Chemistry of the UO_2^{2+} Group. II. Preparation and Properties of $(C_6H_5)_3PO$ and $(C_6H_5)_3AsO$ Complexes of Uranyl Acetate and the Structure of $(UO_2(CH_3COO)_2(C_6H_5)_3PO)_2$

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Monomeric complexes of formula $UO_2(CH_3COO)_2L_2$ and dimeric complexes of formula $(UO_2(CH_3COO)_2L)_2$, with $L = (C_6H_5)_3$ -PO and $(C_6H_5)_3AsO$, were synthesized from uranyl acetate dihydrate and triphenylphosphine and triphenylarsine oxides, respectively. The reaction products have been characterized *via* elemental analyses, conductivity measurements, and infrared spectral data. The crystal structure of *trans*-di- μ -acetato-bis(dioxotriphenylphosphine oxide acetate)diuranium(VI), $(UO_2(CH_3COO)_2(C_6H_3)_3PO)_2$, has been determined from three-dimensional, single-crystal X-ray film data and refined to a final R value of 0.095. The crystals are triclinic, space group PI, with one formula unit in a cell with the dimensions a =8.346 Å, b = 11.022 Å, c = 13.680 Å, $\alpha = 101^{\circ}$ 50', $\beta = 91^{\circ}$ 12', $\gamma = 109^{\circ}$ 12', $D_m = 1.91$ g cm⁻³, and $D_x = 1.93$ g cm⁻³. The structure consists of dimeric molecules in which the uranium atoms are seven-coordinated. The coordination polyhedron is a pentagonal bipyramid with an almost linear uranyl group perpendicular to five oxygen atoms at the corners of an irregular pentagonal base. The dimeric units are realized by bridging acetato groups.

Introduction

Several uranyl complexes with triphenylphosphine and triphenylarsine oxides^{1,2} have been reported.

The reactions of uranyl nitrate hexahydrate with pure triphenylphosphine and triphenylarsine give products of the type $UO_2(NO_3)_2L_2$ (where L = $(C_6H_5)_3PO$ and $(C_6H_5)_3AsO$) as reported in the literature.¹⁻⁵ In such complexes two ligand molecules have displaced two water molecules coordinated to the central uranyl group and the two nitrate groups act as bidentate chelating agents^{3,6} leading to the common hexacoordinated structure. However the chemical behavior of $(C_6H_5)_3PO$ and $(C_6H_5)_3AsO$ toward uranyl acetate dihydrate is not known.

Ir studies⁷ have shown that the acetate as well as the nitrate groups⁸ act as chelating bidentate ligands for the uranyl group. On this basis, analogous behavior of $(C_6H_5)_3PO$ an $(C_6H_5)_3AsO$ toward the above uranyl salts could be expected in principle. Thus, we have examined the reaction products of uranyl acetate dihydrate with triphenylphosphine and triphenylarsine oxides. In addition to the expected species, a new dimeric complex with the formula $(UO_2(CH_3 COO)_2(C_6H_5)_3PO)_2$, di- μ -acetato-bis(dioxotriphenylphosphine oxide acetate)diuranium(VI), was synthesized and its structure determined.

Experimental Section

Preparations. Triphenylphosphine Oxide.—Triphenylphosphine oxide was prepared according to the method described by Canziani, *et al.*⁹

Triphenylarsine Oxide.—Triphenylarsine oxide was obtained as described in the literature.¹⁰

Dimer of Diacetatodioxo(triphenylphosphine oxide)uranium-(VI).—A cold acetone solution of uranyl acetate dihydrate and triphenylphosphine oxide (molar ratio 1:1) was stirred for 20–30 min yielding light yellow microcrystals of diacetatodioxo(triphenylphosphine oxide)uranium(VI). They were washed with acetone and recrystallized from nitromethane-benzene, mp 252– 253°. Anal. Calcd for ($C_{22}H_{21}O_7PU$)₂: C, 39.7; H, 3.2; U, 35.7. Found: C, 39.8, 39.6; H, 3.1, 3.2; U, 36.0, 35.9.

Diacetatodioxobis(triphenylphosphine oxide)uranium(VI).—A nitromethane solution of the same salt and the appropriate ligand (molar ratio 1:2.5) was stirred at room temperature for 20 min and then evaporated to dryness in air. After nearly 2 days green-yellow crystals of diacetatodioxobis(triphenylphosphine oxide)uranium(VI) were obtained. The excess of ligand was removed with cold acetone. This complex, dried under vacuum, melted at 208–210°. It was not recrystallized because every attempt to purify it from hot acetone or dioxane produces the mono(triphenylphosphine oxide) derivative. Anal. Caled for C₄₀H₃₅O₄P₂U: C, 50.8; H, 3.8; U, 25.2. Found: C, 49.9, 50.7; H, 3.8, 3.7; U, 25.1, 25.2.

Dimer of Diacetatodioxo(triphenylarsine oxide)uranium. (VI).—A cold methanol solution of the same salt and triphenylarsine oxide (molar ratio 1:1) was stirred for about 20 min. A green-yellow precipitate was obtained. The solution was filtered off and a yellow crystalline solid was recovered by slow evaporation in air. The product was recrystallized from hot nitromethane, mp 245°. *Anal.* Calcd for $(C_{22}H_{21}O_7AsU)_2$: C, 37.2; H, 2.9; U, 33.6. Found: C, 36.5, 36.7; H, 2.7, 2.8; U, 35.0, 33.6.

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ratio 1:2) was stirred for a few minutes. A green-yellow crystalline precipitate was obtained. It was separated, washed with cold acetone, recrystallized from dichloromethane, and washed with the same solvent, mp 260°. *Anal.* Calcd for $C_{40}H_{36}O_8$ -As₂U: C, 46.5; H, 3.5; U, 23.0. Found: C, 46.5, 45.5; H, 3.7, 3.3; U, 22.9, 23.2.

All of the complexes described above are soluble in many organic solvents but insoluble in water, benzene, ethyl ether, and carbon tetrachloride.

Molecular Weight Determination.—The molecular weights of the reported complexes were determined at 37° in solutions of nitromethane and sym-dichloroethane by a Mechrolab osmometer, Model 302. See Table I.

TABLE I MOLECULAR WEIGHT VALUES OF THE URANYL ACETATE COMPLEXES

		Found		
Complexes	Calcd	Nitro- methane	sym- Dichloro- ethane	
$(UO_2(CH_3COO)_2(C_6H_5)_3PO)_2$	1332	a	1000	
$(UO_2(CH_3COO)_2(C_6H_5)_3AsO)_2$	1420	1143	1160	
$UO_2(CH_3COO)_2((C_6H_5)_3PO)_2$	944	600	615	
$UO_2(CH_3COO)_2((C_6H_5)_3AsO)_2$	1032	819	851	

^a This measurement was not possible because the compound undergoes slow decomposition in this solvent.

Conductance Measurements.—The measurements were carried out at 25° with a Type 3216-B LKB conductivity bridge. The values of molar conductivity are reported in Table II.

TABLE II

Molar Conductance Values at 25° (in ohm⁻¹ cm²) of the Uranyl Acetate Complexes

	<u></u>		
Complexes (10 ⁻³ M)	Nitromethane	Dichloro- ethane	Dichloro- methane
$(UO_2(CH_3COO)_2(C_6H_5)_3PO)_2$	6.4^{a}	2.1	2.5
$(UO_2(CH_3COO)_2(C_6H_5)_3AsO)_2$	12.4	8.1	11.3
$UO_2(CH_3COO)_2((C_6H_5)_3PO)_2$	11.9	2.4	3.2
$UO_2(CH_3COO)_2((C_6H_5)_3AsO)_2$	22.7	11.9	18.8

^a The compound undergoes slow decomposition in this solvent.

Infrared Spectra.—The infrared spectra in the region 1300– 450 cm⁻¹ were recorded by Perkin-Elmer infrared spectrophotometer, Model 337. Nujol mulls between KBr plates were employed. The absence of any absorption in the range 3600– 3200 cm⁻¹ indicated that all of the examined samples were anhydrous compounds. The absorption frequencies of P–O and As–O bonds and the asymmetric stretching frequency of the uranyl group are reported in Table III.

TABLE III

ABSORPTION	FREQUENCIES ((CM ⁻¹) OF THE
URANY	I. ACETATE CON	PLEXES

Compounds	U-Oª	P-O	As-O				
$(C_6H_5)_3PO$		1195^{b}					
(C ₆ H ₅) ₃ AsO	• • •		880°				
$UO_2(CH_3COO)_2(H_2O)_2$	940		• • •				
$(UO_2(CH_3COO)_2(C_6H_5)_3PO)_2$	926, 920	1138, 1120	•••				
$(\mathrm{UO}_2(\mathrm{CH_3COO})_2(\mathrm{C_6H_5})_3\mathrm{AsO})_2$	921, 910	· · · ·	880				
$UO_2(CH_3COO)_2((C_8H_5)_3PO)_2$	912,905	1150, 1132					
$UO_2(CH_3COO)_2((C_6H_5)_3AsO)_2$	895		871, 867				

^a U-O (uranyl) bond asymmetric stretching frequency. ^b F. A. Cotton, R. D. Barnes, and E. Bannister, J. Chem. Soc., 2199 (1960). ^c D. M. L. Goodgame and F. A. Cotton, *ibid.*, 2298 (1961).

Crystal Structure of $(UO_2(CH_3COO)_2(C_6H_5)_3PO)_2$

Crystal Data.—Vellow *a* prismatic crystals of $(UO_2-(CH_3COO)_2(C_6H_5)_3PO)_2$, $C_{44}H_{42}O_{14}P_2U_2$, were grown from acetone solutions. The density was measured by flotation in a mixture of *sym*-tetrabromoethane and *sym*-dichloroethane. The cell dimensions were determined from precession photographs taken with Cu K α radiation (λ 1.5418 Å). Space group PI was chosen from structure determination.

The compound has been formulated as *trans*-di- μ -acetato-bis(dioxotriphenylphosphine oxide acetate)diuranium(VI), mol wt 1332.5, mp 252-253°. The crystals are triclinic and the space group is PI (assumed and verified by the structure): $a = 8.346 \pm 0.012$ Å, $b = 11.022 \pm 0.016$ Å, $c = 13.680 \pm 0.018$ Å, $\alpha =$ $101^{\circ} 50' \pm 10'$, $\beta = 91^{\circ} 12' \pm 10'$, $\gamma = 109^{\circ} 12' \pm$ 10', V = 1147.5 Å³, $D_{\rm m} = 1.91$ g cm⁻³, $D_{\rm x} = 1.93$ g cm⁻³, and Z = 1. The absorption coefficient for Cu K α radiation is $\mu = 133$ cm⁻¹ [F(000) = 632].

Intensity Data.—A single-crystal specimen was selected for the X-ray work, of over-all dimensions about 0.2, 0.1, and 0.1 mm. The three-dimensional intensity data of 0kl-4kl were collected from equiinclination Weissenberg photographs taken about the *a* axis using the multiple-film technique with Cu K α radiation. Reflections hk0 and h0l were collected with the precession method. The intensities were measured with the Joyce–Loebl Flying Spot microdensitometer and corrected for Lorentz and polarization factors. Since the μR value was about 0.7, correction for absorption was made as for a cylindrical specimen. A total of 1717 independent structure factors were finally evaluated. The form factors for the neutral atoms were taken from Forsyth and Wells.¹¹

Determination of the Structure

From the h0l and 0kl Patterson projections the relative positions of the uranium and phosphorus atoms were derived. The two U-P vectors in both projections were related by a twofold symmetry. Moreover, the most probable situation being that the dimeric molecule was centrosymmetric, space group P1 was assumed. The different layers were then placed on a relative scale by comparison with the precession data and in absolute scale by comparison with the structure factors calculated with the heavy-atom coordinates and an over-all temperature factor of 2 $Å^2$. The trial model of the structure was deduced from a three-dimensional Fourier electron density summation which confirmed the choice of the centrosymmetric space group. The reliability index at this stage was 0.16. The obtained parameters were refined by the block-diagonal least-squares approximation using the Cruickshank weighting scheme and individual isotropic temperature factors. Four cycles of structure factors and least-squares improved R to 0.12. Scale factor adjustment reduced R to 0.115. At this point a care-

(11) J. B. Forsyth and M. Wells, Acta Cryst., 12, 412 (1959).

