15	114.24	1486.73	731.18	o
2	5	15	0.02	284.26	665.98	o	2	10	15	0.29	536.04	779.69	o
3	5	15	27.28	446.73	682.28	o	3	10	15	5.43	617.26	755.33	o
4	5	15	386.51	1739.95	609.79	o	4	10	15	121.19	-788.06	788.06	o
5	5	15	3.04	-641.62	722.84	o	5	10	15	145.71	519.99	771.86	o
0	6	15	103.04	-227.48	714.92	o	6	10	15	263.24	-121.91	796.47	o
1	6	15	405.23	390.29	748.06	o	7	10	15	45.91	1007.22	828.52	o
2	6	15	20.87	227.42	706.64	o	8	10	15	73.08	170.59	771.71	o
3	6	15	150.97	-195	731.27	o	0	11	15	3.51	162.44	804.06	o
4	6	15	177.3	-341.28	698.82	o	1	11	15	88.5	1080.44	755.5	o
5	6	15	7.58	349.24	698.49	o	2	11	15	6.32	-121.83	787.82	o
6	6	15	67.8	-747.34	747.34	o	3	11	15	4.16	1340.11	763.45	o
0	7	15	468.11	-32.53	707.51	o	4	11	15	280.97	1154.12	747.74	o
1	7	15	23.82	-755.37	755.37	o	5	11	15	1.35	-901.52	901.52	o
2	7	15	189.62	341.29	723.22	o	6	11	15	57.23	-430.52	787.92	o
3	7	15	312.42	1373.76	747.85	o	0	12	15	324.35	918.51	804.72	o
4	7	15	0.27	487.31	730.96	o	1	12	15	68.74	463.02	820.44	o
5	7	15	274.55	-390.13	731.5	o	2	12	15	130.25	-121.87	836.82	o
6	7	15	141.89	-113.75	731.24	o	3	12	15	207.09	-292.54	812.6	o
7	7	15	29.99	1023.42	731.02	o	4	12	15	0.03	1112.68	828.42	o
0	8	15	5.71	592.9	714.73	o	0	0	16	4620.83	4451.45	789.91	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
1	8	15	440.72	1016.44	731.84	o	0	1	16	116.58	349.35	690.58	o
2	8	15	153.29	-243.75	755.64	o	1	1	16	180.45	284.41	666.33	o
3	8	15	14.2	-495.45	771.6	o	0	2	16	8.98	999	666	o
4	8	15	219.53	251.92	772.02	o	1	2	16	4.11	-682.24	739.09	o
5	8	15	25.45	739.13	763.5	o	2	2	16	262.68	1072.87	723.37	o
6	8	15	55.71	-747.31	747.31	o	0	3	16	247.86	1210.99	690.83	o
7	8	15	157.05	1137.51	788.13	o	1	3	16	65.07	-714.85	714.85	o
8	8	15	6.69	243.66	812.19	o	2	3	16	273.38	560.83	707.14	o
0	9	15	370.52	-471.53	796.72	o	3	3	16	497.41	333.45	756.37	o
0	4	16	1.91	-64.97	698.48	o	5	9	16	2.92	-454.82	730.96	o
1	4	16	267.4	-698.99	772.14	o	6	9	16	6.27	1551.28	730.97	o
2	4	16	114.8	471.21	706.82	o	7	9	16	133.17	259.98	796.2	o
3	4	16	31.95	373.63	722.9	o	0	10	16	342.76	-325.16	821.02	o
4	4	16	248.42	625.8	633.93	o	1	10	16	26.16	-804.11	804.11	o
0	5	16	2404.92	1136.4	809.38	o	2	10	16	17.6	487.33	795.97	o
1	5	16	132.29	1064.34	714.98	o	3	10	16	28.21	40.61	820.36	o
2	5	16	73.21	414.29	739.23	o	4	10	16	64.63	503.63	796.06	o
3	5	16	120.07	-32.5	731.2	o	5	10	16	249.47	251.93	763.92	o
4	5	16	53.23	-389.9	739.19	o	0	11	16	1673.25	1574.1	840.06	o
5	5	16	1368.84	1931.92	741.79	o	1	11	16	118.58	81.24	796.17	o
0	6	16	1518.44	1940.96	750.29	o	2	11	16	13.76	-909.67	909.67	o
1	6	16	4.57	633.51	730.97	o	3	11	16	75.34	-511.77	828.58	o
2	6	16	6.42	495.44	771.58	o	0	0	17	1643.35	1256.36	726.08	o
3	6	16	101.07	-178.73	796.15	o	0	1	17	34.71	105.59	706.66	o
4	6	16	126.93	-178.74	731.21	o	1	1	17	589.97	1407.33	740.27	o
5	6	16	757.45	1009.12	773.12	o	0	2	17	16.14	316.76	771.6	o
6	6	16	285.74	308.86	739.63	o	1	2	17	4.24	-73.1	755.33	o
0	7	16	2.45	511.67	698.48	o	2	2	17	2.88	1421.32	665.99	o
1	7	16	241.42	-178.79	747.69	o	0	3	17	186.52	0	788.21	o
2	7	16	187.24	731.32	771.95	o	1	3	17	291.38	-357.64	723.41	o
3	7	16	1.84	146.19	722.84	o	2	3	17	68.4	901.68	698.6	o
4	7	16	238.9	-715.17	763.93	o	3	3	17	42.17	-796.02	796.02	o
5	7	16	8.32	925.9	771.58	o	0	4	17	295.06	0	772.18	o
