Final Report

Contract NAS8-30618

Covering Period March 15, 1974 - December 15, 1975

for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
GEORGE C. MARSHALL SPACE FLIGHT CENTER
MARSHALL SPACE FLIGHT CENTER, ALABAMA 35812

ANALYTICAL AND EXPERIMENTAL EVALUATION OF TECHNIQUES FOR THE FABRICATION OF THERMOPLASTIC HOLOGRAM STORAGE DEVICES

Principal Investigator - Jerry W. Rogers
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INTRODUCTION

Thermoplastic media was first shown to be suitable for holographic recording by Urbach and Meier.[1] Since that time several researchers have published more detailed data which identify the most pertinent characteristics. [2,3,4] The published favorable characteristics are as follows:

1. Thermoplastic has a sensitivity comparable to Kodak 649 F high resolution film.
2. Thermoplastic has adequate resolution for most applications. (1000 cycles/mm or better)
3. Thermoplastic has a bandpass spatial frequency response which aids in suppressing intermodulation distortion.
4. Development of thermoplastic can be done *in situ*.
5. Readout of thermoplastic recording is efficient and non-destructive.
6. Thermoplastic records are erasable and the medium is reusable.
7. Thermoplastic recording is relatively immune to ambient illumination as well as substrate reflections.

Unfavorable characteristics of thermoplastic media are as follows:

1. The recording apparatus is complex.
2. Thermoplastic records have a random frost noise.
3. Thermoplastic records should be made in a clean environment. The surface of the recording degrades very quickly in a dust or lint-filled room.
4. The charge distribution over the recording surface if proportional to the signal, but the deformation is proportional to

the square of the charge distribution; thus, intermodulation frequencies are produced.

5. The necessity of heating for recording and cooling for storage limits thermoplastic to less than real time operation.

Of course, it should be noted that not all of the desirable or undesirable characteristics are necessarily germane to every application.

The object of this research report is to relate the result of an experimental investigation of recording on thermoplastic. A description of a typical fabrication configuration as well as a recording sequence is given. A detailed description is given of the samples which were examined.

There are basically three configurations which can be used for the recording of information on thermoplastic and each will be described. The most popular technique uses corona which furnishes free charge. The necessary energy for deformation is derived from a charge layer atop the thermoplastic. A description of this technique will be included for completeness. The other two techniques simply use a dc potential in place of the corona for deformation energy. A description of all is included.

THERMOPLASTIC HOLOGRAPHY USING CORONA

Device Configuration and Recording Technique

A hologram which is recorded on thermoplastic film is a surface relief phase grating where the optical thickness variation corresponds to the impinging light intensity pattern. A transparent "sandwich" is configured as shown in Figure 1 where the layers consist of a glass substrate, transparent conductor, photoconductor, and thermoplastic.

