

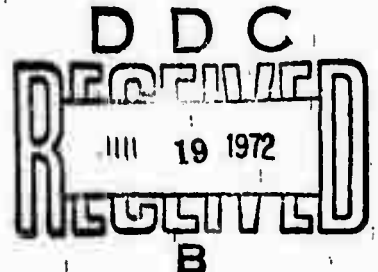
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PREPARATION AND CHARACTERIZATION OF POLYCRYSTALLINE  
HALIDES FOR USE IN HIGH POWER LASER WINDOWS

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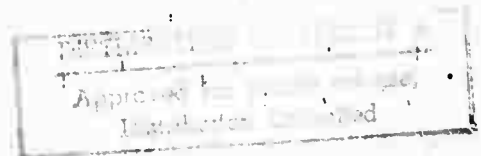
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| <p>Emphasis during the first quarter has been on hot working techniques to produce fully dense polycrystalline KCl from single crystals. Equipment for press forging and extrusion has been set up and is fully operational. A number of press forgings and extrusions have been made and crack free transparent polycrystalline samples have been produced by press forging. Most of the extrusions produced to date were cloudy.</p> <p>Equipment necessary to evaluate the optical properties of polycrystalline halides especially under laser irradiation and external stress, has been constructed and is operational. This includes a calorimeter for absorption coefficient measurements and a holographic interferometric facility for detecting changes in optical and structural homogeneity.</p> |  |   |                 |

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## I. INTRODUCTION

The purpose of this program is to define methods of producing polycrystalline halide materials with improved mechanical properties while maintaining the desirable optical properties of the single crystal materials. In the first quarter of this contract we have emphasized material-processing techniques and the development of test facilities to evaluate the optical and mechanical properties of the materials produced. The materials processing techniques investigated to date are hot press forging and hot extrusion of KCl single crystals. We have succeeded in preparing polycrystalline materials that appear to approach single crystals in optical quality and that have improved mechanical properties. With respect to evaluation techniques we have constructed both conventional and holographic interferometers to study the effect of heating on the optical properties of the materials and have made some preliminary tests using this equipment. In addition, a calorimeter has been built and used to measure the absorption coefficient of a sodium chloride sample.

## II. PROCESSING STUDIES

Two methods of strengthening KCl single crystals have been employed to date, press forging and extrusion. Both techniques rely on recrystallization of the hot worked crystal billet as the strengthening mechanism and are discussed below.

### A. Press Forging

We are examining the effects of both strain rate and working temperature on the structure and properties of large vacuum press forged KCl single crystals. Strain rates ranging from  $5 \times 10^{-3}$  to  $5 \times 10^{-2} \text{ min}^{-1}$  and temperatures ranging from 166 to  $600^{\circ}\text{C}$  have been employed. Cylindrical single crystal billets 1.5 or 2 inches in diameter have been used. All pressing has been along {100}.

Some data and comments concerning these tests are given in Table 1. The strain rates quoted in the Table are engineering strain rates, R/h, where

R is the ram velocity and h is the height of the billet. All pressings except number 29 were carried out at constant ram speed and, therefore, at a continually increasing strain rate. During some press forgings the final load on the press was noted. The stresses given in the Table were calculated from these loads and the final areas of the forging.

As evident in Table 1, we are tending toward lower working temperatures to obtain a finer grain size in the forgings. Metallographic characterization of the grain structure of the forgings is currently in progress and no comments can be made at present concerning either the grain size or homogeneity of the material. We have some evidence, however, that in some cases we are not getting recrystallization during forging. In tests number 12, and 28, for instance, the forgings had a square shape and contained patterns of straight cracks  $90^\circ$  to each other quite reminiscent of {100} cleavage cracks in a single crystal. It is possible even at the relatively high temperature of  $300^\circ\text{C}$  that  $\langle 110 \rangle$ {110} slip was predominant and resulted in the inhomogeneous deformation leading to the final forged shape.

