SINGLE CRYSTAL PEROVSKITES AS ELECTROOPTICALLY ACTIVE MATERIALS

THIRD QUARTERLY PROGRESS REPORT

JANUARY 1, 1967 to APRIL 1, 1967

CONTRACT NO. AF 33(615)-5410

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Sponsored by: Air Force Material Laboratory Research and Technology Division Air Force Systems Command United States Air Force

Written by: Work done by: H. Fay, Principal Investigator

H. Fay W. J. Alford A. M. Broyer

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UNION CARBIDE CORPORATION ELECTRONICS DIVISION

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TABLE OF CONTENTS

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Section		Page
ABSTRACT		1
I.	SUMMARY	2
II.	POTASSIUM TANTALATE NIOBATE (KTN) GROWTH	3
III.	COMPLEX PEROVSKITE COMPOUNDS	7
IV.	OPTICAL AND ELECTROOPTIC TESTS	10
v.	PLANS FOR NEXT QUARTER	14
REFERENCES		15

LIST OF TABLES

	'Table of Jons	16
	Output Lists of Compounds Generated by Computer for $P = 2$, $TC = 3$	17-27
Table I.	Dimensions and Curie Temperatures for KTN Crystals	Follows Fig. 3

A

LIST OF FIGURES

•

Fig. 1	Experimental Schematic
Fig. 2	Unfinished KTN Crystals Photographec in Crossed Polarized Light
Fig. 3	Selected KTN Crystals Photographed in Crossed Polarized Light
Fig. 4	Electrooptic Patterns and Extinction Ratios - Crystal 142-C
Fig. 5	Transmission Photographs of KTN Crystal NF-II-142-C in Polarized Light (5461Å)
Fig. 6	Transmission Photographs of KTN Crystal NF-II-142-C With D-C Electric Field Applied (Parallel Polarizers 5461Å)
Fig. 7	Electrooptic Patterns at 0.546 and 1.06 μ (KTN Crystal NF-II-143F Antireflection Coated for 1.06 μ)
Fig. 8	Experimental Arrangement for In-Cavity Pulsing of Nd:YAG Laser With KTN Electrooptic Switch
Fig. 9	Electrooptic Pulsing of Nd:YAG Laser Loss Modulation With Sinusoidally Driven KTN Retarder
Fig. 10	Electrooptic Pulsing of Nd:YAG Laser Phase Modulation With Sinusoidally Driven KTN Retarder Pulse Mode Locked Output

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ABSTRACT

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The Czochralski growth of KTN was continued in order to determine reproducibility. In addition, a new growth technique was tested. Thirteen perovskite compounds were selected from the computer output data for melt studies on the strip heater. Twelve of the best KTN crystals were fabricated, etched, and tested for electrooptic properties; one crystal was operated as a switch for a Nd:YAG laser.



I. <u>SUMMARY</u>:

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Potassium tantalate-niobate (KTN) crystals were grown primarily from two globar furnaces. No major changes were made in the growth parameters in order to determine the level of reproducibility of crystal quality. Since this turned out to be low, a new growth technique was tried with some hopeful indications.

Computer calculations continued to be used to generate possible perovskite compounds, with additional refinements being added to eliminate undesirable ions.

Thirteen compounds generated for P = 2, TC = 3 were selected for preliminary investigation. Work with the iridium-strip furnace progressed slowly, although preliminary tests indicate that it will be possible to obtain DTA data optically.

Sixteen of the best KTN crystals were selected for careful study, twelve being optically finished for testing as electrooptic elements. In one crystal an extinction of the first half-wave was 20 db over a 4-mm aperture and the crystal showed a conductivity of $65 \pm 5 \times 10^{-14} \text{ n}^{-1} \text{ m}^{-1}$. Another crystal was coated with an antireflection dielectric coating, inserted into a Nd:YAG laser cavity, and used as a loss modulator or phase-modulator for pulsing the laser.

II. POTASSIUM TANTALATE NIOBATE (KTN) GROWTH:

In order to determine the reproducibility of good quality KTN crystals, growth was carried out over a period of several weeks without making any major modifications in the growth conditions. As related in the previous report, several crystals of good optical quality had been grown, and it was our intention to determine whether the growth conditions which were arrived at were good enough to allow the consistent growth of high quality KTN. Briefly, these conditions were:

- A melt composition of KTa_{0.32}Nb_{0.68}O₃ yielding a crystal composition of KTa_{0.63}Nb_{0.37}O₃.
- 2. An excess of four weight percent K_2O added to the melt.
- 3. Maintaining a slight outward taper on the crystal during growth.
- 4. Maintaining careful temperature control during growth, while positioning the crucible at a point where the bottom of the melt is about 50° hotter than the top, thereby providing active thermal stirring of the melt. This is essential in order to remove the Nb⁵⁺ which is rejected at the growth interface and to promote chemical homogeneity in the melt.
- 5. Pull rates of 0.02 to 0.03 inch/hour.
- 6. Rotation of the crystal at 60 rpm during growth.
- 7. Providing an oxidizing atmosphere over the melt in order to prevent reduction of Ta^{5+} and Nb^{5+} to lower oxidation states.
- 8. Very slow cooling of the crystal after growth to minimize cracking.

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Although several good crystals were grown during this period, no reproducibility was obtained. The yield of usable fabricated cubes was less than 10%, and many of these cubes are on the order of 3 mm³ or less. Even though this is a good deal better than had previously been possible, it appeared that the limits of the growth system had been reached after almost two years of experimentation on the globar furnace. In addition, Czochralski growth from flame-heated and platinum resistance furnaces provided crystals which were inferior to those grown in the globar furnaces. In view of this, it was decided to use another growth technique.

After careful deliberation, it was decided to use a modified float-zone growth technique. The furnace consisted of two platinum resistance tube furnaces, each 2 inches I. D. x 12 inches long, one being placed on top of the other. The furnaces are furnished with taps so that it is possible to provide a wide variety of internal temperature profiles. The temperature in the top furnace was made about 150° C hotter than the lower furnace with a sharp gradient at their intersection. A platinum crucible in the shape of cylinder, one end being open and the bottom terminating in a closed cone. The crucible was 1 inch I. D., 5 inches long, and the cone section was 1 inch long. A Pt-13% Rh Pt thermocouple was attached to the tip of the cone.

In operation the top furnace was heated to 1250° C and the lower furnace to 1100° C. The crucible was charged with KTN powder so the melt height was about 2-1/2 inches. The crucible was lowered through the point of steepest temperature gradient (about 150° C over 1/2 inch) at a rate of 0.02 inch per hour. When the tip of the cone reached 1180° C, crystallization began to occur, according to the phase diagram. Since the crystal composition is different from that of the melt, the melt composition begins to change as soon as crystallization is initiated.

-4-

In order to alter this change of melt chemistry, a powder feed system was arranged above the top of the crucible. This system fed an adjustable amount of sintered KTN powder having the same chemical composition as that of crystal growing in the bottom of the crucible. In this way, the composition and volume of the melt are maintained constant, thus eliminating the built-in chemical gradient present in Czochralski growth. Another benefit derived from this system is that crystal grows on the bottom of the crucible and is thus protected from thermal shock, as well as the fact that watential crystal sizes are greater.