TABLE IV

Observed and Calculated Structure Factors $(\times 10)$

	# H K L FOI FG	• H K & Fat Fc	e H K L FOT FC	• 11 jk i FOS FG	анн _с Foi Fo	• H K. (Fat ic	• H # L FO1 gFC	• H K L FOT FC	•, Н. К. Ц. FO1 – FC
	# 02+07+07* 1291+ 1479+ 1 03+00+00+ 1108+ 1126+ 1 84+00+07+ 0754+ 0584+	1 03.07-01- 1891- 0545- 1 04.07-01- 0495- 0525- 0 04.07-01- 0845- 1039-	8 03+01+03- 100A- 1221- 1 03+01-02- 0844- 0859- 1 04+01-03+ 0921- 1035-	1 00+05-041 188A- 0821- 1 01+05+04+ 1014+ 0551+ 1 01+05+04+ 148A- 1424-	0 04+10+65= 0345= 0459= 1 00+11+05+ 0358= 0459= 1 00+11+05+ 0551= 0586=	1 02+05+07= 0508+ 0465+ 1 03+04-07+ 0716+ 0700+ 1 03+05+07- 0725+ 0535+ 0 04+05+07- 0330+ 0380+	1 04+01+09+ 0515+ 0520+ 1 04+01+09+ 0543+ 0511+ 1 00+02+09+ 1020+ 1170+ 01+02+09+ 1020+ 1170+	1 01+10+10+ 0459- 0441+ 0 03+10+10- 0447- 0529- 1 00+11+10+ 0435- 0455-	1 103+02+13+ 0355+ 0382+ 1 03+02+13+ 0414+ 0493+ 0 03+02+13+ 0414+ 0570- 0 04+02+13+ 0297+ 0570-
	1 01-01-00- 1011- 1193- 1 1 02-01-00- 0792- 0908- 1 1 03-01-00- 0817- 0905- 0 9 04-01-00- 0817- 0905- 0	1 00+04+01+ 0348+ 0743+ 1 00+04+01+ 0575+ 0549+ 0 02+08+01+ 0618+ 0549+ 0 03+08+01+ 0450+ 1474+	1 04+01+03- 1044- 1043- 1 04+01-03- 0521- 0589- 1 00+02+03+ 0520- 0433- 1 00+02-03+ 1717- 1923-	1 01+05-04- (1587) 0/32+ 1 02+05-04+ 1613+ 0/18+ 0 02+05-04+ 1174- 1239- 1 02+05+04+ 1301- 1275+	1 02+11+05- (13/4+ 0442+ 1 02+11+05- (13/4+ 0442+ 1 00+12-05+ (12/5+ 0348+ 1 00+0(1+06+ (15)/5+ 0348+	0 04+05-07- 0360- 0371- 1 00+05-07+ 0875+ 0678+ 1 01+05+07- 0628+ 0450+	1 01+02+09+ 10k1+ 04451 1 02+02+09+ 10k1+ 09851 1 02+02+09+ 0361+ 02451	1 02+01+11+ 051- 0326+ 1 04+01+11+ 055- 0593+ 1 05+01+11+ 0554- 0560+	1 04+02+13+ 0414+ 0448- 0 04+02+13- 0672+ 0540- 1 04+03+13+ 0551+ 0489+
	1 00-02-00+ 0514. 0446+ 0 1 01-02-00+ 1445. 1445+ 1 1 01-02-00+ 1552- 1752+	0 04+0H+01+ 0444+ 0485+ 1 01+01+01+ 0446+ 0487+ 1 03+01+01+ 0322+ 0314+	1 01+02+03+ 0221+ 0292+ 1 01+07+03+ 1425+ 1520+ 0 01+02+03+ 2273+ 2208+	1 02+05+04- 05/2+ 05/5+ 1 03+05+04+ 07/2+ 05/5+ 1 03+05+04+ 0844+ 1022-	1 01+00+46+ 0588+ 0502+ 1 01+00+66= 0459+ 0219+ 1 03+00+96+ 0413+ 0234+	0 02+04+07+ 0426+ 0386+ 1 02+04-07+ 07#2+ 0655+ 1 02+04-07- 08#1+ 0662+	1 02+07+09+ 0857. 0899+ 1 03+02+09+ 0840+ 0515+ 1 03+02+09+ 0430+ 0341+	1 01+01+11+ 0598- 0549+ 1 01+01-11+ 04/6+ 6693+ 1 01+01+11+ 0481- 0515+	1 00+03-13+ 0747- 0768- 1 01+03+13+ 0300+ 0347+ 1 01+03-13+ 0626- 0610-
	1 02+02+00+ 0895+ 0884+ 1 02+02+00+ 1245+ 1265+ 1 03+02+00+ 0534+ 0423+	1 00+10-01+ 0540- 0515- 1 01+10-01- 0448- 0498- 1 02+10-01- 0419- 7429-	1 02+02+03+ 1147- 1355- 0 02+02+03- 1358- 1421- 1 03+02-03+ 1254- 1404-	1 03+04+04+ 0947+ 1092+ 1 03+05+04+ 0935+ 1007+ 1 04+05+04+ 0813+ 1005+	1 03+00+06- 0351+ 0346+ 1 04+00+06- (1370+ 0361+ 1 00+01+06+ 0809+ 0850-	1 03+04+07+ 0772+ 0593+ 1 04+06+07+ 0492+ 0494+ 1 04+06+07+ 0497+ 0440+	1 03+02+09+ 0819+ 0865+ 1 04+02+09+ 0819+ 0843+ 1 04+02+09- 0736+ 0702+	1 02+01+11+ 0572- 03;C+ 1 02+01+11- 0504- 0467+ 1 03+01+11+ 0518- 0402+	1 01+01+13+ 0354+ 0454+ 1 02+03+13+ 0544+ 0495+ 1 02+03+13+ 0514+ 0495+
	1 03+02+00+ 1104+ 1246+ 0 1 04+02+00+ 0798+ 0758+ 0 00+03+00+ 0044+ 0083+	0 04+10+01+ 0181+ 0214 1 00+11+01+ 0334+ 0347+ 1 00+11+01+ 0377- 0542- 1 01+11+01+ 0465- 6455-	1 03+02+03+ 0416+ 0358+ 1 04+02+03+ 0416+ 0851-	1 04-03-04- 0683- 1003- 1 00-08-04- 0683- 0603- 1 00-08-04- 10683- 1456-	1 01+01+05+ 0500 + 5544- 0 01+01+05- 5929+ 5544- 0 01+01+05- 5929+ 5924-	1 00.07.07.0 0448.0345. 1 00.07-07. 0825. 0715. 1 01.07.07. 0713. 0588.	1 01+03+09+ 0736+ 1694+ 0 01+03+08- 0619+ 0627+ 1 01+03-08- 0870, 1682+	1 00+02+11+ 6820- 0361+ 1 01+02+11+ 0817- 0483+ 1 01+02-11+ 0478, 0514+	0 02+03+13- 0654- 0760- 1 02+03-13- 0386. 0477. 0 03-03-13- 0426. 0590.
	1 01+03+00+ 097++ 1077+ 1 02+03-00+ 0774+ 0712+ 1 02+03-00+ 0734+ 0712+	0 02+11+01= 0445= 0595- 1 00+12+01+ 0344+ 0402+ 1 00+12+01+ 0528= 0579=	1 04+02+03+ 0415+ 0417+ 1 00+02+02+ 0471+ 0390+ 1 00+02+02+ 0471+ 0390+	1 01+08+04- 1360- 1232- 1 01+06-04- 06-1+ 0542+ 1 02+04+04+ 0382+ 0304+	1 02+01+06+ 0405+ 0565+ 1 03+01+06+ 0651+ 0601+ 1 03+01+06+ 0320+ 0541+	1 01+07+07+ 0654+ 0615+ 1 02+07+07+ 0538+ 0533+ 1 02+07+07+ 0907+ 0926+	1 02+01+09+ 08/1, 099++ 1 02+01+09+ 08/1, 08/9+ 1 03+01+09+ 0712+ 0712+	0 01+02+11= 0404+ 0530+ 1 01+02-11= 0581= 0514+ 1 02+02+11+ 0657= 0361+	1 03+03-13+ 0542- 0672- 0 03+03+13- 05#2- 0666- 1 03+03-13- 0419+ 0502+
	1 03+03+00+ 0467+ 0416+ 1 04+03+00+ 0540+ 0570+ 0 04+03+00+ 6921+ 1020+	0 01+12+01+ 03/3+ 0362+ 0 01+12+01+ 05/7+ 0586- 1 00+00+00+ 1873- 1874-	1 01+01+03+ 0414+ 0459+ 0 01+03+03+ 1245+ 1293- 1 01+03+03+ 1418+ 1854-	1 02+05+04* 0978- 0986- 1 03+06+04* 10:7* 0989* 1 03+06+04* 0616, 0561+	1 03+01+08- 0535- 0557- 1 04+01+08+ 0348- 0399- 1 04+01+08- 0041- 0077-	1 02+07-07- 0476. 0460. 1 03+07+07+ 0464. 0431. 1 03+07+07- 0542. 0815.	1 03+03+09+ 0945, 0851+ 1 04+03+09+ 0588, 0585+ 0 04+03+09+ 0434+ 0454-	1 02+02-11+ 0578+ 0535+ 0 02+02+11+ 0543+ 0532+ 1 02+02+11+ 0543+ 0532+ 1 02+02+11+ 0829+ 0421+	1 04+03-13+ 0492- 0556- 0 04+03+13- 0688- 0615- 1 04+03+13- 0245- 0337+
	1 00+04+00+ 0189- 0212- 1 01+04-00+ 0325+ 0209+ 1 02+04-00+ 0542+ 0470+	1 01+00+02+ 1122- 1625- 1 01+00+02+ 1501- 1604- 1 02+00+02+ 1664- 0577- 1 02+00+02+ 1347- 1440-	1 02+03+03+ 0517+ 0470+ 1 02+03+03+ 1408- 1467- 1 02+03+03+ 1358- 1343-	1 04+06+04+ 6852- 0915+ 1 04+06+04- 1008- 0993- 1 04+08+04- 04+5, 6410+	1 00+02+06+ 1076- 1188- 1 00+02+06+ 1017+ 0074+ 1 01+02+06+ 1016- 0+67- 1 01+02+06+ 1016- 0+67-	1 04+07+07+ 05+0+ 0753+ 1 04+07+07+ 0520+ 0477+ 1 04+07+07+ 0520+ 0477+	1 00+04+09+ 0727+ 0626+ 1 00+04+09+ 0727+ 0626+ 1 00+04+09+ 0727+ 0726+	1 03+02+11+ 0848- 0329- 1 03+02+11+ 0710- 0421+ 1 04+02+11+ 0548- 0351- 0 05-02+11- 0538- 0351-	1 00+04+13+ 0540+ 0542+ 1 00+04+13+ 0544- 0542- 1 01+04+13+ 0544- 0542- 1 01+04-13+ 0544- 0543-
	1 03-04-00- 0547, 04267 1 03-04-00- 0144, 0050+ 0 04-04-00- 0459, 0320+ 0 10-04-00- 0459, 0320+	1 03+00+02+ 05/4+ 0800- 1 03+00+02+ 0849- 0880- 1 04+00+02+ 0742- 0918-	1 03+01+03+ 0777 0593+ 1 03+03+03+ 1543+ 1579+ 1 03+03+03+ 1441- 1532-	1 01+07+04- 074P- 0586- 1 02+07+04- 0510- 0551- 1 03+07+04- 0560- 0551-	0 01+02+44* 0841+ 1106+ 1 01+02-48* 1176= 1075= 1 02+02+46+ 077/* 0474=	1 00+04-07+ 0702+ 0591+ 1 01+04+07+ 0804+ 0705+ 1 01+08+07+ 0849+ 0835+	1 01+04-09+ 0705+ 0618+ 0 01+04+09+ 0618+ 0453+ 1 01+04+09+ 0888+ 0982+	1 04+02+11= 0457= 0258+ 1 00+01+11+ 0957= 0332+ 1 01+01+11+ 0841= 0332+	1 01+04+13= 0622= 0647- 1 01+04+13= 0521+ 0631+ 0 02+04+13= 0518+ 0632+
	1 01+05+00+ 0518- 0433- 1 02+05+00+ 0412- 0295- 1 03+05+00+ 0445+ 0362+	1 04+00+02* 0607- 0968- 0 00+01+02* 0964- 1198- 1 00+01+02* 0968- 1011-	1 03-07-03-0394+ 0474+ 1 04-07-03+ 0448+ 0426+ 1 04-03-03+ 0977- 1170-	\$ 04+07+04= 054A= 0581= \$ 04+07=04= 0421+ 0411+ \$ 00+04=44* 0705= 0857=	1 02+07+06+ 0374+ 0204+ 1 02+07+06+ 0671+ 0616- 1 03+07+06+ 0657+ 0616-	1 01+05-07- 0440, 0487+ 1 02+04+07+ 0517+ 0574+ 1 02+04+07- 0805; 0784+ 1 02+04+07- 0805; 0584+	1 02+04+09+ 0775, 0724+ 1 02+04+09+ 0842+ 0890+ 1 02+04+09+ 0522+ 0488+ 1 02+04+09+ 0522+ 0488+	1 01+03-114 0405. 0407. 0 01+03-11- 08404 0710. 1 01+03-11- 0930- 0740-	1 02+04+13+ 0735- 0774- 1 02+04+13- 0555- 0565- 1 02+04+13- 0555- 0502+
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	0 04+04-00 0455- 0356- 1 00+07+00+ 1074- 0856- 1 01+07+00+ 08/0- 0711-	1 02+01+02+ 0840+ 0828+ 1 03+01+02+ 1108+ 1307+ 1 03+01+02+ 0858+ 0560- 1 03+01+02+ 0258+ 052-	1 02+04+03+ 1162+ 0963+ 1 02+04+03+ 0962+ 1018- 1 02+04+03+ 1455+ 1365- 02+04+03+ 1655+ 0561+	1 00+10+04+ 0525+ 0502- 1 01+10+04+ 0572- 0459- 0 01+10-04+ 0525- 0526- 0 02+10-04+ 0500- 0504-	1 00+03+06+ 0446, 1090+ 1 01+01+06+ 1128+ 1(33+ 1 01+03+06+ 1200+ 1261- 0 01+03+06+ 1200+ 1261-	1 01+09+07+ 0846+ 0362+ 1 01+09+07+ 0846+ 0364+ 0 01+09+07+ 0653+ 0525+ 1 02+09+07+ 0653+ 0585+	1 01+05+09+ 0544 0575+ 1 01+05+09+ 0576+ 0508+ 1 01+05+09+ 0576+ 0545+	1 04+03-11+ 0520+ 0536+ 0 04+03+11+ 0758+ 0595+ 1 04+03+11+ 0511+ 0514+	0 01+05+13+ 0504+ 0559+ 1 01+05+13- 0551- 0524- 1 01+05+13- 0704+ 0580+
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	0 02+04-40+ 0747+ 0839+ 1 03+08+00+ 0841+ 08784 8 03+08+00+ 0784+ 0957-	1 01+02-02- 1301- 1450- 1 02+02+02+ 1408- 1518- 1 02+02-02+ 0729- 0861-	1 01+05+03+ 1040+ 0974+ 0 01+05+03+ 0805- 0923- 1 01+05+03- 0558- 0528-	1 03+00+06+ 1158+ 1293+ 1 03+00+05= 0844+ 0977+ 1 04+00+05+ 1248+ 1294+	1 04+03+06+ 0748+ 0822+ 0 04+03+08= 0748+ 0547+ 1 04+03+08= 0821= 0875=	0 04+10-07- 0577+ 0585+ 0 02+11+07- 0248+ 0286+ 1 00+00+08+ 03#6- 0366-	1 00+05-09+ 0972- 0852- 1 01+0A+09- 0958- 0822- 1 02+0A+09- 0858- 0725-	1 03+04+11+ 0448- 0550- 1 03+04+11+ 0581+ 0593+ 1 03+04+11+ 0981+ 0920+	1 02+06+13- 0317- 0512- 1 02+06+13- 0432- 0512- 1 02+06-13- 0475- 0458- 1 02+06-13- 0475- 0458-