As noted in the comments in Table 1 many of the forgings cracked. The fractures originated from a number of causes, most of which we feel can be controlled. First, when working at constant ram speed, the increasing strain rate leads to work hardening and a flow stress which might exceed the fracture stress of the forging. This can be controlled by pressing at a constant strain rate as done in forging number 29. Second, it is important to have flat and polished platens. Both excessive friction and flaws in the platens can initiate cracks. Third, any internal cracks in the crystal (e.g., resulting from cleavage) can propagate during forging. This was the case in numbers 21, 25, 27, and 29. We had been water polishing the last few billets, but evidently not to the extent required.

To date our most successful press forgings have been made at  $300^\circ\text{C}$  at strain rates of .011 and .023  $\text{min}^{-1}$  (numbers 19 and 24). Both forgings were crack free and transparent. A photograph of forging number 24 is shown in Figure 1. The forging is slightly oval which indicates a slight inhomogeneity of deformation. This sample has been examined in polarized light

and the observed birefringence indicated the residual stresses in the forging to be quite uniform. A photograph of forging number 21 showing the radial cracks mentioned in Table 1 is included in Figure 2.

The final forging stresses listed in Table 1 decrease somewhat continuously with temperature and increase with final strain rate as expected. Within the strain rate range used, the stresses are seen to be much more sensitive to temperature than to strain rate.

The mechanical properties of one of the crack free forgings (number 19) produced at 300°C have been determined. The yield and fracture strengths were determined from 3-point bend tests on a Table Model Instron Machine using samples with nominal dimensions of 1/8 x 1/4 x 1 inch and a span of .75 inches. The beams were cut from the forging with a diamond wheel on a high speed precision surface grinder using a constant, slow feedrate and a copious amount of methyl alcohol as a coolant. The surfaces were then hand polished prior to testing. An average of three tests produced a yield strength of 1060 psi and a fracture strength of 1690 psi.

#### B. Extrusion

The extrusion technique provides a radically different method of producing polycrystalline material than press forging because the deformation is much more localized and inhomogeneous and comparisons of material produced by both techniques should be useful. The strains and strain rates are generally higher in extrusion and it is possible that, at least at high extrusion ratios, the resulting material may be more structurally homogeneous.

All extrusions produced to date were made with a 15° die with no land. The billet container was 1.5 inches in diameter and the extrusion ratio (ratio of billet cross sectional area to extrusion cross sectional area) was 16:1. Extrusions were made at three ram speeds at temperatures ranging from 215 to 400°C. Data and comments on the nine extrusions made to date are contained in Table 2. The strain rates quoted in the Table are equivalent longitudinal true strain rates defined as the true longitudinal strain

( $\ln 16$ ) times the ram speed. At  $400^{\circ}\text{C}$  no problems were encountered and the extrusion (excepting the first 3 inches) was transparent and had a good surface. Rattailing is a common extrusion defect and occurs when die friction is excessive. An example is shown in Figure 3. A photograph of an extrusion exhibiting a good surface is shown in Figure 4.

In general, extrusions produced at  $300^{\circ}$  or less were cloudy. The cloudiness increased with both increasing extrusion rate and decreasing temperature. Metallographic examinations of the extrusions are currently underway to determine the origin of the cloudiness. Most likely it results from grain boundary separation.

#### C. Platen Materials

The choice of platen materials for the press forging of halides is very important, since usually the material cools to room temperature in contact with the platen. This is a problem since alkali halides have large thermal expansion coefficients that cannot be easily matched to materials used for platens. If any reaction occurs between the platen and the halide sample sticking may result and the difference in expansion coefficients will crack the forging. We have conducted tests to determine suitable materials for use in halide forging. Platen materials we have tried include metals such as gold, nickel, platinum, steel and nonmetals such as graphite, vitreous carbon and magnesium oxide. The tests were conducted by forging small KCl samples between platens made of the test materials in a vacuum at temperatures higher than  $250^{\circ}\text{C}$ . The load was left on the material during cooling. We found that with the exception of the carbons all other materials reacted to some extent with the KCl and the resultant sticking of the KCl to the platen was sufficient to produce cracking.