Six runs were made ranging from six to ten days in length. Several problems were encountered, such as variation of the crucible lowering rate, clogging up of the powder feed tube and the development of leaks in the crucible. The first four runs resulted in polycrystalline growth and showed that seeding would be necessary to promote monocrystalline growth. In order to achieve this, the crucible was modified as shown below:



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In the two seeded growth attempts, the crucible was positioned so that the seed was at the point of maximum thermal gradient and the furnaces were slowly brought to temperature. After reaching the proper conditions, the crucible was lowered at 0.02 inch/hour. In both runs, mechanical problems were encountered, but the results were somewhat more encouraging in that some single crystal growth took place. However, the growths were small and badly cracked, indicating that either the thermal gradients are too large or that in cooling, stresses develop around the crystal due to differences in contraction between the crucible and the solidified melt.

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Our plans for the coming period are to pursue the new growth technique, as it seems to offer the best probability of yielding high quality KTN.

III. COMPLEX PEROVSKITE COMPOUNDS:

The objective of the work on complex perovskite compounds is to find new materials that have an electrical Curie point in the vicinity of room temperature that could be used as electrooptical elements. Computer calculations have been used to generate and test new hypothetical compounds. Growth tests are then made to see if the compounds can be synthesized. Compounds that can be synthesized and identified will then be tested for dielectric properties. Finally crystal growth development studies will be made to produce sizeable crystals of the most promising materials. We are now completing the computer studies and making growth tests on selected compounds.

Computer Study of Complex Perovskites

Many tests of computer programs for generating complex perovskites have been made and the number of potential compounds has been found to be very large when all possible ions are included. Since many ions are undesirable, because of oxidation-reduction problems, etc., it has been decided to preselect the ions that are considered most suitable for crystal growth. Ions known to contribute to high dielectric properties were also retained. By making such a reduction in the ion list, the number of generated compounds is reasonably small. The resultant compounds are also substances which should be amenable to crystal growth, since ions of unusual valence have been removed.

An example of the edited ion list is shown in the "Table of Ions" at the end of this report. Following this table, we have reproduced the output lists of compounds generated by the computer for P = 2, TC = 3. There are 156 compounds. The output for P = 3, TC = 3 is also shown. There are 126 compounds. In these programs we have introduced a new test, the P-test, based on the electronegativity of the ions, which is read in as part of the ion input. The electronegativities (EN) were taken from Cotton and Wilkinson⁽¹⁾. Two P-factors are calculated as follows:

-7-

$$PP = \frac{EN_{O_2} - EN_A}{r_A^2} + \frac{EN_{O_2} - EN_B}{r_B^2}$$
$$PN = \frac{EN_{O_2} - EN_A}{r_A^2} - \frac{EN_{O_2} - EN_B}{r_B^2}$$

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The ratio PP/PN should be a measure of the "forces" within the crystal. This ratio is found to be between 1.7 and 3.0 for the known perovskites. We have used these limits in the programs.

Data on some 450 known complex perovskite compounds has been coded on punched cards so that average ionic radii, calculated cell size, T-factors, and P-factors may be computed. Listings of these known compounds have been made but the output formats are still being improved. These listings will be presented in the annual summary report.

Thirteen of the compounds generated for P = 2, TC = 3 have been chosen for preliminary growth tests. They are:

K₂ZrWO₈ KLaZr₂O₆ RbYZr₂O₆ Sr₂MgWO₆ Ba₂CaWO₆ Ba₂ZnWO₈ Ba₂ScTaO₆ Ba₂YTAO₆ Ba₂GaTaO₈ Pb₂GaTaO₈ La₂LiTaO₆ Growth tests are also being performed on the known ferroelectric compound $Pb_3Nb_2MgO_8$. The results of these tests will be reported later.

Work with the iridium-strip furnace is progressing slowly. Some of the growth tests have been performed with the strip furnace while others have been made in crucible furnaces, which more nearly approximate the conditions to be used in the single crystal growth work to be done later. It was intended to employ the strip furnace to get preliminary fusion and reaction data, using special optical detection equipment. By observing both the sample and the strip, it should be possible to obtain DTA (differential thermal analysis) information optically. Preliminary tests have shown that this can be done. A sketch of the experimental arrangement is shown in Fig. 1. By a simple system of lenses, the image of the iridium strip is focused at the imaging screen so that the sample image falls on one hole and a portion of the strip on the other hole in the screen. The light which is impolarized passes through a split field polarizer so that the two beams are orthogonally polarized. The "chopper" polarizer sinusoidally modulates each beam so that the light signals have a relative phase 180° to each other. A third polarizer acts as a zero control by attenuating the beams by an amount dependent on the angular setting. Both light paths impinge on a wide screen, infrared-sensitive photomultiplier (7102). The average light level at the photomultiplier should be proportional to the total radiation which is mainly dependent on the temperature. This level may be measured by recording the d-c output from the photomultiplier. When the two light beams are at equal intensity there is no a-c component at the photomultiplier. An intensity difference, however, produces an a-c signal proportional to this difference and therefore proportional to the difference in radiation of the sample and the strip. The a-c signal can be amplified, rectified and recorded. An auxiliary light and phototube can be used to obtain a synchronizing signal for use with a lock-in amplifier. An experimental set-up of the type shown in Fig. 1 (except for the zero-control polarizer) has shown that the system does work to detect differential radiation. We are now building a more permanent detector assembly.

-9-

IV. OPTICAL AND ELECTROOPTIC TESTS:

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Some 60 crystals of KTN have been examined in crossed polarized light to determine their potential quality as electrooptic elements. These crystals were only roughly finished and not optically polished. Photographs of the crystals are shown in Fig. 2. Ideally, KTN should be isotropic and appear black in the photographs. In no case is this found; there is some birefringence in all the crystals shown. In some crystals the pattern is complex and definitely indicates inhomogeneities in the crystal. In many crystals, however, the pattern is symmetric and appears similar to what might be expected if the crystal were strained.

Sixteen of the crystals were selected for more careful study. They are shown in Fig. 3 and the crystal dimension and Curie temperatures are given in Table I. We have learned, from previous work of Van Uitert at Bell Telephon: Laboratories, that external strain can be eliminated by etching the crystals in molten KOH. Twelve of the crystals were etched by immersion in KOH at 400°C for one minute and re-examined. The lower photograph in Fig. 3 shows the result. In most cases the birefringence is markedly decreased, indicating that it was due to surface strain. This strain is apparently introduced when the crystals are cut, ground and polished. The etching relieves this strain but it also produces a textured surface. For optical applications, the faces must be polished and the strains produced in the polishing operation will cause some birefringence to reappear. We have found that the strain-relieved pattern is not appreciably altered if only the two faces required to transmit the light are polished. The other faces are not reworked but are left with etched surfaces.