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	1 03+09+00+ 0814- 08847 6 03+09-00+ 0814- 0775- 1 64+09+00+ 0878- 0840-	1 04+02-02+ 0664- 0875- 1 04+02+02-0283+ 0216+ 0 04+02-02=037+ 1110-	1 04+05+03+ 0911- 1112- 1 04+05-03- 0550: 0922+ 1 00+06+03+ 1057+ 0948-	1 02+01+05+ 0574+ 0739+ 1 02+01+05+ 1358+ 1512+ 0 02+01+05+ 1351+ 1714+	1 02+04+06+ 1047+ 1127+ 1 02+04+0E= 1240+ 1152+ 1 02+04-06= 1123= 1161=	1 01+01+08+ 0551- 0595- 0 01+01+08- 0731+ 0893- 1 02+01+08+ 0640- 0559-	1 01+07+09+ 0750- 0782- 1 02+07+09- 0778- 0758- 1 03+07-09+ 0827- 0845-	\$ 01+05+11+ 0882- 0890- \$ 01+05+11+ 0877+ 0857+ \$ 01+05+11+ 0840+ 0732+ \$ 01+05+11+ 0840+ 0732+	1 01+00+14+ 0768. 0440+ 1 01+00+14+ 0483. 0779- 1 01+00+14+ 0583. 0582- 1 02+00+14+ 0483. 0411-
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	0 02+10+00+ 0581+ 0/47+ 0 02+10+00+ 0381+ 0/47+ 1 00+11+00+ 0459+ 0453+	1 01+03=02= 0968= 1467= 1 02+01+02= 0821= 0865= 1 02+01+02= 0821= 0865=	1 03+06+03+ 0711+ 0770+ 1 03+0#+03+ 0847+ 0848+ 1 04+06+03+ 0720+ 0741+	1 03+01+05+ 1247+ 1385+ 1 04+01+05+ 0644+ 0711+ 1 04+01+05+ 1214+ 1328+	04-04-06-0945-1045- 04-04-06-0801-0455- 104-04-05-0782-0782-	0 01+02+08- 1020- 1012+ 1 02+02+08+ 0547+ 0600+ 1 02+02+08+ 0841- 0073+	0 01+08+08+ 0421- 0444- 1 01+08+08- 0790- 0748- 1 02+08+09- 0475- 0486-	1 02+05+11+ 0567+ 0668+ 1 03+05+11+ 0817+ 0881+ 1 03+05+11= 0898+ 0929+	1 04+00+14+ C3A6+ 0487- 1 04+00+14+ C407+ 0448- 1 00+01+14+ 055A+ C683-
	1 01+11+00+ 03+0+ 0419+ 0 03+11+00+ 0420+ 0440+ 0 02+00+01+ 01+0+ 0205+	1 02+03-02- 0741- 0908- 1 03+03+02+ 0514- 0581- 1 03+03-02+ 0301- 0289-	1 04+06+05+ 1198+ 1154+ 1 00+07+03+ 0773+ 0732+ 1 01+07+03+ 1335+ 1138+	1 04+01+05+ 1080+ 1186+ 1 04+01+05+ 08#1+ 0845+ 5 00+02+05+ 0640+ 0585+	1 00+05+06+ 10+5+ 0785- 1 00+05-06+ 1077+ 1005+ 1 01+05+06+ 1101- 0910-	0 03+02+08- 0914- 10/3* 1 03+02+08+ 0848- 0845* 0 03+02+08+ 0918- 0994+ 1 04+02+08+ 0755- 0818-	1 03+08+09+ 0455- 0508- 0 04+08+09+ 0576- 0565- 1 04+08+09+ 0478- 0484-	1 04+05+11+ 0573+ 0414- 1 04+05+11+ 0543+ 0599+ 1 04+05+11+ 0578+ 0578+ 04+05+11+ 0578+ 0578+	1 01+01+14+ 0511+ 0499- 1 01+01+14+ 051+ 0465+ 0 01+01+14+ 051+ 0465+
	0 02+04+01+ 0952+ 0942+ 1 04+05+01+ 0231+ 0207+ 0 00+51+01+ 0318+ 3282+ 1 00+51+01+ 0318+ 3282+	1 04+03+02+ 04+6+ 024- 1 04+03+02+ 03+6+ 025+ 1 04+03+02+ 0325+ 0237+ 04+03+02+ 025+ 0237+	1 01+07+03- 0714+ (1824+ 1 02+07+03- 0714+ (1824+ 1 02+07+03+ 0914+ 0818+ 1 02+07+03- 0818+ 0905+	1 01+02+05+ 00+3+ 0945+ 1 01+02+05+ 0718+ 0752+ 0 01+02+05+ 1456+ 1423+	1 01+05+05-0771-0720- 1 02+05+05+05-0771-0720- 1 02+05+05+05+0550-0850-0850-	0 04+02+08- 0582- 0734- 1 00+03+08+ 0744+ 0638+ 1 00+01+08+ 0943- 1030-	1 00+09-09+ 0678- 0605- 1 01+09-09- 0454- 0498- 1 02+09+09- 0454- 0498- 1 02+09+09- 0398- 0349-	1 00+05+11+ 0427- 0440- 1 00+05+11+ 0881+ 0817+ 1 01+05+11+ 0522- 0505-	1 01+01-14- 0717+ 0489- 1 02+01+14+ 0542+ 0557- 1 02+01+14+ 0474+ 0464+
	1 01+01+01+ 0736+ 0556- 1 01+01+01+ 2055+ 2324+ 1 02+01+01+ 0561+ 0557-	1 04+03-02- 1020- 1045- 1 00+04+02+ 1092- 1183+ 1 01+04+02+ 0770- 0733-	1 03+07+03+ 0869+ 0595+ 1 03+07-03- 0758+ 0843+ 1 04+07+03+ 0746+ 0769+	1 01+02+05+ 0745+ 0903+ 1 02+02+05+ 0497+ 0534+ 1 02+02+05+ 1024+ 1197+	1 02+05+06+ 0838+ 0653+ 1 02+05+06+ 0870+ 0924+ 1 03+05+06+ 0984- 0915-	1 D1+03+U8+ C7A2+ C669+ 1 01+03-U8+ 1245- 13A3- 0 01+03+08- 1081- 1054-	0 02+09-09- 042A- 0454- 0 03+09-09- 049A- 042A- 0 04+09-09- 0351- 0332-	1 01+05+11= 08/8+ 0718+ 1 02+05=11+ 0546+ 0535+ 1 02+05=11= 0851+ 0518+ 02+05=11= 0851+ 0518+	1 02+01+14+ 0607+ 0512- 0 03+01+14+ 0428+ 0520- 1 03+01+14+ 0605+ 0470- 0 03+01+14+ 0605+ 0470-
	1 02+01+01+ 0414 0432+ 1 02+01+01+ 0414 0432+ 1 03+01+01+ 03+6- 0362-	0 03+04-02+ 1938+ 0721+ 1 01+04+02- 0849+ 0907+ \$ 01+04-02- 1212- 1322-	1 04+07+03+ 0925+ 1054+ 1 00+03+03+ 0875+ 0403+ 1 01+03+03+ 0875+ 0403+ 1 01+03+03+ 0875+ 0403+	0 02:02:05: 1213: 1318: 1 03:02:05: 05:5: 05:5: 1 03:02:05: 07:3: 05:41:	1 03+05+05+ 0875+ 0684+ 1 03+05+05+ 1075+ 0859+ 1 03+05+06- 0875- 0673-	1 02+03+08+ 0201+ 0538+ 1 02+03+08+ 0201+ 0617+ 1 02+03+08+ 1187- 1170- 0 02+03+08- 0983- 1121-	1 00+10+04+ 0472+ 0388+ 1 00+00+10+ 0754+ 0762+ 0 01+00+10+ 0849+ 0569+ 1 01+00+10+ 1094+ 0984+	1 03+05-11+ 0316- 0386- 1 03+05-11+ 0722+ 0689+ 1 03+05+11- 0610+ 0571+ 1 03+05-11- 0610+ 0571+	1 03+01-14- 0345+ 0599- 1 04+01-14+ 0316+ 0480- 1 04+01-14+ 0521+ 0580-
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	1 00+07+01+ 0457- 0442- 0 00+02+01+ 1214+ 1237+ 1 01+02+01+ 1261- 1128+	1 03+04+024 0543+ 0388+ 1 03+04+02= 0832+ 0594+ 1 03+04+02= 0793= 0863=	1 02+68-03- 0647+ 0815+ 1 03+08+03+ 0615+ 0682+ 1 03+08+03- 0573+ 0451+	1 04+07+05+ 0814+ 0888+ 1 06+07+05+ 1046+ 1047+ 1 01+07+05+ 0697+ 0813+	1 00+05+05+ 0954- 0865- 1 00+05+05+ 1043- 0+14+ 1 01+05+05+ 0821- 0710-	1 04+03+08+ 0545+ 0481+ 1 04+03-08+ 0955- 0910- 0 04+03+08+ 1120- 1131-	1 03+00+10- 0875+ 0915+ 1 04+00+10+ 0584+ 0502+ 1 04+00+10- 0595+ 0727+	1 01+07+11= 0819+ 0731+ 1 02+07+11= 0445+ 0547+ 1 03+07-11+ 0474+ 0440+	0 02+02+14+ 0543+ 0475- 1 02+02+14+ 0574+(0585- 0 03+02+14+ 0587+ 0458- 1 03+02+14+ 0587+ 0458-
	0 01+02-01+ 1843+ 1674+ 0 01+02+01+ 1412+ 1331+ 1 01+02+01+ 02+4+ 0814+	\$ 04-04-02+ 02+ 0828- 0518- \$ 04-04-02+ 0822+ 0505+ \$ 04-04+02- 0542+ 0575+ \$ 04-04-02- 0542+ 0575+	1 03-03-03- 0650+ 0437+ 1 04-03-03+ 0412+ 0493+ 1 04-04-03- 0575+ 0530+ 0 04-08-03- 0541+ 0580+	0 01+04+05+ 11#0+ 0893+ 1 01+03+05+ 1355+ 0391+ 1 02+03+05+ 0517+ 0402+	1 01+06+06+ 1052+ 0874+ 1 01+04-06+ 0741+ 0737+ 1 02+06+08+ 0811- 0538-	1 00+03+08- 049+ 0347+ 1 00+04-08+ 0896+ 0453+ 1 00+04-08+ 0685+ 0641- 4 01+04-08+ 1153+ 0415+	1 00-01-10- 0972- 1203- 1 01-01-10- 0917- 0909- 1 01-01-10- 0917- 0909- 1 01-01-10- 0942- 0756-	1 03-07-11- 0273: 0227- 1 03-07-11- 0414- 0289- 1 04-07-11- 0828- 0571- 5 64-07-11- 0828- 0571-	1 03+02+14+ 0514, 0500- 1 04+02+14+ 0317, 0540- 1 04+02+14+ 0317, 0540- 1 04+02+14+ 0514, 0565-
	0 02+02+01+ 1911+ 1920+ 1 02+02+01+ 1911+ 1920+ 1 02+02+01+ 08+7- 0929+ 1 02+02+01+ 0814- 0812-	1 00+03+02+ 05+5- 6555- 1 00+05-02+ 1020+ 0982+ 0 01+05-02+ 0704, 0812+	1 00+09+03+ 0523+ 1530+ 1 00+09+03+ 0773+ 0525+ 1 01+09+03- 0774+ 0495+	0 U2+0'+05- 0776+ 0795+ 1 03+0'+05+ 0872+ 0999+ 1 03+0'+05+ 0972+ 1003+	1 02+05+06- 0675- 0617- 1 02+05+06+ 0645- 0740- 1 03+05+06+ 0645- 0740- 1 03+05+06+ 1073+ 1227+	1 01+04+08+ 1007- 1191- 0 01+04+08- 1217- 1031- 1 01+04-08- 0843+ NH88+	0 01+01+10= 0849+ 0887+ 1 01+01+10= 0937+ 1018+ 0 02+01+10= 0959+ 0964+	6 62-10-11- 6160- 6163- 0 00-00-12- 0113- 0797-	1 00+03+14+ U5+4+ 0457+ 1 01+03+14+ 0575+ 0356+ 1 01+03+14+ 0458+ 0716-
	1 03.07.01. 0714- 0832* 1 03.02-01. 1073. 1245. 1 03.02-01. 1073. 1245.	1 01+05+02+ 1134+ 1015+ 0 02+05+02+ 0603+ 0683+ 1 02+05+02+ 1223+ 1037+	0 01+09+03- 0577. 0594. 1 02+09+03+ 0486. 0404. 1 02+09+03- 0573. 0593. 0 02+09-03- 0573. 0593.	1 03+03-05- 0553+ 0458+ 1 04+03+05+ 0322- 0254- 1 04+03-05+ 0648+ 0764+	1 03+0A+0A+ 0941+ 0794+ 1 03+0A+06= 0815+ 0728+ 1 04+05+06+ 0474+ 0509+	5 02+04+08+ 0755+ U548+ 1 02+04-08+ 1110- 1057- 0 02+04+01- 0273- 0204- 1 02+04+01- 0245- 0292+	1 02+01+10+ 0518. 08684 1 03+01+10+ 0388. 0240+ 1 03+01+10+ 0773. 0847+ 1 04+01+10+ 0402+ 0391+	1 01+00+12+ 0405- 055- 1 01+00+12+ 0405- 0551- 1 02+00+12+ 0525- 0565- 1 02+00+12+ 0515- 0560-	1 02+01+14+ 0551. 0402+ 1 03+03+14+ 0551. 0402+ 1 03+03+14+ 0551. 0529+
	1 03+02+01+ 0312- 0367- 1 04+03+01+ 0811- 0826- 1 04+02+01+ 0706+ 0/661	1 03+01+07= 0730= 0787+ 1 03+05+02= 0749+ 0787+ 1 03+05=02= 0772= 0799= 1 04+05=02= 0722, 0701	\$ 03+09+03+ 0455+ 0492+ \$ 03+09+03+ 0577+ 0669+ 0 03+09-03+ 0774+ 0710+	0 04+03+05- 0428+ 0284+ 1 L0+04+05+ 0898+ 0715+ 1 01+04+05+ 0186- 0204-	1 04+0A+0A+ 0548+ 0548+ 1 04+0A+0A+ 0548+ 0874+ 1 04+0A+05-0842+ 0890- 1 04+0A+05-0842+ 0898-	1 03+04+08+ 0899+ 0754+ 1 03+04-08+ 0960- 1036- 1 83+04+08- 1881- 0873-	1 04+01-10+ 0643+ 0898+ 1 04+01+10- 0730+ 0683+ 1 04+01+10- 0576+ 0683+	1 03+01+12+ 0747- 0762- 1 03+00+12- 0844- 0864- 1 04+00+12+ 0597- 0512-	0 02+04+14+ 1452+ 1803+ 1 02+01+14+ 0313+ 1813+ 1 00+05+14+ 1562+ 0539+
	1 00-03-01+ 04/4+ 0455- 0 00-03-01+ 153/1+ 1355+ 1 01-03-01+ 140- 145-	1 04+05+02- 0817+ 0811+ 1 04+05-02- 0355- 0300- 1 00+05-02+ 1397+ 1413+	1 04+09+03- 04#0: 0546+ 0 04+09-03- 03/0: 0375+ 1 00+10+03+ 0358: 0375+	1 01+04+05- 0812+ 0736+ 0 01+04-05- 0214- 0140- 02+04-05- 0368+ 0294+	1 00+07+06+ 0642+ 0627+ 1 01+07+06+ 0400+ 0330+ 1 01+07+06+ 0550+ 0619+	1 04+04+08+ 0204+ 0677+ 1 04+04-08+ 0977- 0944- 0 04+04+08+ 0805- 0794-	1 00+02+10+ 1075+ 1103+ 1 01+02+10+ 0800+ 0898+ 0 01+02+10+ 0841+ 0899+ 0 01+02+10+ 0841+ 0899+	1 00+01+12+ 0744+ 0801+ 1 00+01+12+ 0744+ 0731+ 0 01+01+12+ 0744+ 0584+ 1 01+01+12+ 0544+ 07654+	0 02+05+14- 04+2- 0544- 1 03+05+14- 0226+ 0434- 1 04+05-144 0241- 0569-
	0 01+01+01+ 03+01+ 0491+ 1 01+01+01+ 12+0+ 122+ 1 01+04+01+ 1648+ 0840-	0 01+04-02+ 05:14+ 0712+ 1 01+04+02+ 1037+ 090+ 0 02+04-02+ 13:42+ 13:30+ 1 02+04-02+ 13:42+ 13:30+	1 00+10+03+ 075+ 075+ 1 01+10+03- 0971+ 0821+ 1 02+10+03- 0517+ 0552+ 1 03+10+03- 0570+ 0551+	1 03+04+05- 0540+ 0405- 1 03+04+05- 0540- 0405-	1 02+07+1/6+ 04+4+ 0565+ 1 02+07+04+ 0474+ 0407+ 1 03+07+06+ 0534+ 0417+	1 00+05+06+ 1109+ 1142+ 1 00+05+08+ 1109+ 1142+ 1 00+05+08+ 0651- 0583- 1 01+05+08+ 1157+ 0961+	1 02+02+10+ 0817+ 0792+ 0 02+02+10+ 0948+ 0964+ 1 02+02+10+ 0542+ 0520+	0 01+01+12= 0845= 0727= 1 01+01=12= 0916= 0860= 1 02+01+12= 0725= 0682=	1 00+04-141 06/14- 0411+ 01+05+14- 0451- 0455- 1 02+06+14- 0458- 0430-