Conventional graphite as well as pyrolytic graphite were found to be not dense enough for this application. In this case, the problem was not one of chemical reaction between the halide and the graphite, but rather one of mechanical locking caused by the fact that the halide is so plastic at the working temperature it extrudes into the surface voids of the

graphite. Upon cooling, the lower expansion coefficient of the graphite causes severe cracking of the KCl.

Vitreous carbon was by far the best platen material. This material is very dense and we observed no evidence of any sticking of the KCl. We have been able to successfully press forge crack free KCl with vitreous carbon platens.

In extrusion the reactivity of the halide with the die is not as serious because the material cools in air without any contact with the mold. In the case of extrusion we have been able to use steel dies sprayed with colloidal graphite to extrude materials successfully.

### III. OPTICAL EVALUATION

The three methods to be used in this program for optical evaluation are calorimetry for measuring absorption coefficients, interferometry for examining thermally induced changes in the optical properties, and microscopic examination with polarized light to determine the structure and birefringence in processed polycrystalline window materials.

#### A. Calorimetry

A calorimeter has been constructed consisting of a cylindrical container with two windows. The sample whose absorption coefficient is to be measured is held by a low conductance support. A #30 gauge copper-constantan thermocouple is attached to the sample with Dow Corning rubber cement and an identical thermocouple is attached to the wall of the brass container. The two thermocouples are operated in a differential mode and the signal is measured with a Keithley Model 153 microvolt meter. The output of the microvolt meter can be displayed as a function of time on a strip chart recorder. The sensitivity of the measurement is limited by noise at  $\pm 5$  millidegrees centigrade. This is the peak-to-peak equivalent temperature noise of the entire system consisting of thermocouple, microvolt meter and recorder. We have determined the source of this noise to be the microvolt meter since shorting its input results in the same noise level. Should it be necessary

to improve the sensitivity of this instrument, a nanovoltmeter with a lower input impedance can be used.

The absorption coefficient  $\beta$  of a commercially available spectrophotometer NaCl window has been measured using a 10 watt CO<sub>2</sub> laser operating in a TEM<sub>00</sub> mode with the calorimeter both air-filled and evacuated. The window had a square cross section of 1 inch by 5 mm thick. The measured value is  $\beta = (2.8 \pm .3) \times 10^{-3} \text{ cm}^{-1}$ . The method of determining  $\beta$  is that described by Skolnik<sup>(1)</sup>. We wish to acknowledge very useful discussions we had with Len Skolnik of AFCRL as well as personnel at the Air Force Materials Laboratory at Wright-Patterson Air Force Base concerning calorimetry.

We conclude that the calorimetric technique is a straightforward and very useful method of measuring absorption coefficients in halides and that the results obtained are fairly independent of whether the calorimeter is evacuated or filled with air.

#### B. Interferometry

In this program interferometry will be used for characterization of the homogeneity of changes in the optical thickness of polycrystalline halide samples subjected to external agents such as a laser beam or an applied stress. The primary technique of interest in this case is holographic interferometry because it allows one to make a differential measurement that gives directly the changes produced by the external agencies without regard to the surface condition of the sample. We judge this to be an important feature since alkali halides are difficult to polish to a very good surface.

We have constructed two interferometers using a Spectra-Physics Model 120, 5 milliwatt, helium-neon laser operating in the TEM<sub>00</sub> mode as a light source. The illuminated aperture is approximately 1" in diameter. Initially we constructed a conventional Michelson interferometer and used it both to examine some of the optical components to be used in the holographic interferometer as well as to establish the noise spectrum of the granite table

(1) AFCRL Quarterly Report #3, LQ10 Program Sept. 1971.

on which the interferometer was mounted. Examination of the noise spectrum showed that for photographic work the granite table was sufficiently stable, but the use of electronic sensing to examine changes in interference patterns required stabilization of the granite slab. This was accomplished by mounting the slab on inner tubes. The Michelson interferometer was also used to examine the homogeneity of readily available optical windows as well as to determine the distortion of the optical path of a quartz window when it was irradiated with the beam from a 10 watt carbon dioxide laser.