The twelve crystals in Fig. 3 (lower) have been optically finished with plane parallel polished faces so that they could be tested as electrooptical elements. Before final finishing, the usual electrooptic pattern was photographed. The extinction ratios, which were not very large before finishing, were greatly improved when the finished crystals were tested. While most of the crystals have been superficially examined, these tests are continuing and will be reported later.

-10-

More complete data has been obtained on Crystal 142-C which was chosen for transmittal to the Air Force under the terms of the contract. The electrooptic patterns and measured extinctions are shown in Fig. 4. Despite the fact that there is some residual passive birefringences, seen by the dip at the origin, the behavior of the crystal is quite good. The measured extinction ratios for the first and third half-wave are shown in the graph, as a function of aperture. Even with a 4-mm aperture the extinction of the first half-wave is 20 db.

The transmission photographs reproduced in Fig. 5 show the crystal to be fairly uniform. There are a few definite "spots" that may be local internal or surface points of strain. Otherwise, the crystal transmits and extinguishes uniformly. The effect of applying a d-c field to the crystal between parallel polarizers is shown in Fig. 6. The applied voltage corresponded approximately to that required for halfwave retardation. Initially, the crystal extinguishes nearly uniformly and completely, except at the strain spots. However, the extinction degrades with time, as shown in the lower photograph. Interference bands appear to originate at one of the electrodes and migrate through the crystal. The crystal is thus not stable under d-c bias voltages but changes. We have not maintained the voltage for long periods and do not know whether or not the drifting of the bands persists or if a steady state establishes. In any event, this crystal gives evidence of charge carrier migration and thus should exhibit a finite conductivity. This conductivity has been measured by applying a fixed voltage to the crystal and measuring the current passed through it after the initial "charge up". A value of $65 \pm 5 \times 10^{-14}$ m⁻¹ was obtained. This conductivity, while quite low, is apparently enough to cause migration of the interference bands. We note that crystal NF-II-151, which was measured and reported in the last progress report, had a conductivity two orders of magnitude less (6 x 10^{-15}). That crystal was apparently bias stable for 5 min. or more. It appears that true bias stability will demand very low values of conductivity. At present the identification of the charge carriers and their relation to the crystal chemistry is unknown.

-11-

One of the dozen selected crystals (NF-II-143F) has been coated with an antireflection dielectric coating (by Lambda Optics) for maximum transmission at 1.06 μ . The purpose of this was to make it possible to insert the crystal inside the cavity of a c-w, Nd:YAG laser, without extinguishing the laser. The electro-optic patterns for the crystal are shown in Fig. 7. The electrooptic coefficients are only about one third as large at 1.06 μ than at .546 μ . The first half-wave at 1.06 μ thus requires about 1.7 times the voltage required at 0.546 μ . The patterns in Fig. 7 also indicate that the crystal has some passive birefringence; there is a slight dent at the origin.

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The experimental arrangement is shown in Fig. 8. The Nd: YAG laser was operated as an approximately confocal-cavity oscillator. The laser was polarized with a single quartz Brewster plate. The KTN crystal was placed near the center of the confocal cavity. Insertion of the KTN caused the laser to extinguish or drop appreciably in output. After retuning the cavity, however, the laser could be operated c-w at a low level. Much of the insertion loss is attributed to the passive birefringence which degrades the transmission at the origin. The KTN crystal could be used as an electrooptic switch or modulator in either of two ways. When the Brewster polarizer was aligned at 45° to the field direction, the crystal acts as a birefringent loss modulator. When the E-field is increasing, the crystal becomes a retarder. The emergent light is elliptically polarized and reflects back from the mirror in the opposite "handedness". When the crystal is a quarter wave retarder, the laser should be completely spoiled. However, much lower losses cause this laser to extinguish. In fact, the laser usually only comes on near the origin as shown in Fig. 9. This produces a repetitively pulsed output. This behavior results when the crystal is driven such that the voltage approaches, but does not equal, the single pass half-wave voltage. When driven to or beyond this half-wave voltage the crystal will turn on again at this level. This results in unequal pulses of twice the frequency, as shown in Fig. 9. Many other more complex patterns are obtained for a variety of other driving conditions.

-12-

If the Brewster polarizer is aligned parallel to the E-field, there should be no effects from birefringence, passive or induced, and the crystal acts as a pure phase-modulator. At appropriate driving levels the laser will become "mode locked" and produce a pulsed output. Such behavior is shown in Fig. 10. Rather sharp pulses are obtained even though the crystal is driven sinusoidally. These experiments demonstrate that the best of our KTN crystals, even with some passive birefringence, can be used as a loss-modulator or phase-modulator for pulsing a laser.

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V. <u>PLANS FOR NEXT QUARTER:</u>

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During the next quarter, we shall de-emphasize the KTN growth effort and increase the effort on complex compounds. The computer work will be completed and presented in the annual summary report.

Electrooptic tests on KTN will be continued. Crystals designated for delivery to the Air Force will be retained until the data are complete unless more rapid delivery is requested.

REFERENCES:

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 F. A. Cotton and G. Wilkinson, "Advanced Inorganic Chemistry". Interscience, New York, 1966 p. 92.



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P FACTOR	2.000	2.067	2.013	1.724	1.834	1.898	1.846	2.066	2.208	2.303	2.226	1.971	2.105	2.201	2,123	1.989	2.127	2,245	2.148	2,180	2,335	24452	2.357	466.1	2.062	2,136	2.076	2.444	2.634	2.858	2.672
T FACTOR	.890	•905	. 893	•955	• 932	.948	466 .	• 888	. 868	• 882	.870	.912	• 890	• 905	.893	• 923	106.	.916	E06 •	.878	.859	.872	•860	• 903	.882	.897	•88•	•880	•860	+84	•862
CELL	8,129	8.059	8.119	7.839	7.939	7.869	7.929	8,139	8,239	8,169	8.229	8,029	8,129	8.059	8,119	7.979	8.079	6,009	8,069	8,189	8.289	8.219	8.279	8,069	8.169	8,099	8.159	8.179	8.279	8.209	8.269
B RAD	.75	.71	.74	.61	. 66	• 62	.65	.75	•80		68.	.70	. 75	.71	.74	• 68	.73	69.	.72	. 79	.83	• 79	.82	.72	. 77	. 73 5	. 77	. 77	.82	. 79	•82
A RAD	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20	1.20
SNB	14	51	73	23	4	2	13	53	4	51	73	59	4		12	53	4	51	52	53	14	51	5	23	4	51	13	53	4	51	1
INB	21	21	21	31	16	5	1	6	90	39	39	49	49	•	4	51	- 11	5	5	4	49	49	64	11	12	12	11	83	69	83	68
ANZ	82	32	82	28	62	58	2		28	82	82	82	6	28	28	6	6	82	28	69	82	28	82	82	82	82	82	58	28	82	82
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| +5<br>VALENCE  | N 4<br>1 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           |
| +4<br>VALENCE  | TI                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |
| +3<br>VALENCE  | AI                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |
| +2<br>VALENCE  | MG                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |
| +1<br>VALENCE  | 26. E L L 9<br>26. E L 4 9<br>1 9<br>1 9<br>1 9<br>26. I 1 9<br>27. I |