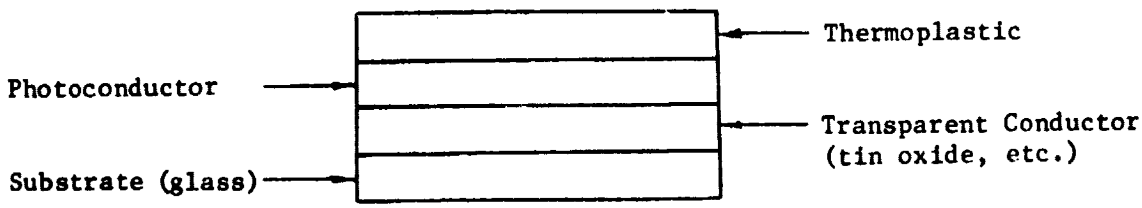
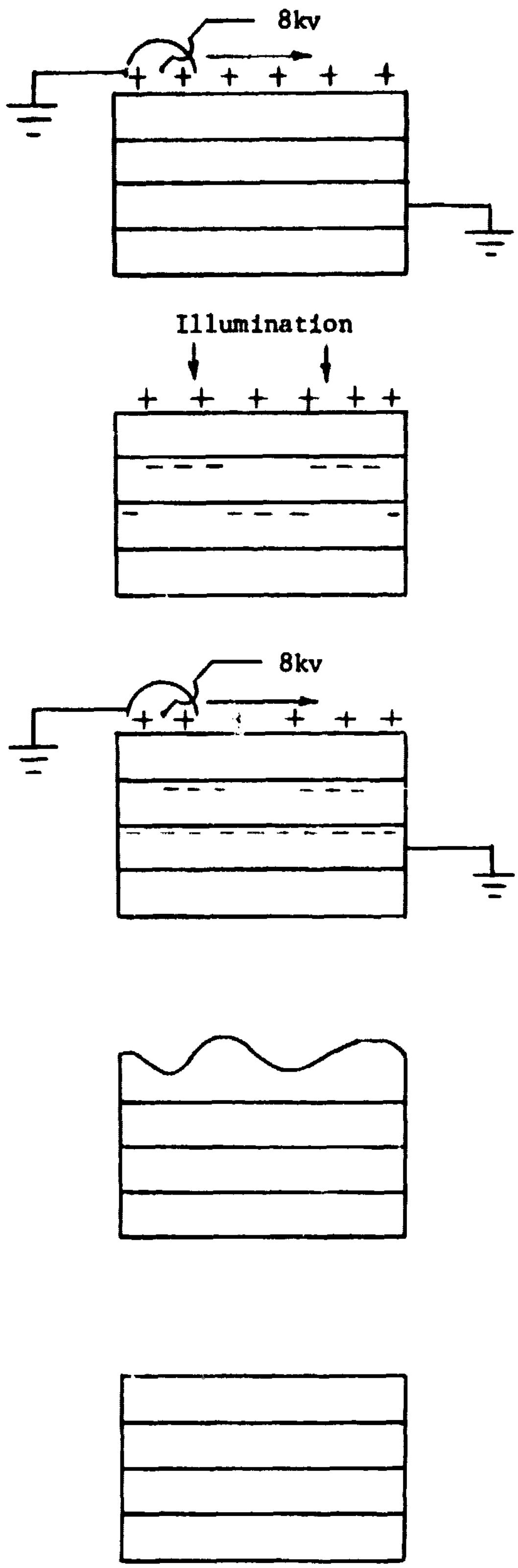


Figure 1. Thermoplastic Film Structure

The recording process itself can best be described by referring to Figure 2. First, a uniform charge is established on the thermoplastic layer by passing a corona device over the surface as illustrated in step 1.

Next, the thermoplastic is exposed to spatial intensity variations of light. These variations correspond, according to some prescribed format, to the information one wishes to record. At the high irradiance locals of the spatial variations, charge will migrate across the photoconductor whereas at the dark local areas, the charge remain static.



Step 1
First Charging

Step 2
Exposure

Step 3
Second Charging

Step 4
Heat Develop

Step 5
Heat Erasure

Figure 2. Recording Process

The final result of this event is a spatial potential variation over the thermoplastic face which corresponds to the illumination variations.

Step 3 consists of passing the corona device over the surface as was done in step 1. The thermoplastic surface again becomes an equipotential surface, but the local electric field across the thickness of the thermoplastic ceases to be a constant. At high local irradiance the electric field across thermoplastic thickness is high and correspondingly low at low irradiance locals.

Heating is accomplished as shown in step 4 by passing a current through the transparent electrode (tin oxide, etc.). Resistive heating raises the temperature of the thermoplastic to its melting point whereupon the electric field variations are translated into "hills and valleys". The molten thermoplastic will be thinner in high electric field areas and thicker otherwise. The intensity variation (information) is stored as a surface relief phase grating. Lowering the temperature of the thermoplastic will "freeze" this pattern for later perusal.

Finally, erasure can be accomplished by simply raising the temperature again to the melting point of the thermoplastic, and the surface tension will cause the thermoplastic surface to smooth, thereby, yielding a film suitable for reuse.