The holographic interferometer has now been completed and some preliminary work with it has been done. Although the interferometer is to be used to make transmission holograms of the alkali halide windows, reflection holograms can be made with a simple rearrangement of the components. Kodak 649F glass plates have been used and we have determined that exposures of approximately 1/40 second produce suitable density in the film using an 8 to 1 ratio of reference to object beam intensity.

Using the interferometer we have examined the deformation of the holographic interference pattern resulting from the application of a soldering iron to a 5mm thick sodium chloride window. The results are shown in Figure 5. To date we have not been able to produce any detectable alteration of the interference pattern by illuminating this window with a 10 watt CO<sub>2</sub> laser. We now have put into operation a 70 watt CO<sub>2</sub> laser and expect to be able to produce measurable changes with the higher power. Changes in the optical path in quartz windows irradiated with a 3 watt CO<sub>2</sub> laser have been observed and are shown in Figure 6.

#### IV. SUMMARY

Emphasis during the first quarter has been on hot working techniques to produce fully dense polycrystalline KCl from single crystals. Equipment for press forging and extrusion has been set up and is fully operational.

A number of press forgings and extrusions have been made and crack free transparent polycrystalline samples have been produced by press forging. Most of the extrusions produced to date were cloudy.

Equipment necessary to evaluate the optical properties of polycrystalline halides especially under laser irradiation and external stress has been constructed and is operational. This includes a calorimeter for absorption coefficient measurements and a holographic interferometric facility for detecting changes in optical and structural homogeneity.

#### V. FUTURE PLANS

Emphasis during the next quarter will be on the optimization of processing parameters to produce material with maximum strength and on the determination of the structure and the optical and mechanical properties of the material produced. Some annealing studies will be conducted and alloy crystals will be obtained. Studies of a third processing technique, hot rolling, will be initiated.

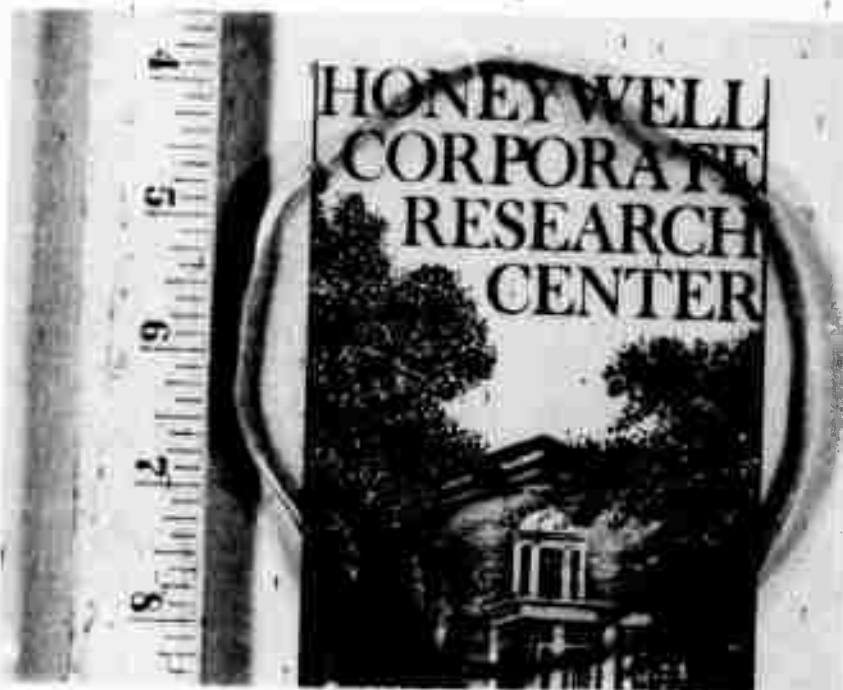
Table 1. Data and Comments on Press Forgings Made To Date