|             | TOTAL A          | A CATION V | ALENCE    | m<br>#     | -   | OTAL B | CATION | VALENCE = 1: |       |             | 36       | 0        |
|-------------|------------------|------------|-----------|------------|-----|--------|--------|--------------|-------|-------------|----------|----------|
| Сн          | MICAL FORMULA    | NA         | 'I ANZ    | ANA        | ING | BN2    | BN3    | A RAD        | 8 RAD | כבור        | T FACTOR | P FACTOR |
| х:<br>Х:    | K I NO 6 NO 6 SC | 3 1        | 9 19      | 19         | 42  | 42     | 21     | 1.33         | A.A.  | 0<br>0<br>0 |          |          |
| K 1 K 1     | K 1 MO 6 MO 6 Y  | 3          | 9 19      | 19         | 42  | 64     | 96     | 1 . 33       |       |             | . 7 . 5  | 788.1    |
|             | K 1 MO 6 MO 6 IN |            | 9 10      | 0          | 107 |        |        |              | 2.    | 660.00      | 954.     | 2.009    |
| K l X l     | K 1 MU 6 MU 6 GU | ÷ ۹        |           | 1          | 1   | N I    |        | 1.33         | • 68  | 8.025       | .973     | 1.941    |
| - ×         |                  | • •        |           | <u>, 1</u> |     | 24     | 40     | 1.33         | - 74  | 8.132       | . 949    | 2.082    |
| <br><br>    |                  |            | 61 6<br>6 | 61         | 24  | 42     | 11     | 1.33         | • 70  | 8.052       | .967     | 1.921    |
|             |                  |            | 4 I9      | 19         | 42  | 42     | 83     | 1.33         | . 73  | 8.125       | . 950    | 2,221    |
| •<br>•<br>• |                  |            | 4<br>19   | 19         | 46  | 4      | 21     | 1.33         | . 68  | 8.025       | .973     | 1.922    |
|             |                  |            | 61 6      | 61 :       | 42  | 44     | 39     | 1.33         | .72   | 8.099       | .956     | 2-056    |
|             |                  |            | 5 I .     | 61         | 42  | 74     | 64     | 1.33         | . 68  | 8.025       | .973     | 1-986    |
|             |                  |            | 5 C       | 51.        | *1  | 4      | 64     | 1.33         | ÷2 •  | 8.132       | 646.     | 2-133    |
|             |                  |            | 5 C       | 23         | ti  |        | 17     | 1.33         | .70   | 8.052       | .967     | 1.963    |
| •           |                  | -<br>-     | 7 I.Y     | 51         | t   | 41     | 83     | 1.33         | .73   | 8.125       | .950     | 2.288    |

TOTAL A CATION VALENCE = 3

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มข้อมูลรูงแขนมาให้หมูล สุขภาพให้สุขชุดที่แขนหมูล