	1 02+01+01+ 1010+ 1030+ 0 02+01+01+ 1503+ 1/41+ 1 02+01+01+ 1306+ 1543+	03+04-02+ 0848+ 0985+ 03+04+02+ 0926+ 0936+ 04+08+02+ 0728+ 0728+	0 03+10=03= 0244. 0301+ 1 00+11=03+ 0741+ 0794+ 1 01+21+03= 05+3+ 0543+	1 04+04+05+ 0212+ 0283- 1 04+04+05+ 0624+ 0624+ 1 00+05+05+ 0508+ 0443-	1 04+07+08+ 0546+ 0423+ 1 04+07+08+ 0546+ 0423+ 1 04+07+05= 0570- 0552- 1 02+04-06- 0410+ 0420+	1 01+05-08+ 0727- 0847- 1 01+05-08- 0904- 0704- 1 01+05-08- 0664, 0722+	1 03+02+10+ 0578+ 0977+ 0 03+02+10+ 0575+ 0828+ 1 04+02+10+ (313+ 0289+	1 02+01+12+ 0708+ 0639- 0 02+01+12- 0808- 0648+ 1 02-01-12- 0808- 0777-	0 03+05+14= 03+5= 0407= 1 04+05+14+ 03+0= 0318= 0 04+05+14= 06+9= 0612= 1 00+05+14= 06+9= 0612=
	1 03+03+01+ 1249- 1056- 1 03+03+01+ 1417+ 1688+ 1 03+03+01+ 1544+ 1684+	1 00+07+02+ 1544+ 1504+ 1 00+07+02+ 1118+ 1044+ 1 01+02+02+ 0741+ 0565+	1 02+11+03- 0556+ 0670+ 1 00+12-03+ 0508+ 0564+ 0 01+12+03- 0466+ 0478+	0 00+05+05+ 030#+ 0368+ 1 01+05+05+ 04##+ 0410- 1 01+05+05+ 04##+ 0410-	0 04+04+06+ 024F+ 020F+ 0 01+10+05+ 0307+ 0272+ 0 01+10-06+ 0305+ 0298+	1 02+05+08+ 022+- 0992- 1 02+05+08- 0644- 0601- 1 02+05+08- 0644- 0601- 1 02+05+08- 1024- 0996+	1 04+02+10+ 0707+ 0763+ 0 04+02+10+ 0600+ 0585+ 1 04+02+10+ 0247+ 0303+ 1 00+02+10+ 0247+ 0303+	1 03-01-12- 062- 0623- 1 03-01-12- 062- 064- 1 03-01-12- 062- 054- 1 04-01-12+ 0541- 0624-	1 01+07+14= 0526- 0546- 0 02+01+14= 0358= 0445- 0 03+07+14= 0358= 0445-
	1 03+03+01+ 0504- 0550- 1 04+03+01+ 0582- 0557- 1 04+03+01+ 1217+ 1412+	1 02+07+02+ 0474+ 0437+ 0 02+07+02+ 0474+ 0437+ 0 02+07+02+ 1130+ 1258+ 1 02+07+02+ 0462+ 0+06+	0 01+00+04+ 1067+ 1005+ 0 01+00+04+ 0602+ 0598+ 02+00+04+ 0141+ 0161+	1 03+05+15+ 0742= 0649= 1 03+05+05= 0603= 0509= 1 04+05+05* 0441= 0379=	1 01-11-05- 1377- 0402- 0 02-11-01- 03+1- 0418- 1 00-12-06- 0341- 0472-	1 03+05+08+ 06+7+ 0729+ 1 03+05+08+ 0815- 0795- 1 03+05+08+ 0851- 0625-	0 01+03-10+ 0849+ 0410+ 0 01+07+10- 0849+ 0564+ 1 02+03-10+ 0811+ 0855+	1 04+01+12+ 0820- 0641- 1 04+01+12- 0717- 0884- 1 04+01-12- 0857- 0842-	1 00+08-14+ 0540- 0557- 0 01+03+14- 0440- 0536+ 0 04+04+14- 0522- 0463+
	1 04+01+01+ 0617+ 0564+ 1 00+04+01+ 1444+ 1650+ 1 00+04+01+ 1345+ 1345+	1 03+07+02+ 0847+ 0573+ 1 03+07+02+ 0925+ 1170+ 1 03+07+02+ 0913+ 1082+	1 03+00+04+ 0518+ 0877+ 1 03+00+04- 0513+ 0590+ 1 04+00+04+ 0615+ 0541+	1 04+05-05+ 0451+ 0425+ 1 04+05-05+ 0405- 0/59- 1 00+05+05+ 0471- 0587-	1 00+01+07+ 0821+ 0975- 1 01+07+07+ 1334- 1125- 1 01+07+07+ 1334- 1125-	1 03-05-08- 0791. 0788. 1 04-05-08- 0578. 0655. 1 04-05-08- 077- 0701- 1 04-05-08- 0537. 0570.	0 02+03+10- 0954+ 0905+ 1 03+03+10- 0893+ 0652+ 0 03+03+10- 0843+ 0535+	1 00+02+12+ 0564- 0544- 0 00+02+12+ 0565+ 0615+ 1 01+02+12+ 0446+ 0419- 1 01+02+12+ 0446+ 0465-	0 01+09+14- 05+0+ 0413- 4 00+00+15+ 0553+ 0492+ 1 01+00+15+ 0303+ 0408+
	1 01+04+01+ 1511- 1451- 0 01+04+01+ 0644+ 0710+ 1 01+01+01+ 1440+ 1523+	1 04-02-02+ 0371+ 0377+ 1 04-02+02+ 0711+ 0762+ 1 00-04+02+ 0843+ 0705+ 2 00-04+02+ 0843+ 0705+	1 00+01+04+ 09551 1057 1 00+01+04+ 0341+ 0193+ 1 01+01+04+ 05+7+ 0768+ 01+01+04+ 05-74+ 0455+	1 01+06+05+ 0815- 0598- 1 01+06+05+ 0815- 0598- 1 01+06+05+ 0641- 0846- 1 02+05+05+ 1048- 0923-	1 02+00+07+ 1132- 1201- 1 02+00+07- 1344- 1452- 1 03+00+07+ 0814- 0800-	1 04+05-08-0509+ 0475+ 1 08+08+08+ 1051+ 1026+ 1 08+08+08+ 0375+ 0465+	0 04+03+10= 0717- 0783+ 1 00+04+10+ 0444. 0432+ 1 01+04+10+ 0512= 0418=	0 01+02+12= 0640= 0626= 1 01+02=12= 0575= 0537= 1 02+02+12= 0543= 0476=	1 02+00+15- 0473+ 0454+ 0 03+01+15* 0495+ 0414+ 1 03+00+15* 0495+ 0533+
	1 01+04-01- 1077- 1173- 1 02+04-01+ 1518- 1498- 0 02+04-01+ 1548- 09214 1 02+04-01+ 1448	1 01+0++U2+ 7543+ U482+ 0 01+05+U2+ 1085+ 1084+ 1 01+05+02+ 7547+ U586+	1 01+01-04+ 1141+ 1374+ 1 02+01+04+ 0905+ 1113+ 1 02+01+04+ 0243+ 0257+	1 02+05+05= 0617= 0550- 0 02+05= 05= 0600= 0584= 1 03+05+05+ 0715= 0715=	1 04+00+07+ 1028- 1020- 1 04+00+07+ 0754- 08+2- 1 00+01+07+ 1244- 1529-	1 01+04+0#+ 0713+ 0758+ 1 01+08-08- 0842+ 0918+ 1 02+08+08+ 0874+ 0871+	1 01+04+10+ 0748+ 0857+ 0 01+04+10+ 0545+ 0512+ 1 01+04+10+ 0544+ 0461-	1 02+02-12+ 0412+ 0465- 0 02+02+12+ 0735+ 0534+ 1 02+02+12+ 0773+ 0741+	1 04+00+15- 0215, 0203+ 1 00+01+15+ 0408, 0465+ 1 01+01+15+ 0627, 0646+ 0 01+01+15+ 0627, 0646+
	02+04+01* 0809- 0973- 1 03+04+014 1125- 0931- 1 03+04-014 1381+ 1485+	0 01+03+02- 05/1+ 0843+ 1 02+04+02+ 0818+ 0685+ 1 02+04+02+ 1047+ 1035+	1 02+01-04- (1724, 0739+ 1 03+01+04+ 0511+ 1055+ 1 03+01-04+ 1055+ 0503+	1 03+05+05= 6640- 0564- 1 03+04+05= 6610= 0564- 1 04+06+45+ 4440= 0529- 1 04+06+45+ 0467- 0529-	1 00+01+07+ 0955- 1955* 1 01+01+07+ 0970- 1071- 1 01+01-07+ 0874- 0877-	1 02+08-08-08-08-0 1 02+08-08-08-08-08-08-08-08-08-08-08-08-08-0	1 02+04+10+ 0301+ 0317+ 1 02+04+10+ 0507+ 0519+ 0 02+04+10+ 0511+ 0447+ 1 02+04+10+ 0479+ 0384+	1 03+02+12+ 0548- 0579- 1 03+02+12- 0509+ 0536- 1 03+02+12- 0509+ 0545-	1 02+01-15+ 0475+ 0512+ 0 02+01+15+ 0476+ 0507+ 0 03+01+15+ 03#0+ 0427+
	1 03+04+01- 13(1+ 1191+ 1 03+04-01- 1300- 1200- 1 04+04+01+ 0218- 0525-	1 03+04+02- 0746+ 0736+ 1 03+04-02- 0643+ 0635+ 1 03+04-02- 0643+ 0635+ 1 04+08+02- 0647+ 0675+	0 04+01-04+ 0543+ 0425+ 1 04+01+04+ 1166+ 0182+ 1 04+01+04+ 1166+ 0182+	1 00+07+08+ 0804+ 0708- 1 00+07+08+ 0840+ 0808- 1 01+07+05+ 1051+ 1021-	0 03+01+07- 1318- 1417- 1 03+01+07- 1325- 1318- 1 02+01+07+ 0767- 0867- 1 02+01+07+ 087- 0865-	1 03+06+08- 0445- 1431- 1 03+08-08- 0818+ 0405+ 1 04+06-08+ 0494- 0477-	1 03+04-10+ USAR+ 0545+ 1 03+04+10- 0527+ 0535+ 1 04+04+10+ 0454- 0446-	1 04+02+12+ 0448- 0495- 0 04+02-12+ 0572+ 0576- 1 04+02-12+ 0490- 0454-	0 03+01+15= 0441, 0525+ 1 03+01=15= 0405, 0377+ 0 04+01=15= 0331, 0404+
	1 04+04+01+ 0852+ 0811+ 1 04+04+01+ 0703+ 0685+ 1 00+04+01+ 1517+ 1550+	0 04+08-02- 0360+ 0408+ 1 00+09-02+ 0614+ 0615+ 1 01+04+02+ 0653+ 0617+	1 00.02-04- 1495- 1772- 1 00-02-04- 0470- 0429- 1 01-02-04- 1928- 1547-	1 01+0/+05* 0648* 0542* 0 01+07+05* 0610* 0878* 1 02+07+75* 1020* 0738*	1 02+01+07+ 1108+ 1951- 1 03+01+07+ 0978+ 1014- 1 03+01-07+ 0847+ 1071-	1 04+08-08- 0844, 1823+ 1 00+07+08+ 0813+ 0589+ 1 01+07+08+ 0407+ 0521+ 1 01+07-08+ 0243+ 0921+	1 04+64-10* 0744+ 0733+ 0 04+64+10- 0643+ 0527+ 1 00+05+10* 0547- 0551- 1 01+05+10* 0713+ 0593-	1 00+01+12+ 0447- 0341- 1 01+01+12+ 0447- 0360- 1 01+03+12+ 0447- 0360- 0 01+03+12+ 0447- 0369-	1 04+01+15- 0240+ 0242+ 1 04+01+15- 0240+ 0262+ 1 00+02+15+ 0447+ 0468+ 5 01+02+15+ 0647+ 0681+
	1 00+05-01+ 0772+ 0841+ 1 61+05+01+ 1103- 1481+ 6 01+05-41+ 1243+ 140+	0 01+09-02+ 0544. 07764 1 03+04-02+ 0842+ 0784+ 0 01+04-02+ 0847+ 0784+ 0 01-04-02+ 0847+ 0751+	0 01+02+04+ 1402+ 0588 1 02+02+04+ 1455+ 1556+ 1 02+02+04+ 1455+ 1556+ 1 02+02+04+ 1455+ 1556+	1 02+07+05- 0545- 0524- 1 02+07+05+ 0545- 0524- 1 03+07+05+ 0545- 0598- 1 03+07+05+ 0545- 0598-	1 03+01+07- 1078- 1205- 1 04+01+07+ 0734- 0599- 1 04+01+07+ 0984- 0941-	1 02+07+08+ 0558+ 0585+ 1 02+07-08- 0575+ 0538+ 0 03+07+08+ 0447+ 0509+	1 01-65-10- 0445- 0439- 1 02-95-10- 0512- 0452- 1 02-05-10- 0573- 0490-	1 01+01+12+ 03AB+ 0331+ 1 02+01+12+ 03A7+ 0350+ 0 02+03+12+ 0475+ 0363+	0 01+02+15- 0534+ 0567+ 1 02+02+15+ 0679+ 0704+ 0 02+02+15- 0502+ 0643+
	1 01+05+01+ 121A+ 131A- 1 02+05+01+ 113A- 133A- 0 02+05+01+ 113A- 113A-	1 02+03+02+ 0709+ 0575+ 0 02+04-02+ 0575+ 0689+ 1 03+09+02+ 0565+ 0511+	1 03-02-04- 1001- 0991- 0 03-02-04- 0755- 0710- 1 03-02-04- 0832- 1006-	1 03+07+05- 0755- 0710+ 1 04+07+05+ 05+2- 0614- 1 04+07-05+ 04+4- 0361-	1 04+01+07+ 0505+ 0703+ 1 04+01+07+ 0505+ 0703+ 1 00+02+07+ 1105+ 1122+ 5 00+02+07+ 0655+ 0870+	1 03+07+08- 0778, 0784+ 1 04+07-08- 0598, 0584+ 1 01+04-08- 0445, 0454+	1 02+05-10- 0526- 048- 0 03+05+10+ 0487- 0541+ 1 03+05-10+ 0358+ 0358+	1 02+03-12- 0298- 0371- 1 04+03-12+ 0212- 0345- 1 04+03+12+ 0353- 0293+	0 03+02+15+ 0383+ 0523+ 0 03+02+15+ 0457+ 0462+ 0 04+02+15+ 0391+ 0498+
	1 02+05+01= 1311+ 1224+ 1 02+05+01= 0885- 10/2= 1 03+05+01+ 1116- 1042=	0 03+09-02+ 0573+ 0594+ 1 04+09+02+ 0575+ 0514+ 1 04+09+02+ 0575+ 0514+ 1 04+09+02+ 0443+ 0458+	1 04+02+04+ 08+1+ U326+ 1 04+02+04+ 0848+ 1062+ 1 00+03+04+ 1504+ 1726+ 2 00+03+04+ 1504+ 1726+	1 04+07+05+ 0414+ 0508+ 1 04+07+05+ 0644+ 0725+ 1 00+08+05+ 0970+ 0943-	1 01+02+07+ 1018- 0981+ 1 01+02+07+ 0585+ 0714+ 0 01+02+07+ 0585+ 0707+	0 02+09+08+ 05/0. 0438+ 1 02+08-08- 05/1. 048+ 1 03+08-08- 0443. 0505+	1 04+05+10+ 0480- 0438- 0 04+05-10+ 0353+ 0574+ 0 04+05-10- 0361- 0727-	1 00+04+12+ 0507+ 0476+ 1 01+04+12- 0400+ 038+ 1 01+04+12- 0455+ 0278+	1 00+03-15+ 0515+ 0522+ 1 01+03-15+ 054+ 0505+ 1 01+03+15+ 0530+ 0558+
• • • • • • • • • • • • • • • • • • •	0 03+05-01. 1181. 1110. 1 03+05-01. 1034. 0639. 1 03+05-01. 1034. 1121.	1 00+10+02+ 05+3+ 0597+ 1 01+10+02+ 0544+ 0622+ 1 01+10+02- 0492+ 0539+	1 01+01+04+ 1504+ 1563+ 1 01+07+04+ 0543- 0555- 0 01+07+04+ 0519+ 0599-	1 01+04+05+ 0941- 0989+ 1 01+04+05+ 0947- 0904- 1 01+04-05- 0675- 0605-	1 02+02+07+ 0415+ 0685+ 1 02+02+07+ 0412+ 0890+ 1 02+02+07+ 0412+ 0890+	D 04+08+08+ 0471+ 0619+ 1 00+09-08+ 0482+ 0432+ 1 03+04+08- 0474+ 0525+	1 00.05.10+ 0854- 0546- 1 01.05-10+ 0545- 0458- 1 01.05-10+ 0544- 0502- 03.05-10+ 0544- 0502-	1 00+03-12+ 0507+ 0526+ 1 01+07+12- 0560+ 0573+ 0 01+07+12- 0560+ 0483+	1 02+03-15+ 0873+ 0698+ 1 02+03-15- 04#5- 0540+ 0 03-03-15+ 04#0- 0594+