| Forging Number | Billet Height (in) | Billet Diameter (in) | Temperature (°C) | Initial Strain Rate (min <sup>-1</sup> ) | Final Strain Rate (min <sup>-1</sup> ) | % Reduction in Height | Approximate Final Stress (KPSI) | Comments  |
|----------------|--------------------|----------------------|------------------|--|--|-----------------------|---------------------------------|---|
| 6              | .372               | 2.0                  | 500              |  |  | 46                    |                                 | No cracks in forging.   |
| 10             | .370               | 2.0                  | 500              |  |  | 54                    |                                 |   |
| 11             | .380               | 2.0                  | 500              |  |  | 50                    |                                 | No cracks in forging.   |
| 12             | .316               | 2.0                  | 300              | .005                                     | .077                                   | 32                    |                                 | Apparent {100} cracks in forging suggest recrystallization did not occur.   |
| 18             | .465               | 2.0                  | 300              | .05                                      | .25                                    | 80                    |                                 |   |
| 19             | .698               | 2.0                  | 300              | .023                                     | .080                                   | 71                    |                                 | No cracks in forging.   |
| 21             | .730               | 2.0                  | .80              | .011                                     | .040                                   | 72                    |                                 | Forging had square outline and extensive radial cracks.   |
| 24             | 1.07               | 2.0                  | 280              | .011                                     | .074                                   | 80                    |                                 | No cracks in forging.   |
| 25             | 1.00               | 2.0                  | 244              | .012                                     | .056                                   | 78                    | 4.8                             |   |
| 27             | 1.12               | 2.0                  | 196              | .007                                     | .033                                   | 78                    | 7.0                             | Water polished billet. Three cracks in forging.   |
| 28             | 1.06               | 2.0                  | 166              | .006                                     | .027                                   | 79                    | 10.6                            | Water polished billet. One corner of billet contained a cleavage crack. Forging had square outline and was cracked in one corner. |
| 29             | 1.57               | 2.0                  | 194              | .011*                                    | .011                                   | 87                    | 5.2                             | Water polished billet. One crack in forging.  |

\* Strain rate maintained constant.

Table 2. Data and Comments on Extrusions Made to Date

| Extrusion Number | Billet Length (in) | Temperature (°C)  | Equivalent True Longitudinal Strain Rate (min <sup>-1</sup> ) | Comments   |
|------------------|--------------------|-------------------|---|--|
| 14               | 1.0                | 400               |   | First 3 inches of extrusion rat tailed   |
| 15               | 1.0                | 310               | 1.51  | Total extrusion rat tailed.  |
| 16               | 1.0                | 300               | .22<br>.71<br>1.51  | Three strain rates used. Extrusion cloudy. Cloudiness increased with strain rate.  |
| 17               | 1.0                | 250               | .22<br>.71<br>1.51  | Three strain rates used. Total extrusion rat tailed.   |
| 20               | 1.0                | 300               | .078  | Extrusion had smooth surface. Slightly cloudy.   |
| 22               | 1.5                | 280               | .078  | Extrusion had smooth surface. Slightly cloudy.   |
| 23               | 1.5                | 255               | .078  | Extrusion had smooth surface. Slightly cloudy.   |
| 24a.             | 1.5                | 240<br>250<br>260 | .078  | Three temperatures used. Started at lowest T. Extrusion rat tailed at all T.   |
| 26               | 1.5                | 260 to<br>216     | .078  | Temperature continually decreased. Extrusion was smooth until last 2 inches rat tailed. Cloudiness increased as T decreased. |

