RADIATION PROTECTION STUDY FOR PHOTOGRAPHIC SPACE FILM

FINAL TECHNICAL REPORT

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SPACE DIVISION CHRYSLER CORPORATION

RADIATION PROTECTION STUDY

FOR

PHOTOGRAPHIC SPACE FILM

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I. ABSTRACT

A Cobalt-60 source has been used to irradiate several types of space photographic film. From the data obtained, characteristic curves have been drawn for these films and a proton-gamma equivalence determined. The effects of variations in the development processing parameters have been investigated and the feasibility of reduced film storage temperature has been studied as a means to inhibit film fogging in space.

II. INTRODUCTION

The analytical and experimental work described in this report was sponsored by the National Aeronautical and Space Administration under contract NASW-1741. The original purpose of this study was to determine the effect of gamma fogging exposure on several types of Kodak film, and to use this data to determine uncertainties in film density due to variations in processing parameters and to determine if the effects of protons on film could be simulated by gamma rays. After the study had been in progress for some time, it was decided to divert a part of the work scope into an investigation of the effects of exposure temperature on film fogging due to gamma irradiation.

This study was motivated by the desire to establish a solid experimental basis in order to determine the optimum means to protect film in the space environment.

At the time the proposal for this contract was written, all of the films listed for study were being considered for possible use in the ATM mission. At present, due to the expected fogging in space from nuclear radiation, only the least sensitive films will be used.

The Cobalt-60 exposures were performed at the Ochsner Foundation Hospital in New Orleans. Eastman Kodak Company has donated considerable technical assistance to us throughout the program, particularly on the cross-calibration study, and we gratefully acknowledge this courtesy.

III. TECHNICAL STUDIES

III.A. GAMMA EXPOSURE

All Chrysler exposures throughout the study were made with a Picker V4M60 Cobalt-60 teletherapy apparatus. This equipment is located at Ochsner Foundation Hospital in New Orleans, and is part of the hospital's radiation therapy unit. Use of this facility by Chrysler Corporation was arranged on a per-hour rental basis. Source strength as of September 1968 was 3340 curies.

1. Experimental Procedure

Those films strips not requiring special handling were enclosed in double-envelope light tight paper containers and placed between 0.5 gm/cm^2 layers of plexiglass, which served as electron equilibrium layers of air equivalent material.

Special handling was required for the ultraviolet films -- types SWR, 101-01 and SC-5 -- which are very sensitive to pressure. For the SWR film, strips of masking tape were placed along the inside of the plexiglass slabs and used as rails to support the outer edge of the bare film. This prevented the central portion of the strips from coming in contact with the slabs. A light tight envelope was then placed around the plexiglass-film combination. The 101-01 film is manufactured with beads already imbedded along the sides to prevent the emulsion side of the film from touching a flat object placed over the film, but extra caution was necessary in handling the film and placing it in the envelope. The SC-5 comes prepackaged in a container with raised lids, so there was no special problem with this film.

Although there was no discernible backscatter, a plexiglass plate was used for all films as a convenient back support for the strip. The entire film strip package was placed on a hospital therapy bed and the head of the cobalt unit adjusted for a 100 centimeter source-to-film distance (SFD).

The size of the gamma field is adjustable for the source unit and was maintained at 7x20 cm. for all tests. All exposures were performed on 35 mm x 12 inch film strips, except for the SC-5 film, which is delivered precut at a 35 mm x 7 inch size. Film strips were centered in the gamma field. Exposure dose was checked periodically throughout the program with a Victoreen r-meter, calibrated in roentgens. The conversion from roentgen to rads in air absorbed dose for Co-60 is 0.877 rad per roentgen. Rads in air was used as the basic unit throughout the study in order to have a common unit to correlate with other reports.

In general, the films were processed in accordance with Table III.A.1, which lists the processing parameter values recommended by the film manufacturer, Eastman Kodak Company. All processing was done in deep tanks with manual agitation provided by moving the film hangers. The films were usually developed the day after exposure. All development work and density readings were performed by the same technician throughout the program. Unexposed strips of each type of film were included in each processing to determine background density and to serve as a processing control. Background fog of the SWR and 101-01 films was reduced as recommended by a presoak of two minutes in distilled water at 68° F.

Film density was measured manually with a Macbeth Quantalog TD-102 Densitometer. Accuracy over the entire scale is 0.02 density units. Each density value recorded was an average of three readings taken along the center line of the long axis of the film, using a 3 mm aperture for the densitometer. A calibrated sensitometric strip was read before each series of observations to insure consistency of the densitometry reading throughout the experiment.