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TOTAL B CATION VALENCE = 12

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|------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|--------------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|--------------------------------------|-------------------------------|-------------------------------|-------------------------------|--------------------------------|--------------------------------|--------------------------------|--------------------------------|-------------------------------|--------------------------------|--------------------------------|-------------------------------|-------------------------------|-------------------------------|------------------------------|-----------------------------|----------------------------|-------------------------------|------------------------------|-------------------------------|------------------------------|-------------------------------|
| P FACTOR         | 2,327                       | 2,279                       | 2,943                       | 2.810                       | 2.337                       | 2.106                       | 2.701                       | 2.813                       | 2.637                       | 2.122                                | 2.729                         | 2.844                         | 2.663                         | 2.096                         | 2.683                         | 2.793                         | 2.619                         | 440 • Z                       | 2.589                         | 2.690                         | 2.530                         | 2.005                         | 2.518                         | 2.613                         | 2.463                         | 1.951                         | 2.424                         | 2.510                         | 2.997                                | 2.374                         | 2,051                         | 2.600                         | 2.703                          | 2.541                          | 1.895                          | 2.328                          | 2.406                         | 2.842                          | 2.282                          | 2.938                         | 2.996                         | 2.061                         | 2.085                        | 2,374                       | 2.408                      | 2.220                         | 2.252                        | 2.262                         | 2.300                        | 810.7                         |
| T FACTOR         | .907                        | . 697                       | .855                        | .853                        | • 905                       | .936                        | .888                        | .922                        | .892                        | .928                                 | .881                          | -915                          | . 885                         | • 909                         | . 863                         | . 896                         | .867                          | 246.                          | •864                          | .928                          | . 898                         | .932                          | •88•                          | .918                          | . 888                         | .931                          | .883                          | 116.                          | .922                                 | .887                          | 146.                          | . 893                         | .927                           | .897                           | 116.                           | .921                           | .957                          | .962                           | .926                           | .867                          | . 267                         | • 948                         | .948                         | .916                        | .916                       | .948                          | .948                         | • 963                         | .963                         | 204.                          |
| CELL             | 7.741                       | 7.700                       | 7.900                       | 7.895                       | 7.736                       | 7.860                       | 8.080                       | 7.920                       | 8.060                       | 7.829                                | 8.049                         | 7.889                         | 8.029                         | 7.752                         | 7.972                         | 7.812                         | 7.952                         | 1.885                         | 0.105                         | 7.945                         | 8.085                         | 7.844                         | 8.064                         | 7.904                         | 8.044                         | 7.839                         | 8.059                         | 7.899                         | 7.879                                | 8.039                         | 7.880                         | 8.100                         | 7.940                          | 8.080                          | 8.003                          | 8.223                          | 8.063                         | 8.043                          | 8.203                          | 7.828                         | 7.828                         | 8.168                         | 8.168                        | 8.314                       | 8.314                      | 8.168                         | 8.168                        | 8.101                         | 8.101                        | 8.381                         |
| B RAD            | .68                         | . 68                        | .78                         | . 78                        | . 68                        | • 68                        | . 79,                       | .71                         | . 78                        | .68                                  | • 79                          | .11                           | . 78                          | • 68                          | • 19                          |                               | • 78                          | 89 <b>.</b>                   | 61.                           | .71                           | . 78                          | . 68                          | • 79                          | .71                           | . 78                          | . 68                          | • 79                          | .71                           | .70                                  | . 78                          | . 68                          | . 79                          | .71                            | . 78                           | .68                            | . 79                           | .71                           | . 70                           | .78                            | .75                           | . 75                          | . 75                          | . 75                         | <b>.82</b>                  | .82                        | . 75                          | • 75                         | .71                           | 12.                          | CQ •                          |
| A RAD            | 1.15                        | 1.12                        | 1.12                        | 1.12                        | 1.15                        | 1.23                        | 1.23                        | 1.23                        | 1.23                        | 1.21                                 | 1.21                          | 1.21                          | 1.21                          | 1.16                          | 1.16                          | 1.16                          | 1.16                          | 1.24                          | 1.24                          | 1.24                          | 1.24                          | 1.22                          | 1.22                          | 1.22                          | 1.22                          | 1.21                          | 1.21                          | 1.21                          | 1.21                                 | 1.21                          | 1.2%                          | 1.24                          | 1.24                           | 1.24                           | 1.32                           | 1.32                           | 26.1                          | 1.32                           | 1.32                           | 1.12                          | 1.12                          | 1.34                          | 1.34                         | 1.34                        | 1.34                       | 1.34                          | 1.34                         | 1.34                          | 1.34                         | 1.34                          |
| 6N3              | 22                          | 22                          | 72                          | 72                          | 22                          | 22                          | •                           | 50                          | 72                          | 22                                   | 40                            | 50                            | 72                            | 22                            | 4                             | 20                            | 22                            | 22                            | 40                            | 50                            | 72                            | 22                            | 40                            | 50                            | 72                            | 22                            | •                             | 50                            | 52                                   | 72                            | 22                            | 40                            | 50                             | 72                             | 22                             | 40                             | 20                            | 52                             | 72                             | 42                            | 14                            | 42                            | 74                           | <b>4</b> 2                  | 41                         | 42                            | 14                           | 42                            | 2                            | N4                            |
| BN2              | 22                          | 22                          | 72                          | 72                          | 22                          | 22                          | 40                          | 50                          | 72                          | 22                                   | 40                            | 50                            | 72                            | 22                            | 4                             | 205                           | 22                            | 22                            | 4                             | 20                            | 72                            | 22                            | 40                            | 50                            | 72                            | 22                            | 40                            | 50                            | 52                                   | 72                            | 22                            | 40                            | 50                             | 72                             | 22                             | 40                             | 50                            | 52                             | 72                             | 21                            | 21                            | 21                            | 21                           | 9 E                         | 96<br>E                    | 49                            | 64                           | 51                            | 5                            | 40                            |
| INB              | 22                          | 22                          | 72                          | 22                          | 22                          | 22                          | <b>9</b>                    | 20                          | 72                          | 22                                   | 40                            | 50                            | 72                            | 22                            | 4                             | 20                            | 72                            | 22                            | 4                             | 50                            | 72                            | 22                            | 4                             | 50                            | 72                            | 22                            | 40                            | 50                            | 52                                   | 72                            | 22                            | 40                            | SC                             | 72                             | 22                             | 40                             | 20                            | 52                             | 72                             | 21                            | 21                            | 21                            | 21                           | 39                          | 39                         | 49                            | 64                           | 51                            | 51                           | •                             |
| ANB              | 40                          | 20                          | ŝ                           | 22                          | 22                          | 6                           | 8                           | 6                           | 6                           | 22                                   | 22                            | 22                            | 22                            | 32                            | 32                            | 32                            | 32                            | 4                             | 9                             | <b>9</b>                      | 4                             | ß                             | ŝ                             | 50                            | ŝ                             | 52                            | 52                            | 52                            | 52                                   | 52                            | 22                            | 21.                           | 72                             | 72                             | 8                              | 6                              | 8                             | 6                              | 66                             | 38                            | 38                            | 56                            | 56                           | 56                          | 56                         | 56                            | 56                           | 56                            | ŝ                            | \$                            |
| ANZ              | 19                          | 19                          | 19                          | 19                          | 19                          | 61                          | 19                          | 19                          | 19                          | 37                                   | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 5                             | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                                   | 37                            | 9.1<br>1                      | 37                            | 37                             | 11                             | 37                             | 37                             | 37                            | 37                             | 37                             | 38                            | 38                            | 56                            | 56                           | 56                          | 56                         | 56                            | 56                           | 56                            | 3.¢<br>1                     | 20                            |
| ANI              | 19                          | 19                          | 19                          | 19                          | 19                          | 19                          | 19                          | 19                          | 19                          | 37                                   | 37                            | 16.                           | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 37                            | 75                            | 37                                   | 37                            | 37                            | 37                            | 37                             | 37                             | 37                             | 37                             | m<br>I                        | 37                             | 37                             | 38                            | 3 <b>8</b>                    | 56                            | 56                           | 56                          | 56                         | 56                            | 56                           | 56                            | 56                           | 90                            |
| CHEPICAL FORMULA | K 1 K 1 2R 4 TI 4 TI 4 TI 4 | K I K I SN 4 TI 4 TI 4 TI 4 | K 1 K 1 SN 4 HF 4 HF 4 HF 4 | K ] K ] TE 4 HF 4 HF 4 HF 4 | K 1 K 1 HF 4 TI 4 TI 4 TI 4 | K 1 K 1 TH 4 TI 4 TI 4 TI 6 | K 1 K 1 TH 4 2R 4 2R 4 2R 4 | K I K I TH 4 SN 4 SN 4 SN 4 | K 1 K 1 TH 4 HF 4 HF 4 HF 4 | <b>RB 1 RB 1 TI 4 TI 4 TI 4 TI 4</b> | RB 1 FB 1 TI 4 2R 4 2R 4 2R 4 | RB 1 RB 1 TI 4 SN 4 SN 4 SN 4 | RB 1 RB 1 TI 4 HF 4 HF 4 HF 4 | R8 1 R8 1 GE 4 TI 4 TI 4 TI 4 | RB 1 RB 1 GE 4 ZR 4 ZR 4 ZR 4 | RB 1 RB 1 GE 4 SN 4 SN 4 SN 4 | RB 1 RB 1 GE 4 HF 4 HF 4 HF 4 | RB 1 RB 1 ZR 4 TI 4 TI 4 TI 4 | RB 1 RB 1 ZR 4 ZR 4 ZR 4 ZR 4 | RB 1 RB 1 2R 4 SN 4 SN 4 SN 4 | RB 1 RB 1 ZR 4 HF 4 HF 4 HF 4 | R8 1 R8 1 SN 4 TI 4 TI 4 T2 4 | RB 1 RB 1 SN 4 2R 4 2R 4 2R 4 | RB 1 RB 1 SN 4 SN 4 SN 4 SN 4 | R8 1 R8 1 SN 4 HF 4 HF 4 HF 4 | RB 1 RB 1 TE 4 TI 4 TI 4 TI 4 | RB 1 R3 1 TE 4 ZR 4 ZR 4 ZR 4 | RB 1 R3 1 TE 4 SN 4 SN 4 SN 4 | <b>RB 1 R3 1 TE 4 TE 4 TE 4 TE 4</b> | R8 1 R3 1 TE 4 HF 4 HF 4 HF 4 | RB 1 RB 1 KF 4 TI 4 TI 4 TI 4 | RB 1 RI 1 HF 4 ZR 4 ZR 4 ZR 4 | RB I RIJ I HF 4 SN 4 SN 4 SN 4 | R8 1 R13 1 HF 4 HF 4 HF 4 HF 4 | RB 1 RIJ 1 HH 4 TI 4 TI 4 TI 4 | R8 1 RIJ 1 TH 4 2R 4 ZR 4 2R 4 | RB 1 RIJ 1 H 4 SN 4 SN 4 SN 4 | RB 1 RIJ 1 TH 4 TE 4 TE 4 TE 4 | RB 1 R() 1 TH 4 HF 4 HF 4 HF 4 | SR 2 SH 2 SH 2 SC 3 SC 3 MO 6 | SR 2 SR 2 SR 2 SC 3 SC 3 15 6 | 84 2 84 2 84 2 SC 3 SC 3 MO 6 | BA 2 BA 2 BA 2 SC 3 SC 3 H 6 | 8A 2 8A 2 8A 2 Y 3 Y 3 M0 6 | 8A 2 8A 2 8A 2 Y 3 Y 3 Y 6 | BA 2 BL 2 BA 2 IN 3 IN 3 MO 6 | 8A 2 8A 2 8A 2 IN 3 IN 3 W 6 | 8A 2 8A 2 8A 2 58 3 58 3 MO 6 | BA 2 BA 2 BA 2 SB 3 SB 3 W 6 | 8A Z 8A Z 8A Z 6U 3 6U 3 MU 0 |