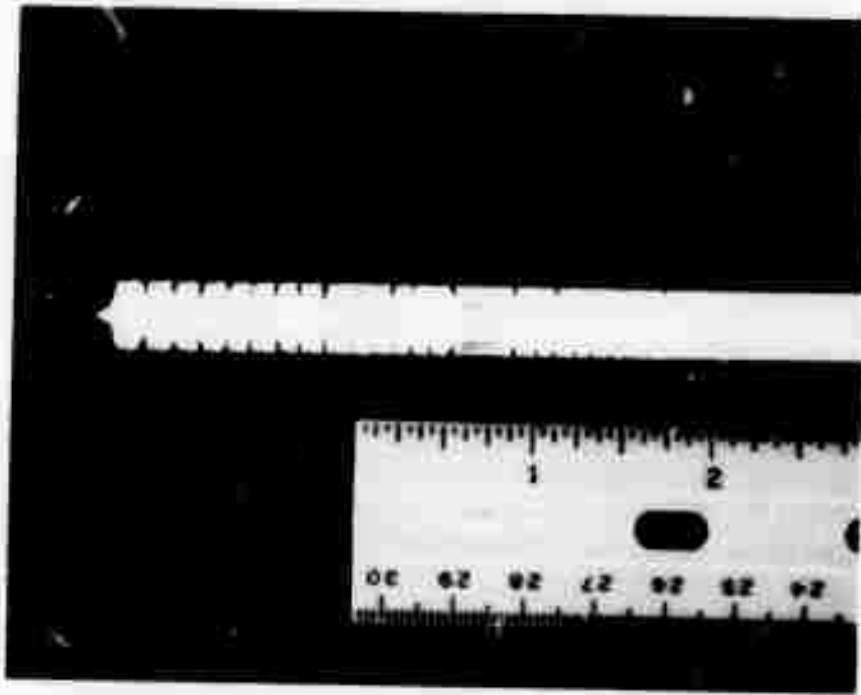


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Figure 1. Photograph showing a successful crack free transparent KCl press forging. Forging was carried out at an initial strain rate of  $.011 \text{ min}^{-1}$  and at a temperature of  $280^{\circ}\text{C}$ .



Figure 2. Photograph showing a KCl press forging containing radial cracks. The irregularities at the outside edge of the forging indicate the cracks may have originated from surface flaws in the initial billet. Forging was carried out at an initial strain rate of  $.011 \text{ min}^{-1}$  and at a temperature of  $190^{\circ}\text{C}$ .



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Figure 3. Photograph showing a typical extrusion resulting from excessive die friction. (Extrusion number 1. See Table 1.)