Thermoplastic Film Fabrication

A 2" x 2" optically smooth glass plate was selected as the substrate, and tin oxide as the transparent conductor. These plates were commercially available.

The electrode configuration was produced by applying silver paint over a mask. The electrodes were subsequently baked for one hour at 100° C. The electrodes furnish a conductor access to recording squares.

The substrate with its silver electrodes was then dipped for the photoconductor. The first trial was made using dioxane and dichloromethane as photoconductor solvents; however, the results were not satisfactory. In normal building conditions, the photoconductor would "fog". The technique then was simply to dry them rapidly with warm air such as furnished by a hot air dryer. Later, the solvent was changed to tetrahydrafuron and trinitrofluorenone which did not have "fogging" tendencies. The photoconductor used was polyvinyl carbazole sensitized with 2, 4, 7 trinitrofluorenone at a molar ratio of 8:1. Tetrahydrafuron was used as a solvent and adjusted such that a 2.0 μm. thickness resulted from being pulled at 2 cm/min.

Finally, after a few minutes of air dry, the "sandwich" was immersed in thermoplastic and drawn at a rate of 10 cm/min which yielded a thickness of 1.0 μm. The thermoplastic was natural tree resin where 12 1/2 grams was dissolved in 50 ml of naptha.

Experimental Recording Configuration

The recording arrangement for both the recording and reconstruction is shown in Figure 3.

The recording was made by heating the thermoplastic with the corona device in place. Reconstruction was accomplished by removing the corona device and covering up the information beam (i.e., the transparency). A test pattern of several images was made and reconstructed.

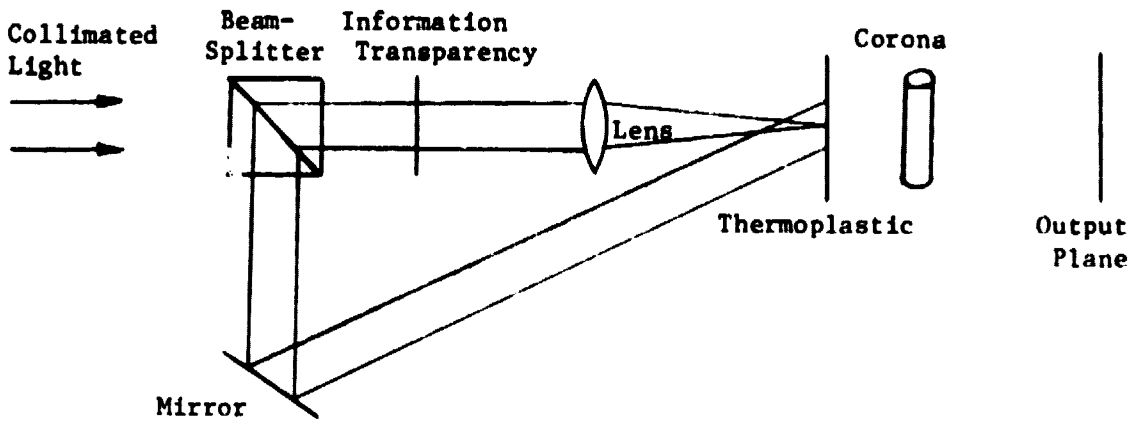


Figure 3. Recording and Reconstruction Arrangement

The image quality demonstrated very graphically bandpass characteristics of thermoplastic.

A typical slide transparency was also recorded and reconstructed in order to test the procedure as well as the subjective image quality. Diffraction efficiency was in the order of 6% corresponding to that reported by others. Each recording pad was cycled 5 - 10 times with no appreciable surface degradation. The heating of the first ones was done by hand; that is, the voltage of the heater supply was raised and lowered by hand. It was evident that serious degradation of adjacent records resulted from this crude procedure; however, a pulsing power supply completely eliminated this problem.