6	7	16	42.49	1259.01	747.28	o	1	4	17	261.84	1276.01	731.47	o
7	7	16	204.76	1129.52	755.72	o	2	4	17	12.81	-795.96	795.96	o
0	8	16	365.23	-113.81	804.83	o	3	4	17	144.42	-519.99	739.36	o
1	8	16	48.85	-300.54	787.91	o	4	4	17	10.1	414.22	722.86	o
2	8	16	169.22	495.65	771.91	o	0	5	17	163.28	219.38	739.4	o
3	8	16	266.66	227.57	780.23	o	1	5	17	125.2	-796.2	796.2	o
4	8	16	0.96	-812.18	812.18	o	2	5	17	1.74	-739.08	739.08	o
5	8	16	268.27	-552.66	804.61	o	3	5	17	31.13	-682.28	731.02	o
6	8	16	179.61	203.14	828.8	o	4	5	17	168.87	-73.13	780.03	o
7	8	16	45.4	414.26	820.39	o	5	5	17	57.71	1291.55	666.08	o
8	8	16	225.02	40.63	812.63	o	0	6	17	192.27	650.07	796.34	o
0	9	16	21.71	-804.1	804.1	o	1	6	17	115.5	-763.68	763.68	o
1	9	16	114.45	-666.18	820.54	o	2	6	17	10.46	227.42	747.22	o
2	9	16	73.59	276.19	763.59	o	3	6	17	71.14	-812.33	812.33	o
3	9	16	70.03	-796.08	796.08	o	4	6	17	274.47	-121.91	747.73	o
4	9	16	142.66	-365.61	763.73	o	5	6	17	0.01	357.36	795.93	o
6	6	17	23.8	576.68	747.25	o	2	5	18	4.7	-763.46	828.43	o
0	7	17	54.66	113.72	787.92	o	3	5	18	53.34	568.6	763.55	o
1	7	17	134.85	-511.85	763.71	o	4	5	18	20	747.24	812.22	o
2	7	17	3.64	397.97	755.33	o	5	5	18	35.56	925.96	771.64	o
3	7	17	293.82	-235.71	829.05	o	0	6	18	0.72	860.91	763.45	o
4	7	17	16.8	1088.36	755.36	o	1	6	18	6.96	-495.44	820.31	o
5	7	17	92.28	1316.04	771.75	o	2	6	18	1.07	0	820.3	o
6	7	17	69.49	-714.84	828.57	o	3	6	18	6.6	-544.17	795.95	o
7	7	17	0.25	-519.79	844.66	o	4	6	18	56.84	926.01	771.68	o
0	8	17	205.42	-284.41	820.73	o	5	6	18	2.39	-341.12	763.45	o
1	8	17	98.61	-528.05	836.75	o	0	7	18	161.88	585.01	885.63	o

h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)		h	k	l	Fcal^2	Fobs^2	σ(Fobs^2)	
2	8	17	9.64	-690.37	787.83	o	1	7	18	0.35	-129.95	779.69	o
3	8	17	29.66	1112.77	755.38	o	2	7	18	63.83	-763.57	844.8	o
4	8	17	67.24	121.85	812.31	o	3	7	18	97.62	-89.36	877.37	o
5	8	17	78.38	40.62	844.83	o	4	7	18	70.44	1421.56	812.32	o
6	8	17	159.36	130	812.5	o	0	8	18	3.82	-836.55	836.55	o
0	9	17	21.82	-462.97	812.22	o	1	8	18	71.16	-836.69	836.69	o
1	9	17	70.18	333.05	836.69	o	0	0	19	347.34	-536.51	788.51	o
2	9	17	6.74	-333	852.8	o	0	1	19	5.94	1153.31	812.19	o
3	9	17	73.31	804.2	796.08	o	1	1	19	100.43	1771	804.26	o
4	9	17	28.42	560.44	763.5	o	0	2	19	170.49	-121.88	828.78	o
0	10	17	37.17	-430.49	852.86	o	1	2	19	90.48	2485.83	763.62	o
1	10	17	146.27	430.61	869.35	o	2	2	19	50.78	1039.72	820.4	o
0	0	18	440.86	796.86	756.2	o	0	3	19	275.09	-170.68	788.36	o
0	1	18	38.99	414.25	779.77	o	1	3	19	34.07	1592	796	o
1	1	18	34.01	113.71	755.39	o	2	3	19	342.2	1723.3	772.23	o
0	2	18	164.76	-763.78	763.78	o	3	3	19	1485.72	-40.76	855.97	o
1	2	18	54.09	381.78	706.69	o	0	4	19	30.44	40.61	779.75	o
2	2	18	26.34	-795.99	795.99	o	1	4	19	144.1	-844.97	844.97	o
0	3	18	69.53	739.21	796.08	o	2	4	19	83.97	-170.59	852.97	o
1	3	18	152.78	-268.13	747.5	o	3	4	19	8.68	-211.17	828.44	o
2	3	18	0.69	1128.93	747.2	o	0	5	19	60.96	511.75	836.67	o
3	3	18	231.66	-81.27	820.79	o	1	5	19	71.1	-259.94	861.06	o
0	4	18	281.42	861.54	764	o	2	5	19	1.42	739.08	836.54	o
1	4	18	0	-81.22	795.93	o							
2	4	18	132.48	-796.21	796.21	o							
3	4	18	60.29	-804.18	804.18	o							
4	4	18	209.58	-812.61	812.61	o							
0	5	18	5.72	-804.07	804.07	o							
1	5	18	33.27	162.45	763.51	o							