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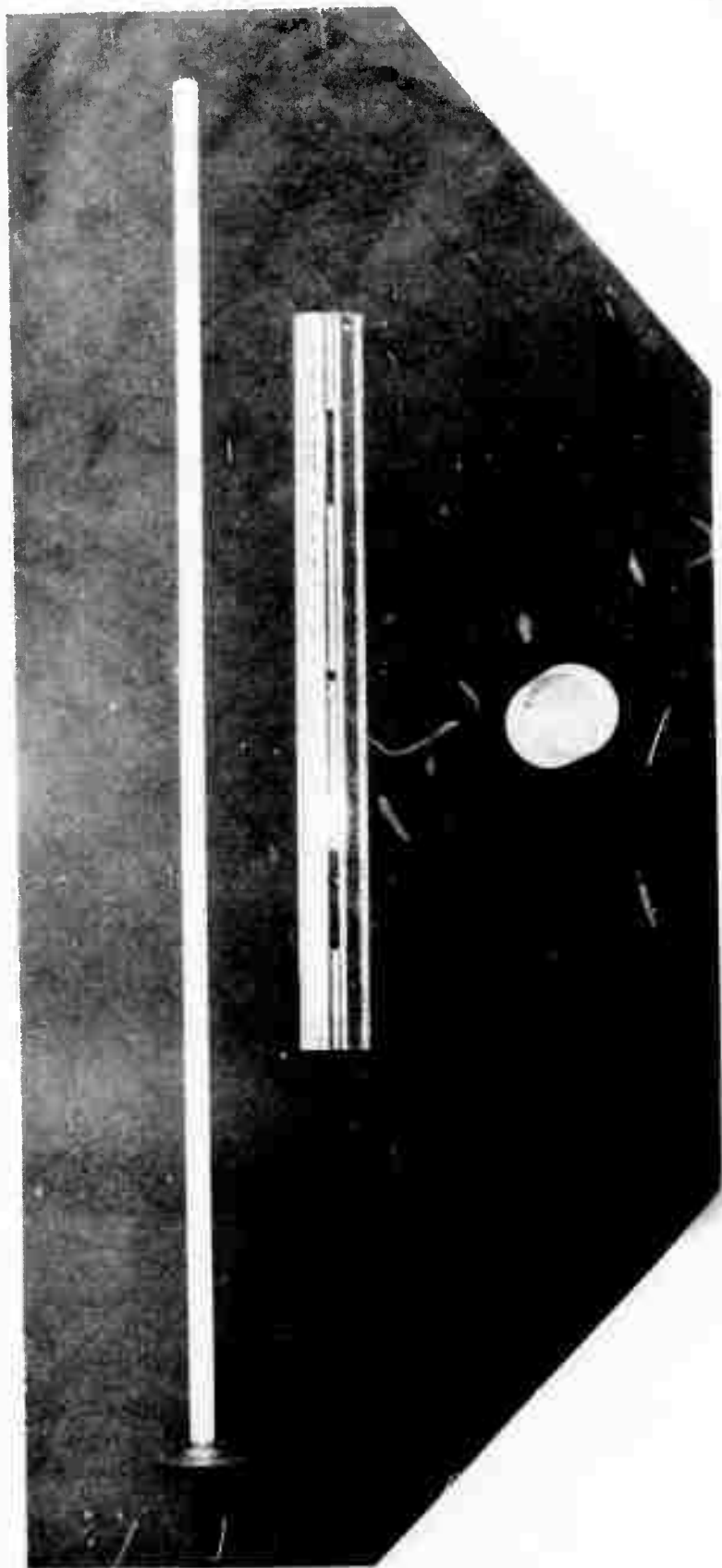
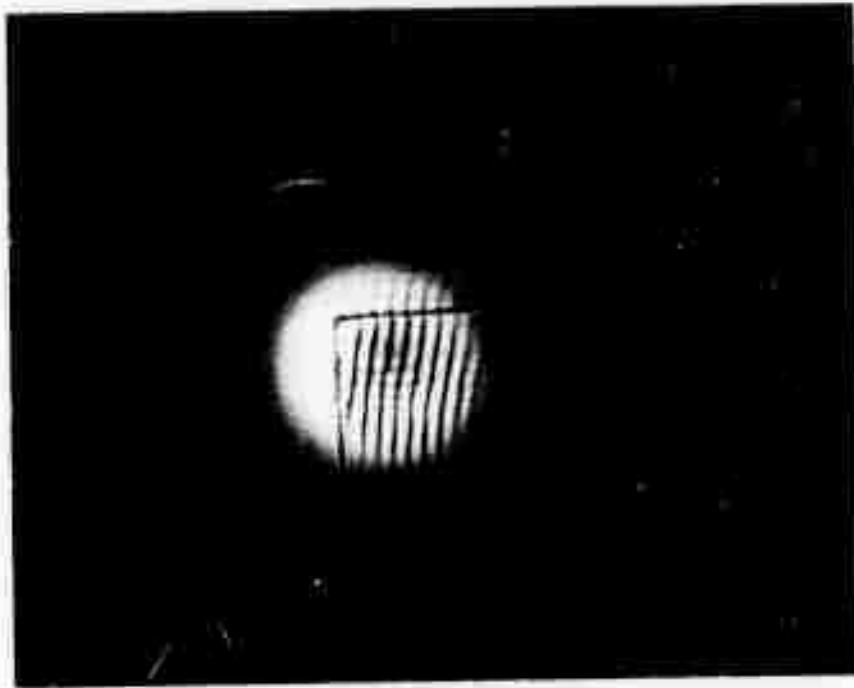
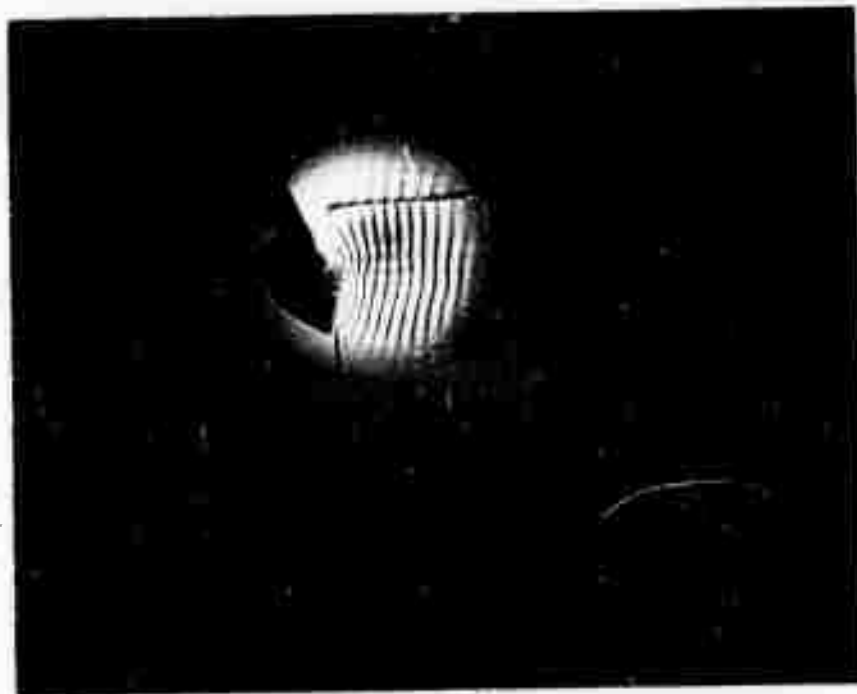