As cycling was extended, the thermoplastic "wears out." The tendency was for it to retain "shadows" of past images. This phenomena has been observed by other researchers, too. The reason for this is not clear, but it is felt to be a result of surface damage done by the surface charge and the high local fields. Because of this, a technique for recording without using corona was explored.

Two different configurations can be devised to produce thermoplastic deformation without using corona. One uses parallel plates and the field produced by an applied potential between the plates as energy source for deformation. An alternate "sandwich" configuration which obviates the corona device requires a transparent conductor coating atop the thermoplastic. The coating thus placed would serve to produce a constant potential surface. A power supply electrically connected to the conductive coating would furnish the energy needed for the electrostatic deformation. Using this arrangement, the surface relief pattern would hold as long as the voltage was present or would "freeze" at cooling. In addition, it can be noted that conductor transparency is not necessary if a reflection hologram is desired. The first method mentioned will be considered next in more detail.

THERMOPLASTIC HOLOGRAPHIC RECORDING WITHOUT CORONA

Theoretical Analysis

Consider first the case of two parallel conducting plates. Assume that these plates are separated by d meters and the volume between the plates is occupied by two dielectrics as shown in Figure 4. A difference of potential, V, exists between the two plates.

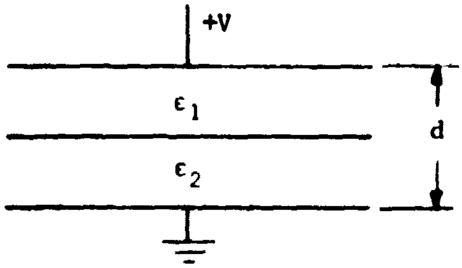


Figure 4. Two Dielectrics Between Parallel Plates

Assume too that the surface area of the plates are large such that the field vectors are normal to the boundary surface. As a result of a pressure at the boundary, dielectric ϵ_2 moves into the volume occupied by dielectric ϵ_1 (virtual displacement). Through the use of virtual change in the energy located on the field resulting from the virtual displacement, the following relationship can be formed,

$$\delta W_e = \left(1/2 \frac{D_2^{\,2}}{\epsilon_1} - 1/2 \frac{D_2^{\,2}}{\epsilon_2}\right) \Delta x \;\delta x \tag{1}$$

where

δW_e = virtual change in energy

Δs = small part of boundary surface

δx = virtual displacement

From equation (1), the pressure at the surface is found to be

$$P = -\frac{1}{\Delta s}\frac{\delta W_e}{\delta x} = 1/2\left(\frac{1}{\varepsilon_1} - \frac{1}{\varepsilon_2}\right)D_n^{\,2} \qquad (2)$$

The configuration shown in Figure 4 and the resulting equation (2) will form the basis for a slightly more complex configuration.

Consider the arrangement shown in Figure 5 where

d = plate separation (Glass substate with tin oxide coating)