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|             | P FACTOR         | 2.619                        | 2.151                         | 000 · [                                                     | 2.021                        | 2.286                       | 2.317                      | 2.145                         | 2.175                        | 2.184                         | 2.219                        | 2.471                         | 60C • 7        | 20102                              | 2.976                         | 2.837                  | 2.773                         | 2.816                           | 2.017                       | 2.094                       | 2.104                       | 1.805                          | 2.210                           | 1.989<br>2.301                | 215.0                                                               | 2.341                           | 2.477                         | 2.510                         | 2.233                          | 2.008                           | 166.2                         | 020.1                                                            | 1.778                       | 2.030                       | 2.038                       | 1.762                          | 2.136                         | 1.932                         | 022.5                         | 2,232                         | 1.015                                 |                                    | 260.02                                                            | 2.410                         | 1.773                         | 2.158                         |
|-------------|------------------|------------------------------|-------------------------------|-------------------------------------------------------------|------------------------------|-----------------------------|----------------------------|-------------------------------|------------------------------|-------------------------------|------------------------------|-------------------------------|----------------|------------------------------------|-------------------------------|------------------------|-------------------------------|---------------------------------|-----------------------------|-----------------------------|-----------------------------|--------------------------------|---------------------------------|-------------------------------|---------------------------------------------------------------------|---------------------------------|-------------------------------|-------------------------------|--------------------------------|---------------------------------|-------------------------------|------------------------------------------------------------------|-----------------------------|-----------------------------|-----------------------------|--------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|---------------------------------------|------------------------------------|-------------------------------------------------------------------|-------------------------------|-------------------------------|-------------------------------|
|             | T FACTOR         | .902                         | .936                          | 896 -                                                       | .896                         | .866                        | .866                       | . 896                         | .896                         | 116.                          | 116.                         | 50.                           | . 675          | .885                               | .855                          | .877                   | • 884                         | .887                            | .958                        | .961                        | • 968                       | 619.                           | 676.                            | 006.                          | 107.                                                                | 676.                            | .952                          | .958                          | 166.                           | .969                            | • C 7 C                       | 046.                                                             | 946                         | 606 "                       | .915                        | .926                           | .878                          | • 16 •                        | .88.                          | 000.                          | . 47<br>808                           |                                    | 555 •<br>100 ·                                                    | . 906                         | .929                          | .881                          |
|             | CELL             | 8.381                        | 8.221                         | 122.0                                                       | 7.952                        | 8.098                       | 8•098                      | 7.952                         | 7.952                        | 7.885                         | 7.885                        | 8.165                         | 8.100<br>0.005 | 8.005                              | 8.152                         | 7.782                  | 7.748                         | 7.735                           | 8.121                       | 8.108                       | 8.081                       | 8.034                          | 8.434                           | 8.088                         | 112°0                                                               | 8.161                           | 8.148                         | 8.121                         | 8.241                          | 8.074                           | 8.228                         | 8.2015<br>7 00.5                                                 | 7.738                       | 7.892                       | 7.865                       | 7.818                          | 8.038                         | 7.872                         | 8.025                         | 1.998                         | (2) - 1                               |                                    | 1.118                                                             | 7_905                         | 7.805                         | 8.025                         |
| J           | B RAD            | • 85                         |                               |                                                             | .15                          | .82                         | ۰ 8 2                      | . 75                          | . 75                         | 17.                           | 12.                          | . 8.                          |                |                                    | .85                           | .72                    | .71                           | • 70                            | .72                         | • 72                        | • 10                        | • 68<br>• 5                    | 5. ·                            | 1/*                           |                                                                     | 42.                             | 12                            | .12                           | . 78                           | • 70                            | 8/.                           | • •                                                              | 49                          | .72                         | • 70                        | 。 <b>6</b> 3                   | • 19                          | . 71                          | • 78                          |                               | .0.                                   | •                                  | • • • •                                                           | .72                           | . 67                          | .78                           |
| AALENCE + 1 | A RAD            | 1.34                         | 1.34                          |                                                             | 1.20                         | 1.20                        | 1.20.                      | 1.20                          | 1.20                         | 1.20                          | 1.20                         | 1.20                          | 1.20           | 1.20                               | 1.20                          | 1.12                   | 1.12                          | 1.12                            | 1.34                        | 1.34                        | 1.34                        | 1.34                           | 1. JA                           | 1.34                          |                                                                     | 46.1                            | 1.34                          | 1.34                          | 1.34                           | 1.34                            | 1.34                          |                                                                  | 1.20                        | 1.20                        | 1.20                        | 1.20                           | 1.20                          | 1.20                          | 1.20                          | 1.20                          | 1.20                                  | 1.20                               | 1.20                                                              | 1.20                          | 1.20                          | 1.20                          |
|             | <b>BN3</b>       | 74                           | 4;                            | 53                                                          | :2                           | 42                          | 14                         | 42                            | 74                           | 42                            | 14                           | 2                             | ÷ ۲            | 44                                 | . 4                           | 202                    | 30                            | 30                              | 20                          | 4                           | 20                          | 12                             | 02                              | 0                             |                                                                     |                                 | 0 60<br>4                     | 20                            | 20                             | 30                              | 80 (<br>•                     | 00                                                               |                             | 4                           | 50                          | 12                             | 20                            | 30                            | 4<br>1                        | 20                            | 210                                   | 200                                | 0                                                                 | 05                            | 12                            | 20                            |
|             | 8N 2             | <b>64</b>                    | 2;                            | 17                                                          | 21                           | S.                          | 39                         | 49                            | 49                           | 51                            | 21                           | 5                             | <b>*</b> •     |                                    | - 10<br>- 10                  | 1<br>1<br>2            | 14                            | 73                              | 23                          | 23                          | 53                          | 4                              | 4                               | 4                             | -                                                                   |                                 | 15                            | 51                            | 73                             |                                 | 51                            | 50                                                               | , .                         | 23                          | 23                          | 14                             | 4                             | 14                            | 14<br>1                       | 41                            |                                       | 71                                 |                                                                   | 215                           | 13                            | 73                            |
| -           | BNI              | <b>64</b>                    | 21                            | ::                                                          | 23                           | 39                          | 39                         | 64                            | 49                           | 51                            | 51                           | •                             | * ;<br>0 f     | 15                                 | - 00                          | 53                     | 41                            | 73                              | 23                          | 23                          | 53                          | 41                             | 4                               | 4                             |                                                                     | <b>;</b> ;                      | 15                            | 51                            | 73                             | 23                              | 2;                            | 20                                                               | 32                          | 23                          | 23                          | 14                             | <b>!</b>                      | 14                            | 4                             | 71                            | 5                                     | 71                                 | ñ 1                                                               | 15                            | 13                            | 13                            |
| ٥           | ANB              | 56                           | <b>9</b> 5                    | 0 6                                                         | 82                           | 82                          | 82                         | 82                            | 82                           | 82                            | 82                           | 82                            | N C            | 0 0                                | 3.6                           | 38                     | 38                            | 38                              | 56                          | 26                          | 56                          | 26                             | 2                               | 9 X                           | 0 4                                                                 | 0 v<br>0                        | 2.5                           | 56                            | 56                             | 56                              | 8                             | 0                                                                | 82                          | 82                          | 82                          | 82                             | 82                            | 82                            | 82                            | 82                            | 20                                    | 20                                 | 20                                                                | 32                            | 82                            | 82                            |
|             | AN2              | 56                           | 9 i<br>20                     | 000                                                         | 82                           | 82                          | 82                         | 82                            | 82                           | 82                            | 82                           | 82                            | 200            | 20                                 | . 6                           | 38                     | 38                            | 38                              | 56                          | 56                          | 56                          | 26                             | 2                               | 90                            |                                                                     |                                 | 56                            | 56                            | 56                             | 95                              | 0 i                           | 8<br>2<br>2                                                      | 828                         | 82                          | 82                          | 82                             | 82                            | 82                            | 82                            | 82                            | 22                                    | 200                                | 2 0                                                               | 82<br>82                      | 82                            | 82                            |
| I TON AVEC  | ANI              | 56                           | 56                            | 000                                                         | 82                           | 82                          | 82                         | 82                            | 82                           | 82                            | 82                           | 82                            | 20             | 20                                 | 82                            | 38                     | 38                            | 38                              | 56                          | 56                          | 56                          | 56                             | 96                              | 202                           |                                                                     | 0 V<br>0                        | 56                            | 56                            | 56                             | 56                              | 20                            | 000                                                              | 20                          | 82                          | 82                          | 82                             | 82                            | 82                            | 28                            | 82                            | 82                                    | 28                                 | 20                                                                | 82                            | 82                            | 82                            |
|             | CHEMICAL FORMULA | 84 2 84 2 84 2 GD 3 GD 3 W 6 | BA 2 BA 2 BA 2 LU 3 LU 3 MO 6 | DA 2 DA 2 BA 2 LU 3 LU 3 W 4 0<br>DA 3 DA 3 DA 3 21 2 M 4 4 | P8 2 P8 2 P8 2 SC 3 SC 3 H 6 | PB 2 PB 2 PB 2 Y 3 Y 3 MO 6 | PB 2 PB 2 PB 2 Y 3 Y 3 W 6 | PB 2 PB 2 PB 2 IN 3 IN 3 NO 6 | P8 2 P3 2 P8 2 IN 3 IN 3 W 6 | P8 2 P8 2 P8 2 58 3 58 3 MG 6 | P8 2 P8 2 P8 2 58 3 58 3 W 6 | P8 % P8 2 P8 2 G0 3 G0 3 M0 6 |                | PR 2 PR 2 PR 2 11 3 11 3 14 2 14 0 | PB 2 PR 2 PR 2 PI 3 BI 3 WU 6 | SR 2 SR 2 V 5 V 5 CA 2 | SR 2 SH 2 SR 2 NB 5 NB 5 ZN 2 | • SR 2 SR 2 SR 2 TA 5 TA 5 ZN 2 | 8A 2 8A 2 8A 2 Y 5 V 5 CA 2 | BA 2 BA 2 B4 2 V 5 V 5 CD 2 | 84 2 84 2 64 2 V 5 V 5 SN 2 | .BA 2 BA 2 BA 2 NB 5 NB 5 MG 2 | . 64 2 64 2 64 2 NB 3 NB 3 CA 2 | 84 2 84 2 84 2 NB 5 NB 5 2N 2 | 04 C 04 C 04 C 10 J 10 J 10 Z<br>64 J C4 J 64 J 10 K 10 K 10 K 10 J | 84 2 84 2 84 2 88 3 88 5 64 2 8 | 8A 2 8A 2 8A 2 58 5 58 5 CD 2 | BA 2 BA 2 BA 2 SB 5 SB 5 SN 2 | ~8A 2 8A 2 8A 2 7/ 5 TA 5 CA 2 | - BA 2 BA 2 BA 2 TA 5 TA 5 2N 2 | 84 2 84 2 84 2 14 5 14 5 CU 2 | 84 2 64 2 84 2 14 5 14 5 84 2 89 2 89 2 89 2 89 2 89 2 89 2 89 2 | PR 2 PR 2 PR 2 V 5 V 5 2N 2 | PB 2 PB 2 PB 2 V 5 V 5 CD 2 | PB 2 PB 2 PB 2 V 5 V 5 SN 2 | -PB 2 PB 2 PB 2 NB 5 NB 5 MG 2 | PB 2 PB 2 PB 2 NB 5 NB 5 CA 2 | PB 2 PB 2 PB 2 NB 5 NB 5 2N 2 | PB 2 P8 2 P8 2 NB 5 NB 5 CU 2 | PB 2 PB 2 PB 2 NB 5 NB 5 SN 2 | 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 | PB Z PB Z PB Z 38 3 38 3 48 2 48 2 | P8 2 F8 2 P8 2 56 5 58 5 28 7 2 8 7 2 8 5 58 5 58 5 2 8 2 8 5 7 3 | PA 2 PR 2 PB 2 28 5 28 5 2N 2 | PR 2 PB 2 PB 2 TA 5 TA 5 NG 2 | PB 2 PB 2 PE 2 TA 5 TA 5 CA 2 |