1 0000000 10400 0010 0010 0010 0010 001	1 04+05+03+ 0917+ 1075+ 1 04+05+01+ 0744+ 0751+ 1 04+05+01+ 1014+ 1042+	0 01+10-02- 0638+ 0776+ 1 02+10+02+ 0550+ 0628+ 0 02+10-02- 0690+ 0827+	01+03+04+ 1317+ 1525+ 02+03+04+ 114A+ 1210+ 02+03-04+ 0379- 0318-	1 02+08+04+ 05+3- 0652+ 1 02+08+05+ 0870+ 0767- 1 02+08+05+ 0796+ 0776+	1 03+02+07+ 0972- 0834- 1 03+02+07+ 0870+ 0940- 0 03+02+07+ 0541- 0520-	1 00+10-08+ 0573+ 0583+ 1 00+10-08+ 0573+ 0583+ 1 01+10+08- 0474+ 0454+ 1 02+10+08- 0574+ 058+	1 02+05-10- 0521- 0597+ 1 03+05-10+ 03A1- 0384- 1 00+07+10+ 0720- 0509+	0 02+07+12+ 0420+ 0401+. 1 02+07+12= 0352+ 0396+ 1 03+07+12= 0473+ 0544+ 03+07+12= 0473+ 0544+	1 04+03-15+ 0447, 0533+ 1 04+03+15- 0462, 0545+ 1 00-04-15+ 0570, 0553+
	1 00+0A+U1+ 0454- 0830- 1 00+0A+U1+ 0610+ 0526+ 1 01+0A+U1+ 1164- 1128-	0 03+10-02+ 04/7+ 0204+ 1 03+10-02- 04/7+ 0204+ 1 04+10-02- 0312+ 0348+	02+03+04- 13/0- 1262+ 02+03+04- 13/0- 1262+ 03+03+04+ 0674- 0803+ 1 03+03+04+ 0472- 0437-	1 03-08-05- 0565- 0664- 1 03-08-05- 0720- 0720-	1 03+02+07- 0992- 0943- 1 04+02+07+ 0558- 6527- 1 04+02-07+ 0751- 2749-	C 03+10+08= 6511+ 6516+ 1 00+11-08+ 0544+ 6586+ 1 01+11+08= 0747+ 0682+	1 01+07+10+ 0444+ 0344- 1 01+07+10= 0444- 0549+ 1 01+07+10= 0571= 0457-	1 04+07+12+ 0328+ 0299+ 1 04+07+12- 0340+ 0472+ 1 00+08-12+ 0905+ 0866+	1 01+04+15- 0481: 0479+ 1 02+04-15+ 0485- 0505+ 1 02+04+15- 0387- 0429+
Operation Operation <t< td=""><td>1 01+06+01= 0948+ 0873+ 1 01+06+01= 0978+ 0873+ 1 01+06+01= 0978+ 1245- 1 02+06+01= 1171+ 1145*</td><td>1 01+11+42+ 0528+ 0611+ 0 03+11+02- 0193+ 0161+ 1 00+12+02+ 0402+ 0473+</td><td>1 03+03+04- 0713- U553- 1 03+03-04- 0804. 0911+ 1 04+03+04+ 1139+ 1112+</td><td>1 04+04+05* 0585* 0645* 0 04+04-05* 0574* 0570* 1 00+03+05* 0574* 0573*</td><td>1 00+01+07+ 0817+ 0864- 1 01+01+07+ 0864- 0562- 1 01+01+07+ 0864- 0562- 1 01+01-07- 0841- 0924-</td><td>0 02+11+00+ 0410+ 05404 0 00+12-00+ 0400+ 0517+ 0 01+12+00+ 0617+ 0499+</td><td>5 02+07+10+ 0487+ 0518+ 5 02+07+10+ 0291+ 0555+ 5 03+07+10+ 0291+ 0407+ 5 03+07+10+ 0491+ 0407+</td><td>1 01+08+12- 0508+ 0544+ 1 02+08+12- 0408+ 0504+ 0 02+08+12- 0506+ 0408+ 0 03+08+12- 0468+ 0408+</td><td>0 03+04-15+ 0423, 0516+ 0 03+04+15+ 0474+ 0475+ 0 03+04+15+ 0258- 0356- 5 04+04+15+ 0258- 0356-</td></t<>	1 01+06+01= 0948+ 0873+ 1 01+06+01= 0978+ 0873+ 1 01+06+01= 0978+ 1245- 1 02+06+01= 1171+ 1145*	1 01+11+42+ 0528+ 0611+ 0 03+11+02- 0193+ 0161+ 1 00+12+02+ 0402+ 0473+	1 03+03+04- 0713- U553- 1 03+03-04- 0804. 0911+ 1 04+03+04+ 1139+ 1112+	1 04+04+05* 0585* 0645* 0 04+04-05* 0574* 0570* 1 00+03+05* 0574* 0573*	1 00+01+07+ 0817+ 0864- 1 01+01+07+ 0864- 0562- 1 01+01+07+ 0864- 0562- 1 01+01-07- 0841- 0924-	0 02+11+00+ 0410+ 05404 0 00+12-00+ 0400+ 0517+ 0 01+12+00+ 0617+ 0499+	5 02+07+10+ 0487+ 0518+ 5 02+07+10+ 0291+ 0555+ 5 03+07+10+ 0291+ 0407+ 5 03+07+10+ 0491+ 0407+	1 01+08+12- 0508+ 0544+ 1 02+08+12- 0408+ 0504+ 0 02+08+12- 0506+ 0408+ 0 03+08+12- 0468+ 0408+	0 03+04-15+ 0423, 0516+ 0 03+04+15+ 0474+ 0475+ 0 03+04+15+ 0258- 0356- 5 04+04+15+ 0258- 0356-
1 03-00-01 100 100 100 100 100 100 100 100 1	0 02+0++v1+ 0/11+ 0781+ 1 02+05+01+ 00+4+ 0913+ 1 02+0++1+ 1190- 1281+	1 00+00+03+ 08/0- 0747- 1 01+00+03+ 1455- 1545- 0 01+00+03+ 0346- 0547- 1 01+00+03- 0346- 0547-	1 04+03+04- 0344+ 0354- 1 04+03+04- 10540- 1 04+03-04- 1057+ 1143+ 1 00+04+04+ 1135+ 1211+	1 00+09+05+ 4941- 0831- 1 01+04+05+ 4680- 4024- 1 01+04+05+ 0888- 0025- 0 61+03+05+ 0688- 0025-	0 02+01+07+ 0711= 0643- 1 02+03-07+ 0309+ 0414- 1 02+03-07+ 0309+ 0414-	1 01+00+08+ 0747+ 0768+ 1 01+00+09= 0724+ 0792+ 1 02+00+04+ 0677+ 0524+	1 04+07+10- 0348- 0360- 1 04+07+10- 0411- 0403+ 1 00+08+10+ 0424- 0748+	0 04+08+12= 0445+ 045+ 1 00+09=12+ 0685+ 0700+ 1 01+09+12= 0341+ 0355+	C 04+04+15* 0342+ 0378+ 1 00+05+15* 0440+ 0435+ 0 02+05+15* 0242+ 0381+
1 04:0-+01+ 08:0- 0778- 1 04:00+03- 0778- 0854- 1 00-00+04-0150-053- 0315-0314- 08:0- 1 04:00-039 0784-08:0- 1 04:00-039 0784-08:0- 00+04-04-00-031-08:0- 00+04-04-00-031-08:0- 00+04-04-00-031-08:0- 00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00-031-08:0-00+04-04-00+04-04-04-04-04-04-04-04-04-04-04-04-04-0	1 03+0+++14 1009- 6591+ 0 03+0+++14 0794, 0910+ 1 03+0+++1+ 0593, 0483+ 1 03+0+++1+ 1148- 1246+	1 02-00-03- 1503- 1489- 1 03-00-03- 0565- 0712- 1 03-00-03- 0563- 0763-	1 00+04-04+ 0799- 0889- 1 01+04-04+ 1294- 1201+ 1 01+04+04+ 1299- 0997-	1 02+09+05+ 05/3- 05/4- 1 02+0+05+ 05/3- 05/4- 1 02+0+05+ 05/1- 07/4-	1 03+01+07+ 0310- 0313- 1 03+01+07+ 0541- 0509- 1 03+01-07- 0719- 0761- 1 04+01+07+ 0494- 0444-	1 02+00+09- 0907. U888+ 1 03+07+09+ 0710+ 0738+ 1 03+00+09- 0511, 0557+	1 00+08+10+ 0825+ 0467+ 0 01+08+10+ 0418- 0850+ 1 01+08+10+ 0430- 0558+ 0 01+08+10+ 0430- 0558+	0 02+09+12- 0392+ 0515+ 0 03+09+12- 0330+ 0501+ 1 00+10-12+ 0394+ 0406+	1 04+05+15+ 0275+ 0347+ 1 01+01+18+ 0471+ 0482+ 1 02+01+18+ 0302+ 0288+ 1 02+01+18+ 0302+ 0288+
1 04-0-01 0 0-7 0 0-01 0-1 0-1 0-1 0-1 0-0 0-00 0-0	1 04-0-+11+ 0810- 0778- 1 04-01-01+ 0607- 0564+ 1 04-04-01= 0550+ 0564+	1 04.00.03. 0724- 0854- 1 04.01.03- 1047- 1095- 1 00.01.03. 1015- 1114-	1 01+04+04+ 1114+ 1189+ 1 02+04+04+ 0987+ 11837+ 1 02+04+04+ 0677+ 01837+ 1 02+04+04+ 1274+ 1184+	0 03+0'+0'5* 0524- 0544- 1 03-0'+05* 0536- 0576- 1 03+0'+05* 0516- 0576- 1 04+0'+05* 0516- 0452-	1 04+01-07+ 0524- 0482- 1 04+01-07+ 0741- 0744- 1 01+04-07- 0550- 0492-	1 04+00+09* 0477+ 0571+ 1 00+01+09* 08#5+ 1900+ 1 01+01+09* 08#5+ 1900+	0 02+0A+10- 034A- 051A+ 1 02+04-10- 0528- 0582+ 1 03+08+40- 0390- 0395+	1 00+01+13+ 0465- 0353+ 1 01+01+13+ 0509- 0482+ 0 02+01+13= 0605- 0609-	0 01+02-16- 0653- 0531- 0 02+02-16- 0429- 0563- 1 03+02-16- 0372- 0342-
1 0440-010 0548 1200 1 0010 0540 0540 1 0240 024 1341, 1345 0 0440 0550 1 0240 0470 0410 0240 0 010100 0574 0410 0240 0 0541 0574 0400 010 0550 1 0040 0410 0550 1 0040 1 0550 1 0040 0550 1 0040 0550 1 0040 1 0550 1 0040 0550 1 0040 0550 1 0040 1 0050 1 00	1 04+0++0)= 0948+ 1207+ 1 00+07+01+ 0710+ 0744+ 1 01+07+01+ 1111+ 0075+ 0 01+07+01+ 1111+ 0075+	1 01+01+03+ 04-04 D580+ 1 01+01+03+ 04-04 D580+ 1 01+01+03+ 04-04 D574+ D 01+01+03+ 13+7+ 1572+	1 02+04-04- 1361+ 1345+ 1 03+04+04+ 1974+ 1972+ 1 03+04-04+ 1769- 0943-	0 04+04+05+ (1541- 0672- 1 00+16+05+ 11498- 0538- 1 00+11-05+ 11697- 0581-	0 02+04+07+ 0410- 0246+ 1 02+04+07- 0512+ 0451+ 1 03+04+07- 0512+ 0451+ 1 03+04+07- 0574- 0430+	0 01+01+09- 0744+ UB2U+ 1 01+01-UD- 0714, 0594+ 1 02+01+U9+ 0544+ U556+	1 04+08+10- 0314- 0399+ 0 04+08+10- 0347+ 0854+ 1 00+09-10+ 0841- 0552+	1 04+01+13+ 0312+ 0291- 0 00+02+13+ 0491+ 0276+ 1 00+02+13+ 0558= 0539-	0 02+03-16- 0980- 0578" 0 03-03-18- 0387- 0338- 1 00-04-18- 0368- 0379-
0 01672-01 273, 00-75 1 01661-05, 073-0 1 03-06-14 0974-0155 4 100-0594 100-0593-1 0445-010-0150 0145-1 01655-0154-0555-1 01674-13 0275-0275-0275-0355-0554-0554- 1 01672-01-01424-0575-1 0475-0137-0554-0554-0554-0554-0554-0554-0554-055	1 01+07+01+ 0444+ 0675+ 1 01+07+01+ 0424+ 0675+ 1 01+07+01+ 0424+ 0627+ 1 02+07+01+ 0770+ 0711-	1 01+01-03- 0710- 0753- 1 02+01+03- 0603- 0759- 1 02+01+03- 0604- 1000-	1 03+04+04* 0974* 0806* 1 03+04+04* 0813. 0793. 1 04+04*04* 0843. 0904+	1 01+10+05+ (13+0)= 0473- 1 01+10+05- (644- 0528- 0 01+10-05- (644- 0528- 1 02+10-05- (644- 0170-	1 04+04+0/+ 0198+ 0198+ 1 04+04+0/+ 0198+ 0190+ 1 04+04+07* 0398+ 0347+ 1 04+04+07* 0549+ 0582*	1 07+01+09+ 0540, 0542+ 0 02+01+09+ 06,44, 0446+ 1 02+01+09+ 1010+ 1106+ 1 03+01+09+ 1010, 0920+	1 01+09+10= 0462= 0652+ 0 01+09=10= 0514= 0608+ 1 02+09+10= 0410= 0412+ 0 02+09=10= 0570= 0766+	1 01+02+13+ 0227+ 0277+ 1 01+02+13+ 0826+ 0534+ 0 01+02+13+ 0528+ 0520+ 1 01+02+13+ 0383+ 034+	0 02+04+18- 0307. 0457. 0 03+04+18- 0307. 0457. 1 00+05+18- 0517. 0540.
0 02/07/UI 0479, 0479, 02/07/07 1557- 1550, 10/07, 10/07 10/07 10/07 10/07 10/07 10/07 057, 0477 0487 1 00/07 057, 0477 0487 1 00/07 057, 0477 0487 1 00/07 057, 0477 0487 1 00/07 057, 0477 0487 1 00/07 057, 0477 0487 1 00/07 057 0478 1 00/07 057 0478 1 00/07 057 0488 1 00/07 057 0488 1 00/07 057 0588 1 00/07 057 0588 1 00/07 057 0588 1 00/07 057 0588 1 00/07 057 0588 1 00/07 057 0588 1 00/07	0 02+07+01+ 0817+ 0512+ 1 02+07+01+ 0874- 0930+ 1 03+07+01+ 0874- 0514- 0 03+07+01+ 0478- 0514-	0 02+01+03+ 1552- 1950- 1 02+01+03+ 0405+ 0481- 1 03+01+03+ 0410+ 0498- 1 03+01+03+ 0680+ 0878+	1 00+05+04+ 0978+ 0865* 1 00+05+04+ 0978+ 0908+ 1 00+05+04+ 0978+ 0908+	D 02+10+07 0440 0448+ D 02+10+07 03+1+ 0450+ 1 03+10+05 05/0+ 05/1+ 0 03+10+05 05/0+ 05/1+	5 00+05+07+ 0547+ 0487+ 5 01+05+07* 0545+ 0525+ 5 02+05+07* 1460+ 0360+	1 03+01+08+ 0543+ 0481+ 1 03+01-08+ 0748+ 0890+ 1 04+01+08+ 0844+ 0790+	0 03.09.10- 0401- 0581. 5 03.09-10- 0587- 0518. 0 04-09-10- 0319- 0710.	1 02+02-13+ 0618- 0595- 0 02+02+13+ 0618- 0595- 0 02+02+13+ 0637+ 0668- 0 02+02+13+ 0468+ 0375+	