a = thermoplastic plus photoconductor thickness

c = photoconductor thickness

h = the thermoplastic deformation

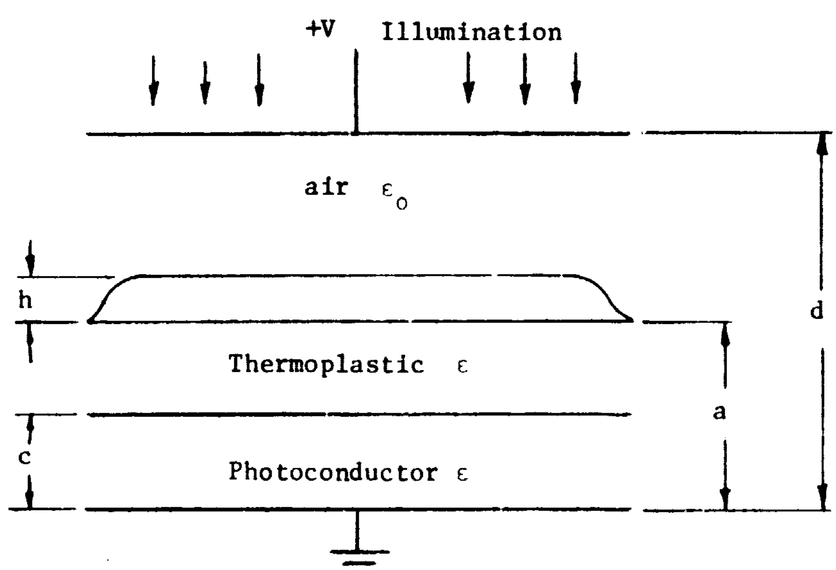


Figure 5. Optical Recording Device

While there is no illumination, the photoconductor is a dielectric and for the purposes of this analysis assumed to be approximately equal to the thermoplastic dielectric constant, ε. During the time it is illuminated from some external source it becomes a conductor. When these assumptions and conditions are combined with equation (2), then

$$P = 1/2(\varepsilon - \varepsilon_o) \frac{\varepsilon_o}{\varepsilon} E^2 \qquad (3)$$

where ε = thermoplastic dielectric constant

ε_o = dielectric constant of free space

E = electric field in the air

The dielectric will move upward by h, and will rise until counter-balanced by weith· f the liquid above the surrounding level. If ρ_m is the mass density, g is the gravitational field, then

$$P = \rho_m h g \qquad (4)$$

There is one assumption that will initially be made and that is that surface tension does not materially affect the results. A bit more will be discussed about this later.

Equation (3) and (4) can be combined to form the equation

$$h = \frac{1}{\rho_m g} \frac{1}{2} (\varepsilon - \varepsilon_o) \frac{\varepsilon_o}{\varepsilon} E^2 \qquad (5)$$

which relates the height of deformation of the thermoplastic to the characteristics and applied field.

Consider the case where a checkered grid of light impinges upon the thermoplastic and photoconductor "sandwich," where the geometry is shown in Figure 5. A cross section reveals that the conductor becomes square wave of height, c, because the photoconductor becomes a conductor where light is present.

Thus, in the area where light is present,

$$V = E[d - (a - c) - h] + E'[a - c + h] \tag{6}$$

where ε_o is the electric field in air and E' is the electric field in the thermoplastic.

Since $D_{n_1} = D_{n_2}$, then

$$E' = \frac{\varepsilon_o}{\varepsilon} E \tag{7}$$

When equation (7) is combined with equation (6) and, the resulting solution for E becomes

$$E = \frac{V\ \varepsilon}{\varepsilon(d - a + c - h) + \varepsilon_o(a - c + h)} \tag{8}$$

Equation 8 relates the electric field to applied voltage and system parameters. Equation (8) and (5) combined furnished the final relationship,

$$h = \frac{1}{2\rho_m g} (\varepsilon - \varepsilon_o) \frac{\varepsilon_o}{\varepsilon} \left[\frac{V\ \varepsilon}{\varepsilon(d - a + c - h) + \varepsilon_o(a - c + h)}\right]^2 \tag{9}$$

Equation (9) can be formed into a third order equation in h and is

$$h^3(\varepsilon_r - 1)^2 + h^2[2(1 - \varepsilon_r)(u\ \varepsilon_r + w] + h[\varepsilon_r u + w]^2$$

$$- \frac{1}{2\rho_m g} \varepsilon_o \varepsilon_r (\varepsilon_r - 1)V^2 = 0 \tag{10}$$

where $u = d - a + c$

$w = a - c$

If one assigns typical value of physical parameter, then equation (10) can be solved as a third order algebraic equation. In order that numerical results can be investigated, assume that the system parameters are as follows:

$c = 1 \times 10^{-6}$ meters

$d = 1 \times 10^{-3}$ meters

$a = 3 \times 10^{-6}$ meters

$g = 9.81$ M/sec^2

$\rho_m = 1115$ KG/m^3

$\varepsilon_r = 1.5$

$\varepsilon_o = 8.854 \times 10^{-12}$ farads/meter

Let h_1 be the height of deformation with the sandwich illuminated (c = 0) and let h_2 be the height of the deformation without illumination. The relative deformation normalized to red light (6328Å) is

$$\text{Rel. Def.} = \frac{h_1 - h_2}{6.328 \times 10^{-7}} \qquad (11)$$

Figure 6 is the result of solving equation (10) for applied voltage from 50 to 500 volts in steps of 50 volts for two typical values of d.