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TOTAL & CATION VALENCE = 12

TOTAL A CATION VALENCE

| 323         |                  | 1.950<br>1.950<br>2.262                                                                         |
|-------------|------------------|-------------------------------------------------------------------------------------------------|
|             | T CALTOO         | - 750-0<br>- 917<br>- 884<br>- 889                                                              |
|             |                  | 7.858<br>8.012<br>7.985                                                                         |
| 12          | B RAD            | • 70<br>• 78<br>• 76                                                                            |
| VALENCE = ] | A RAD            | 1.20<br>1.20<br>1.20                                                                            |
| CATION      | 8N3              | 5 4 M                                                                                           |
| OTAL B      | BN 2             | 73<br>73                                                                                        |
| Ē           | BN1              | 73<br>73                                                                                        |
| Ŷ           | AN3              | 82<br>82<br>32                                                                                  |
| ENCE =      | ANZ              | 82<br>82<br>82                                                                                  |
| LICN VALE   | ANI              | 82<br>82<br>82                                                                                  |
| TOTAL A CA' | CHEMICAL FORMULA | PB 2 PB 2 PB 2 TA 5 TA 5 ZN 2<br>PB 2 PB 2 PB 2 TA 5 TA 5 CD 2<br>PB 2 PB 2 PB 2 TA 5 TA 5 SN 2 |