FINAL ATOMIC PARAMETERS ^a									
Atom	x	y	3	<i>B</i> , Å*	Atom	x	У	g	B, Å2
U	-70(004)	-568(002)	7912 (001)	2.3	C(7)	4593(96)	1430 (66)	4400 (48)	2.2
Ρ	3478(24)	2071 (14)	7265 (10)	2.5	C(8)	6260 (110)	1612 (80)	4483(46)	5.7 ·
O(1)	-1763 (60)	24 (40)	7598(28)	3.7	C(9)	7070 (110)	2016 (81)	5376 (60)	6.2
O(2)	1478 (52)	1209 (36)	8230 (26)	2.9	C(10)	6233 (95)	2230 (70)	6276 (23)	4.3
O(3)	-336 (49)	-1791 (32)	6217 (22)	2.3	C (11)	3060 (90)	3519 (60)	7336 (40)	3.6
O(4)	1819 (44)	-2797(26)	7261 (18)	1.1	C (12)	4100 (65)	4754 (40)	7214 (26)	1.2
O(5)	-1491(48)	-1153 (30)	9284 (20)	1.7	C(13)	3503 (80)	5885(45)	7225 (40)	2.7
O(6)	1037 (54)	1369 (36)	9182 (24)	2.7	C(14)	1877 (100)	5784 (70)	7375 (40)	4.4
O(7)	1873 (44)	905 (29)	7090 (18)	1.2	C(15)	759 (101)	4570 (68)	7550 (41)	4.5
C(1)	-1176 (70)	-1901(40)	9900 (28)	1.5	C(16)	1610 (80)	3 600 (5 0)	7494 (35)	2.5
C(2)	-509(109)	-2964(86)	9600 (60)	6.5	C(17)	4825 (85)	2176 (58)	8359 (40)	3.4
C(3)	-1486(72)	-2969(44)	6317 (30)	1.8	C(18)	5214 (81)	3293 (40)	9121 (35)	2.7
C(4)	-2159 (96)	-4054 (64)	5510 (46)	4.2	C(19)	6160 (70)	3287 (46)	33 (30)	2.0
C(5)	4506 (78)	1977 (50)	6198 (25)	2.4	C(20)	6890 (105)	2220 (66)	52(48)	5.0
C(6)	3645 (80)	1591 (52)	5228 (36)	2.7	C(21)	6439 (85)	1234 (58)	9261 (40)	3.1
			· ·,		C(22)	5474 (90)	1142 (60)	8403 (41)	3.0