Observation of the values of relative deformation shows that the light wave front is phase modulated and is in fact a record of the spatial variation of the light pattern.

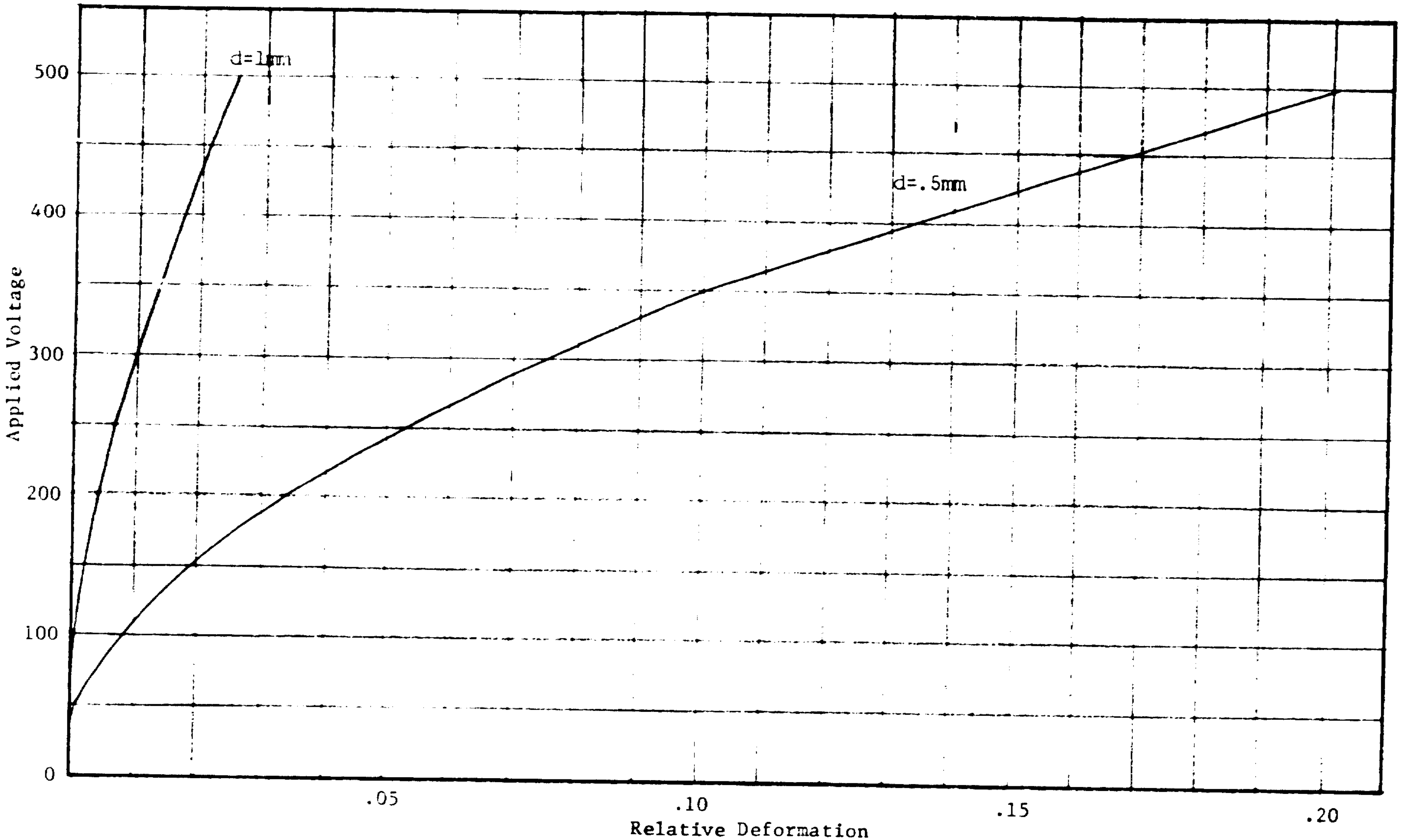


Figure 6. Relative Deformation of Thermoplastic Vs. Applied Voltage

It was mentioned earlier that surface tension was neglected for the purposes of the calculation. If, however, one wishes to consider its effects, then the differential equation,