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| 24         | P FACTOR<br>2.166<br>2.931<br>2.931<br>2.936<br>2.936<br>2.936<br>2.618<br>2.618<br>2.633<br>2.575                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
|------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| ŝ          | <pre>FACTOR     -907     -907     -889     -885     -903     -904     -871     -871     -871     -871     -871     -871     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -874     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97     -97</pre>                                                                                                                                                                                                                                                                                                                                 |
|            | CELL<br>7.746<br>7.746<br>7.746<br>7.813<br>7.813<br>7.746<br>7.719<br>7.719<br>7.753<br>7.753<br>7.859                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |
| 6          | 8 RAD<br>• 67<br>• 69<br>• 72<br>• 72<br>• 78<br>• 78<br>• 78<br>• 75<br>• 75                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |
| VALENCE =  | A RAD<br>1.14<br>1.14<br>1.14<br>1.14<br>1.14<br>1.14<br>1.14<br>1.1                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |
| CATION     | 8<br>8<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9<br>9                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |
| OTAL B     | BN2<br>BN2<br>1220<br>4222<br>220<br>4222<br>220<br>4222<br>220<br>2220<br>2                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
| -          | BN1<br>22<br>30<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52<br>52                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |
| 6          | ANS 52 52 52 52 52 52 52 52 52 52 52 52 52                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |
| ENCE =     | AN2<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55<br>55                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |
| ATIGN VALI | NA<br>122<br>122<br>122<br>122<br>122<br>122<br>122<br>122<br>122<br>12                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |
|            | CHEMICAL FORMULA<br>LA 3 LA 2 LA 3 MG 2 MG 2 MG 3<br>LA 3 LA 2 LA 3 LA 2 NZ 2 V 5<br>LA 3 LA 3 LA 3 ZN 2 ZN 2 NB 5<br>LA 3 LA 3 LA 3 ZN 2 ZN 2 NB 5<br>LA 3 LA 3 LA 3 ZN 2 ZN 2 NB 5<br>LA 3 LA 3 LA 3 ZN 2 ZN 2 NB 5<br>LA 3 LA 3 LA 3 ZN 4 ZN 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 3 LA 3 ZR 4 ZR 4 LI 1<br>A 3 LA 4 LA 4 LI 1<br>A 4 LA 4 LI 1<br>A 4 LA 4 LI 1<br>A |

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Fig. 1 Experimental Schematic

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Fig. 2 Unfinished KTN Crystals Photographed in Crossed Polarized Light

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Before Etching

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| 99A  | 143B | 143E | 101A |
|------|------|------|------|
| 100D | 139C | 143K | 106D |
| 106G | 139B | 145B | 143F |
| 106I | 139A | 142C | 145H |



After Etching for Strain Relief

Fig. 3

Selected KTN Crystals Photographed in Crossed Polarized Light'

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TABLE I

Dimensions and Curie Temperatures for KTN Crystals

|                   |           |               | ΠΩ                    | nensions in |           |              |
|-------------------|-----------|---------------|-----------------------|-------------|-----------|--------------|
|                   | Optically | Curie Temp. C | <i>k</i> <sub>1</sub> |             | \$        | Aspect Ratio |
| Crystal No.       | Finished  | Visual        | (aperture)            | Ł2          | (optical) | 22 41        |
| <b>V66</b>        |           | 17-20         | 4.64                  | 5.55        | 3.70      | 0.80         |
| 100D              |           | 24-25         | 4, 90                 | 6.82        | 5.07      | 1.03         |
| VIOI              |           | 21-23         | 4.45                  | 6.72        | 6. 85     | 1.54         |
| 106D              | ×         | 20-22         | 4.59                  | 5.84        | 4.74      | 1.03         |
| 106G              |           | 3-5           | 4.83                  | 5.36        | 3.91      | 0.81         |
| 1061              | X         | 16-18         | 5.29                  | 5.33        | 2.73      | 0.51         |
| 139 <b>A</b>      | X         | 16-19         | 6.01                  | 9.50        | 10.07     | 1.68         |
| 139B              | X         | 19-21         | 6.12                  | 6.88        | 7.18      | 1. 17        |
| 139C              | ×         | 14-17         | 5.87                  | 6.35        | 9. 25     | 1.58         |
| 142C <sup>1</sup> | ×         | 9-11          | 6, 90                 | 7.67        | 6. 17     | 0.89         |
| 143B              |           | 13-15         | 5.38                  | 6.00        | 1.67      | 0.31         |
| 143E              |           | 14-15         | 6.01                  | 7.29        | 4. 97     | 0.83         |
| 143F              | X (AR)    | 13-15         | 4.74                  | 5.86        | 6.73      | 1. 42        |
| 143K              | X         | 18-19         | 5.37                  | 5.97        | 3.00      | 0.56         |
| 145B              | X         | 13-15         | 5,04                  | 9.31        | 10. 15    | 2.01         |
| 145H              |           | 17-20         | 6.37                  | 11.91       | 5.40      | 0.85         |
|                   |           |               |                       |             |           |              |

<sup>1</sup>Crystal delivered to Wright-Patterson AFB.

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Electropptic Patterns and Extinction Ratios - Crystal 142-C

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## Crossed Polarizers

Parallel Polarizers

Parallel Polarizers with HV-AC Signal Applied to Crystal



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1000V After . 25 sec.

Transmission Photographs of KTN Crystal NF-II-142-C With D-C Electric Field Applied (Parallel Polarizers 5461Å)

Fig. 6

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Fig. 7 Electrooptic Patterns at 0.546 and 1.06µ (KTN Crystal NF-II-143F Antireflection coated for 1.06µ)

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For Loss Modulation: Brewster polarizer 45° to E-field. Electrooptic switch normally open, closes for induced quarter-wave retardation.

Fig. 8 Experimental Arrangement for In-Cavity Pulsing of Nd:YAG Laser With KTN Electrooptic Switch

## **Electrooptic Patterns**

Drive Less Than Full Wave On at Origin Only KTN NF-II-143F Drive to One Full Wave On at Origin and Full Wave ł





Temporal Patterns and 1 KH<sub>2</sub> Drive Signal





Fig. 9Electrooptic Pulsing of Nd:YAG LaserLoss Modulation with Sinusoidally Driven KTN Retarder



| Upper | Trace: | Pulse Mode Locked Output  |
|-------|--------|---------------------------|
| Lower | Trace: | Sinusoidal Drive at 2 KH, |

Fig. 10

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Electrooptic Pulsing of Nd:YAG Laser Phase Modulation With Sinusoidally Driven KTN Retarder Pulse Mode Locked Output