TABLE V

^a Coordinates are ×10⁴. Standard deviations from the least-squares refinement are in parentheses.

TABLE VI INTERATOMIC DISTANCES (Å) WITH THEIR STANDARD DEVIATIONS (× 10²)²

STANDARD DEVIATIONS (X10-)-							
I-O(1)	1.81 (5)	C(5)-C(6)	1.41(7)				
U-O(2)	1.75(4)	C(6) - C(7)	1.41(10)				
UO(3)	2.39(3)	C(7)-C(8)	1.34(10)				
U-O(4)	2.38(3)	C(8)-C(9)	1.30(8)				
U-O(5)	2.33(3)	C(9)-C(10)	1.44(8)				
U-O(6)	2.35(4)	C(10)-C(5)	1.37(7)				
U-O (7)	2.37(3)	C(11)-C(12)	1.40(7)				
		C(12)-C(13)	1.48(9)				
O(3)-C(3)	1.37(6)	C(13)-C(14)	1.35(10)				
O(4)-C(3)	1.32(5)	C(14)-C(15)	1.43(8)				
C(3)-C(4)	1.40(8)	C(15)-C(16)	1.45(10)				
O(5)-3(1)	1.37(6)	C(16)-C(11)	1.26(9)				
O'(6)-C(1)	1.26(7)	C(17)-C(18)	1.38(6)				
C(1)-C(2)	1.45(10)	C(18)-C(19)	1.44(7)				
P-O(7)	1.49(4)	C(19)-C(20)	1.49 (10)				
PC(5)	1.71(5)	C(20)-C(21)	1.30(12)				
P-C(11)	1.73(6)	C(21)-C(22)	1.38(7)				
PC (17)	1.81(5)	C(22)-C(17)	1.42(8)				

^a Mean value in the 18 C-C (phenyl) distances is 1.39 Å.