$$T_o \frac{d^2h}{dx^2} = P(x) + C_o \tag{12}$$

can be shown to relate deformation, h, to pressure, p(x), where T_o is the undisturbed surface tension, and x is measured along the plane of the thermoplastic surface.

Fabrication

The procedure used for the construction of this device is similar to the one described previously. In this case, however, a second 2" x 2" tin oxide coated glass substrate was affixed to a support which placed it approximately 1 mm from the sandwich as shown in Figure 7.

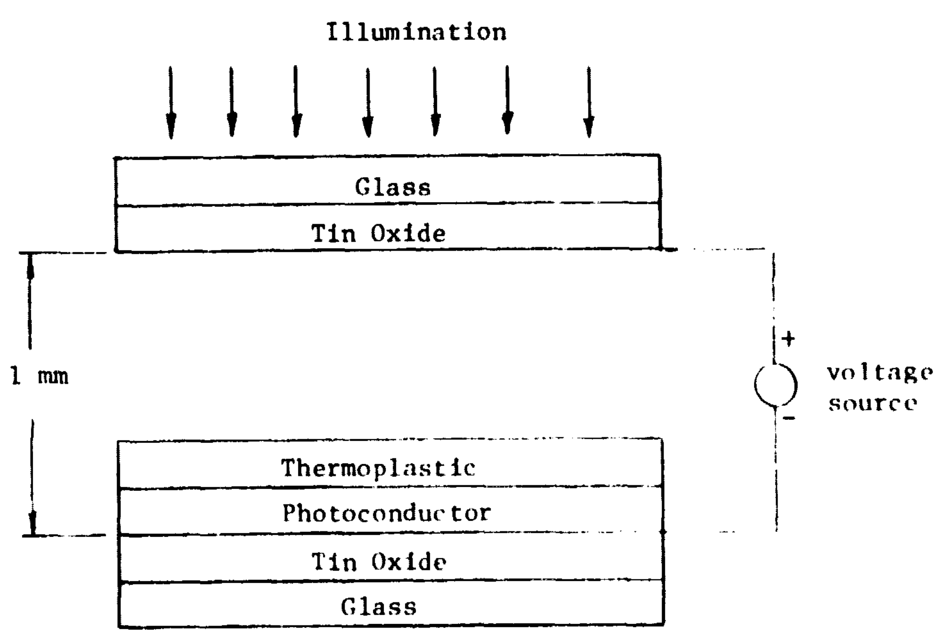


Figure 7. Optical Recording Device Configuration A

The application of the photoconductor and thermoplastic to the glass substrate was the same as used for the corona device and the silver electrodes served to "spot heat" recording squares. The recording equipment configuration was essentially the same as used with the corona device shown in Figure 3.

Many attempts to obtain reasonably good recordings were made. It is evident from Figure 6 that voltage should be in the order of 500 for the spacing of 1 mm plate separation; thus, most trials were made with 300 or more volts. A major problem encountered was that of arcing from one plate to the other at the silver electrode locations. The spacing in these areas were obviously less than 1 mm and were expected to be the critical places. Unfortunately not any of the recordings were good quality.

Any attempts to insulate the two plates with a transparent dielectric were not successful. There was always the passing of optical information from glass-air-dielectric-air-thermoplastic, etc. with the injection of a dielectric. Due to the many changes in dielectrics, the information was degraded arising primarily from front surface effects at each boundary.