Figure 4. Photomicro showing an extrusion having a good surface finish. (Extrusion number 23. See Table 1.) The extrusion is somewhat cloudy. (But identical to one used for extrusion also shown.)



(a)



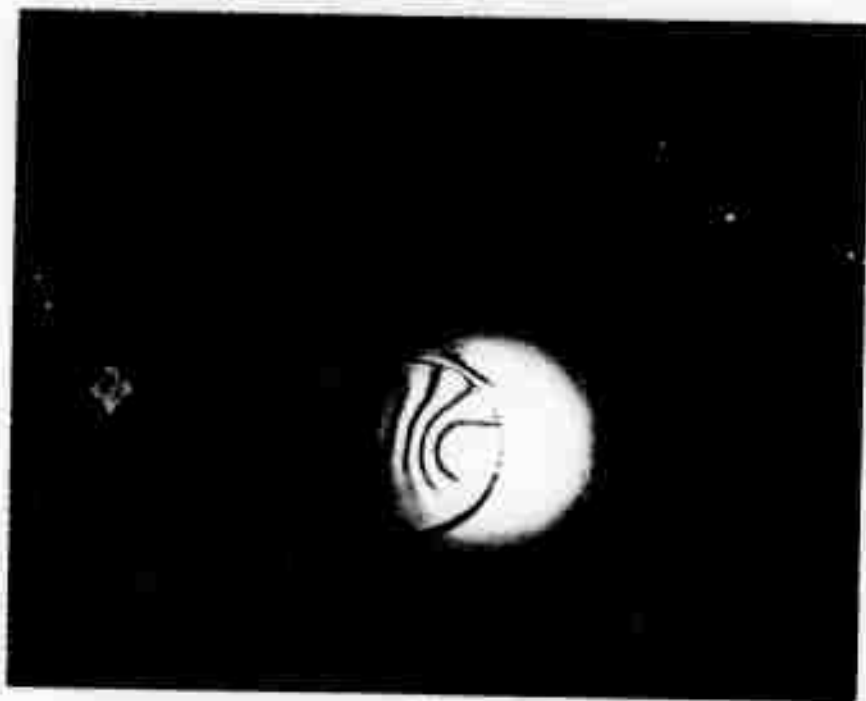
(b)

Figure 5. Changes produced in holographic interferogram of NaCl window when heated with a soldering iron. a) original interferogram; b) interferogram with soldering iron on window.

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(a)



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(b)

Figure 6. Changes produced in interferogram of quartz window when heated near edge with a 3 watt  $\text{CO}_2$  laser. a) original interferogram; b) interferogram after 30 sec. exposure to laser.