TABLE VII

INTRAMOLECULA	r Bond	Angli	es (deg) and thei	r Esd's	(deg)
Atoms	Angle	σ	Atoms	Angle	σ
O(1)-U-O(2)	177	2.7	O(7) - P - C(5)	107.7	2.5
O(1)-U-O(3)	90.4	1.6	O(7)-P-C(11)	111.5	3.5
O(1)-U-O(4)	91.6	1.6	O(7)-P-C(17)	112.6	2.5
O(1)-U-O(5)	88.1	1.6	C(5)-P-C(11)	101.7	3.2
O(1)-U-O(6)	88	1.7	C(5)-P-C(17)	111.8	4.1
O(1)-U-O(7)	90.9	1.6	C(11)-P-C(17)	111	3.2
O(2)-U-O(3)	89.6	1.5	C(6)-C(5)-P	122.8	3.9
O(2)-U-O(4)	85.9	1.6	C(5)-C(6)-C(7)	118.1	4.5
O(2)-U-O(5)	89.6	1.6	C(6)-C(7)-C(8)	123.7	4.4
O(2)-U-O(6)	93.4	1.6	C(7)-C(8)-C(9)	118.2	3.8
O(2) - U - O(7)	92	1.6	C(10)-C(9)-C(8)	122.7	3.7
O(3)-U-O(4)	53.5	1.3	C(9)-C(10)-C(5)	119.3	2.8
O(4)-U-O(5)	77.1	1.1	C(10)-C(5)-P	119.3	3.5
O(5)-U-O(6)	77	1.2	C(12)-C(11)-P	130.8	4.6
O(6)-U-O(7)	77	1.2	C(11)-C(12)-C(13)	124.1	5.8
			C(12)-C(13)-C(14)	120.9	2.8
C(3)-O(3)-U	98.8	2.3	C(13)-C(14)-C(15)	118.1	3.5
C(3)-O(4)-U	101.5	2.6	C(14)-C(15)-C(16)	111.5	6.5
O(3)-C(3)-O(4)	106.2	3.1	C(15)-C(16)-C(11)	128.3	7.2
O(3)-C(3)-C(4)	122.9	4.5	C(16)-C(11)-P	122.2	2.9
O(4)-C(3)-C(4)	130.8	4.5	C(18)-C(17)-P	117.7	3.2
C(1)-O(5)-U	128.3	2.8	C(17)-C(18)-C(19)	115	6.8
C(1)-O'(6)-U'	148	3.1	C(18)-C(19)-C(20)	121	3,9
O(5)-C(1)-O'(6)	113	4.8	C(19)-C(20)-C(21)	116	6.2
O(5)-C(1)-C(2)	125	5.2	C(20)-C(21)-C(22)	125	7.3
O'(6)-C(1)-C(2)	118.4	3.5	C(21)-C(22)-C(17)	118.5	3,1
U-O(7)-P	143	1.5	C(22)-C(17)-P	120	2.5

ful examination of $w\Delta^2$ values showed that $w\Delta^2$ did not pass the test suggested by Cruickshank, *et al.*¹²

No weighting system was found which entirely satisfied the above-mentioned test, however, the best scheme we could deduce was: if $|F_0| > 20$

$$w = \frac{1}{0.0439 |F_o|^2 + |F_o| + 25}$$

if $|F_{\circ}| \leq 20$

w = 0.01

With such a weighting scheme, the average value of $w\Delta^2$, dividing the reflections into groups of increasing F_o or according to increasing $(\sin^2 \vartheta)/\lambda^2$ or being selected at random, was sufficiently constant. After four cycles of isotropic refinement, each followed by a proper rescaling of $|F_o|$'s in each layer from the comparison of the observed with the calculated values, the *R* index fell to 9.5%. All calculations were performed on an Olivetti Elea 6001 3K computer; the block-diagonalmatrix, least-squares refinement was carried out using the program of Panattoni, *et al.*¹³

The observed and calculated structure factors are listed in Table IV. The final positional and thermal parameters are given in Table V.

A perspective drawing of the molecule with the atom numbering used is shown in Figure 1. The bond distances and valency angles with their standard deviations calculated from the least-squares residuals are given in Tables VI and VII and are reported in Figure 2 on a schematic drawing of the dimeric arrangement.

The best planes determined by the least-squares calculations are described by the equations in Table VIII.

⁽¹²⁾ D. W. J. Cruickshank, *et al.*, "Computing Methods and the Phase Problem in X-Ray Crystal Analysis," Pergamon Press Ltd., London, 1961, Paper 6.

⁽¹³⁾ C. Panattoni, G. Grubissich, and E. Frasson, Ric. Sci., Rend., Sez. A, 8, 995 (1965).

(a) Coefficient	$\sin AX + BY + CZ =$	D Referred to the Cry	vstallographic Axes	
Plane	A	В	с	D
α, equatorial plane O(3), O(4), O(5), O(6), O(7)	7.394	-7.209	4.389	3.817
3, bridging plane O(5), O(6), O'(5), O'(6)	7.599	-7.592	0.601	-0.300
y, chelate acetato O(3), O(4), C(3), C(4)	7.518	-7.213	3.721	3.357
b, bridging acetato O(5), O'(6), C(1), C(2)	6.419	3.599	0.277	-1.133
, phenyl group C(5)-C(10)	-1.198	10.813	-3.544	-0.577
r, phenyl group C(11)-C(16)	1.265	-0.446	12.774	9.933
, phenyl group C(17)-C(22)	-6.028	-3.317	7.171	2.345

Table VIII Least-Squares Planes

(b) Deviations of the Atoms from the Planes (Å \times 10³)

	α		γ		δ	·		s		η	
O(3)	46	O(3)	5	O(5)	- 19	C (5)	23	C(11)	17	C(17)	
O(4)	-40	O(4)	5	O(6)	1	C(6)	-7	C(12)	-13	C(18)	40
O(5)	14	C(3)	-17	C (1)	30	C(7)	-13	C(13)	-3	C(19)	-44
O(6)	8	C(4)	6	C (2)	-12	C(8)	19	C(14)	16	C(20)	27
O(7)	-27					C(9)	-3	C(15)	-12	C(21)	-4
\mathbf{U}^{a}	-13					C (10)	-18	C (16)	-5	C(22)	1
$C(3)^a$	3										
$C(4)^a$	73										
				(c)	Angles betw	een the Plan	les (deg)				

(1)						
16.8	∠ <i>ډ</i> ځ	88.2				
2.9	$\angle \epsilon \eta$	61.6				
63.7	$\angle 5\eta$	77.2				
	16.8 2.9 63.7	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$				

^a Atoms not included in the least-squares calculation.



Figure 1.—A perspective view of the molecule.

Results and Discussion

By treating uranyl acetate dihydrate with $(C_6H_5)_3$ -PO and $(C_6H_5)_3$ AsO two different products were obtained, of formula $UO_2(CH_3COO)_2L_2$ and $(UO_2(CH_3-COO)_2L_2)_3$



Figure 2.-- A schematic drawing of the dimeric arrangement.

 $COO)_2L)_2$. The presence of the dimeric species was recognized in solution by molecular weight measurements (Table I). The experimental values are generally below the theoretical, much more than one can expect from conductivity data in the corresponding solvent (Table II). This indicates that molecules of ligand are also partially lost in solution.

The monomeric complexes of the type $UO_2(CH_3-COO)_2L_2$ can be described as hexacoordinated species, congeners to $UO_2(NO_3)_2 \cdot 2(C_6H_5)_3AsO,^5$ where two water molecules are substituted by two ligands in the hexagonal plane normal to the uranyl ion. In fact,

the space group and crystal data determination¹⁴ proved that a trans-planar and centrosymmetric configuration can be assigned to molecules of such a type. Furthermore, the monomeric as well as the dimeric complexes of the phosphine and arsine oxide adducts were found to be isomorphous.¹⁵ The structure of the dimer consists of discrete molecules with two UO2- $(CH_3COO)_2(C_6H_5)_3PO$ units related by a center of symmetry and bridged by acetate groups, so that the resulting coordination number of the uranium atoms is seven. The coordination polyhedron is a pentagonal bipyramid realized by seven oxygen atoms, the uranylic oxygens being apical. The pentagonal base is defined by two oxygen atoms of a chelate acetato group, two oxygens of the bridging acetato groups, and the oxygen of the ligand as shown in Figure 1.

The uranyl U–O distances, 1.81 and 1.75 Å, may be considered equivalent. They may be compared with the distance of 1.78 Å found in the symmetrical uranyl group in rubidium uranyl nitrate¹⁶ and with 1.77 and 1.748 Å found in uranyl nitrate hexahydrate.¹⁷

These two U–O bonds make an angle of 177° with one another and angles ranging from 85.9 to 93.4° with the other coordinating bonds. Considering the presence of systematic errors (approximation in the absorption correction, the neglecting of the anomalous dispersion, and the extinction correction), we assume the uranyl group to be linear and normal to the equatorial plane. The five U–O distances in the equatorial plane are all included between 2.33 and 2.39 Å with a mean value of 2.36 Å.

The angles subtended at the uranium atom in the plane are approximately equal $(75.5-77^{\circ})$, with the exception of the angle subtended by the chelate acetate group, which has a value of 53.5° .

The chelate acetate group appears to be regular. It may be noted that the angles at the oxygen atoms are equal to those previously observed for a bidentate nitrato ion.⁵ The C(3) and C(4) carbon atoms are slightly tilted from the plane of the pentagon, the dihedral angle between this plane and the plane of the acetate group being about 3° . Since the two units of

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- (16) G. A. Barclay, T. M. Sabine, and J. C. Taylor, *ibid.*, **19**, 205 (1965).
- (17) J. C. Taylor and M. H. Mueller, *ibid.*, **19**, 536 (1965).

the dimer are related by an inversion center, the directions of the uranyl groups are parallel; and consequently the equatorial planes are also parallel, but they are not coplanar. The bridging acetates are tilted at 67° with respect to the bridging plane O(5), O(6), O'(5), O'(6). The U'-O'(6)-C(1) angle (148°) is about 20° greater than the U-O(5)-C(1) angle. In addition, the O'(6)-C(1) distance (1.25 Å) is shorter than the corresponding O(5)-C(1) distance (1.37 Å). The difference in these values is not particularly significant considering the standard deviations. However this fact could be associated with the difference in the angles at the oxygen atoms, although there is no evident explanation for this.

A general discussion of the P–O bond has been made by Lindqvist¹⁸ by considering the influence of the substituents in ligands of X_3 PO type and of the acceptor.

The P–O distance of 1.49 Å is considerably less than the single-bond value. Similar short bonds have been found in some P–O and As–O distances^{19–22} and can be ascribed to the strong π character of these bonds.

The U–O–P angle of 143° can be compared with the value of 153° found for the U–O–As angle.⁵ The influence of steric hindrance on these values appears to be excluded since, the substituents being the same, the U–O–As angle is about 10° larger than the U–O–P. Other factors may be responsible for this difference, as for instance the contribution of the back-donation from the oxygen to the d empty orbitals of the arsenic and phosphorus atoms. It may be noted that, if we assume the phosphorus and the arsenic atoms have the same hybridization, the bonding orbitals of the phosphorus are comparatively more electronegative, and this should be in agreement with a U–O–As angle larger than in the phosphine adduct.

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⁽¹⁴⁾ R. Graziani, B. Zarli, and G. Bandoli, Ric. Sci., 37, 984 (1967).

⁽¹⁸⁾ I. Lindqvist, "Inorganic Adduct Molecules of Oxo Compounds," Springer-Verlag, Berlin-Göttingen-Heidelberg, 1963.

⁽²⁰⁾ C. I. Branden, Arkiv Kemi, 22, 485 (1964).