The most probable solution to the above described problems would be to make other electrode arrangements and evacuate the space between the two plates.

Another configuration which received attention is shown in Figure 8. The basic structure of the glass-tin oxide-silver electrodes-photoconductor-thermoplastic was the same as used previously. The thin conductive coating is the only difference. The purpose of the conducting coating was to

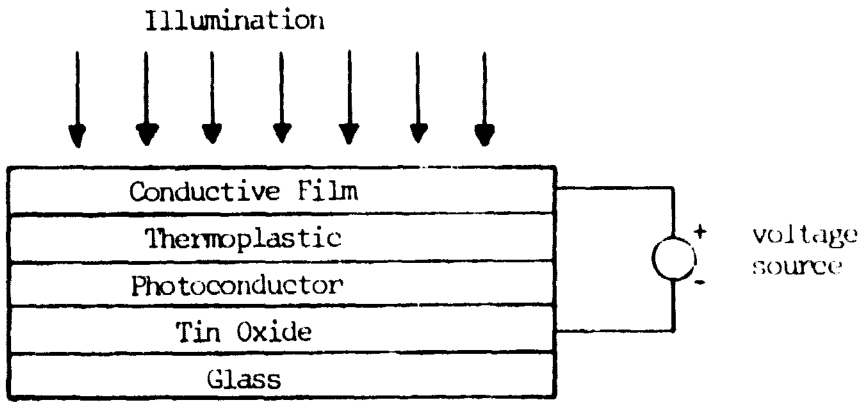


Figure 8. Optical Recording Device Configuration B

produce a constant potential surface over the face of the thermoplastic. This coating was placed on the thermoplastic surface using vacumm evaporation technique. Aluminum was chosen for the conducting coating because it is ductile, conducting and easy to evaporate.

Two cases were examined. First, the coating was thin so as to be transparent. This results in a transmission type recording. Second, the thickness was increased so as to be essentially opaque. This results in a reflection type recording. The recording equipment configuration was the same as the two previous cases.

A mechanical bond was determined to be unattractive so the connection to the aluminum coating was made with mercury. Again the problem of voltage isolation across the thermoplastic was a problem. The evaporation process quite often produced an aluminum connection to the tin oxide. The probable cause was that the environment for evaporation was hot enough to make the thermoplastic liquid, and the aluminum vapor impregnated the thermoplastic at electrode position so as to create a "weak spot."

RESULTS AND CONCLUSIONS

The lack of good quality reproduction of optically recorded information on the non-corona devices was disappointing. However, several things were learned in the experimental phase which should be of some value. First, it would be desirable to construct a simpler arrangement for testing the calculated results. The calculated values were assumed to be reasonable and the device construction went directly to a recording arrangement. A preliminary step where one actually observed and measured thermoplastic deformation without the photoconductor would have been extremely helpful for furnishing recording device design constraints. It would have, for example, helped determine the extent of the effects of thermoplastic surface tension. A second experiment where one used the photoconductor and observed the effects with and without illumination (no spatial variations) and the subsequent charges in thermoplastic deformation would likewise furnished valuable information for device construction.

Another area which requires care is the plate support structure and the minimizing of electrode irregularities. This can be done by using photographic mask and thick film technology.

It is not clear what overall effects will occur where the plates are not within certain tolerances of being parallel. Since the thickness of the photoconductor is approximately 1×10^{-6} to 3×10^{-6} meters, it is expected that the tolerances on the parallel separation is close; however, it should be pointed out that the recording squares are very small and would not be subject to large electric field variation over the recording square.

Arcing was a prevalent problem all through the experiments. Two recommendations are made. First, evacuate the volume between the plate and minimize the electrode runs between the plate. The second recommendation is to look for an alternate recording medium to replace the thermoplastic. For example, a high compliance transparent elastomer would serve. An elastomer would not, however, be able to "freeze" the information in a phase relief and a holding voltage would be necessary. It is also uncertain whether a holding voltage would retain the optical information recording for an extended period.

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