2. Cross-Calibration Procedure

A cross-calibration experiment was performed through the courtesy of Eastman Kodak Company to obtain a film control which could be used throughout the remainder of the program. The film selected was Kodak Plus-X, Aerial Film, Type 3401 (Estar Thin Base). Chrysler exposures were made under the same conditions as normally used for all tests (i.e., 100 cm. SFD with 7x20 cm field). Kodak exposures were performed with a 0.71 curie Cobalt-60 source and a 25 cm SFD. On the same day, Kodak Research Laboratory and Chrysler Corporation Space Division each exposed strips of 3401 film to three different levels of Cobalt-60 radiation, previously agreed on by the two companies. All of the film strips were from the same production batch. Three samples for each exposure level irradiated by CCSD were processed and evaluated by Kodak[1] and the same procedure was repeated by CCSD for films irradiated at Kodak. The experimental results obtained by the two laboratories are illustrated in Figures III.A.1 and III.A.2. The shape of the curve of Figure III.A.2 was drawn from Sr-90 and Y-90 β-ray sensitometric exposures, while the densities obtained from the Cobalt-60 exposures were used to position the curve properly along the exposure axis.

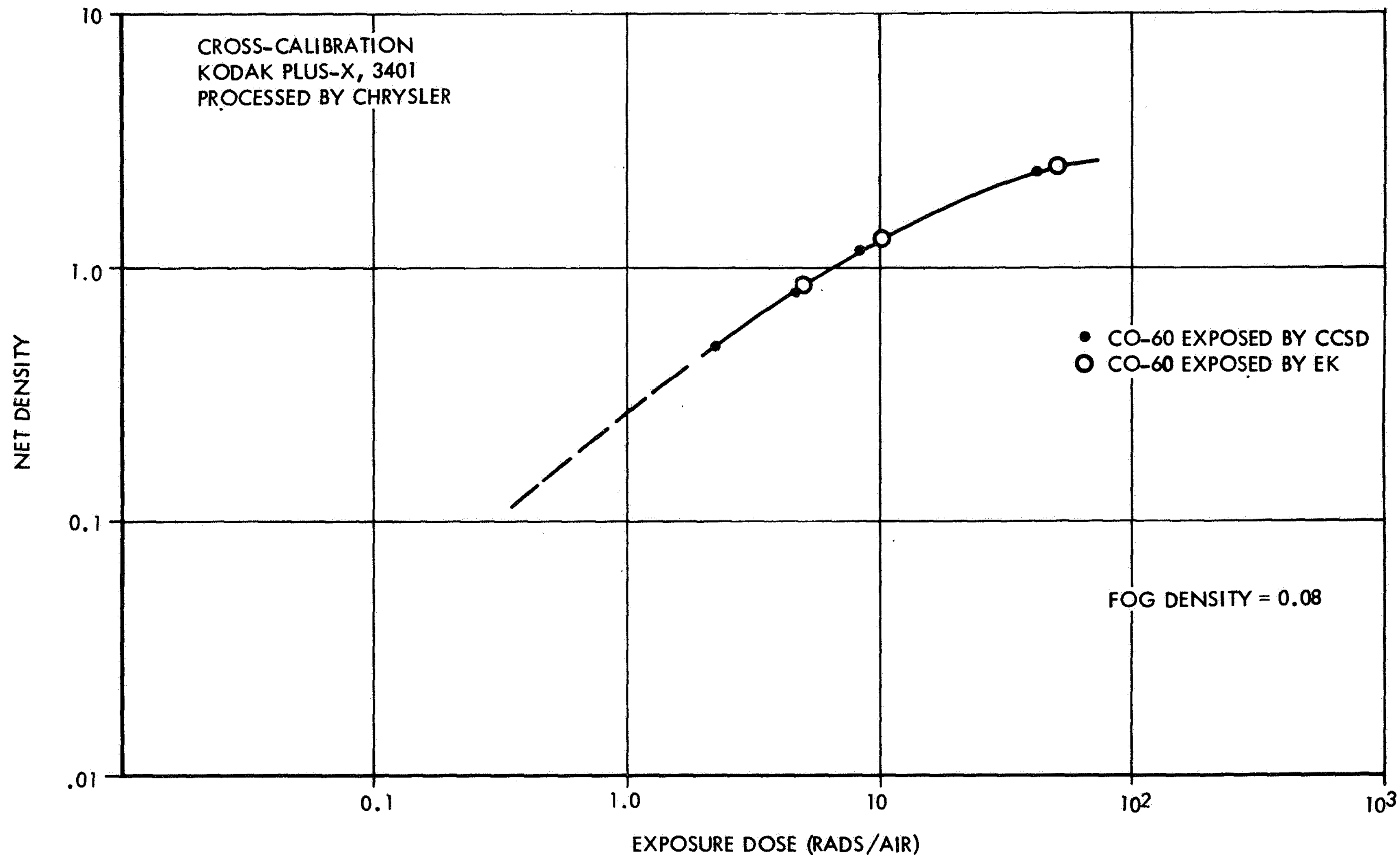

Figure III. A. 1

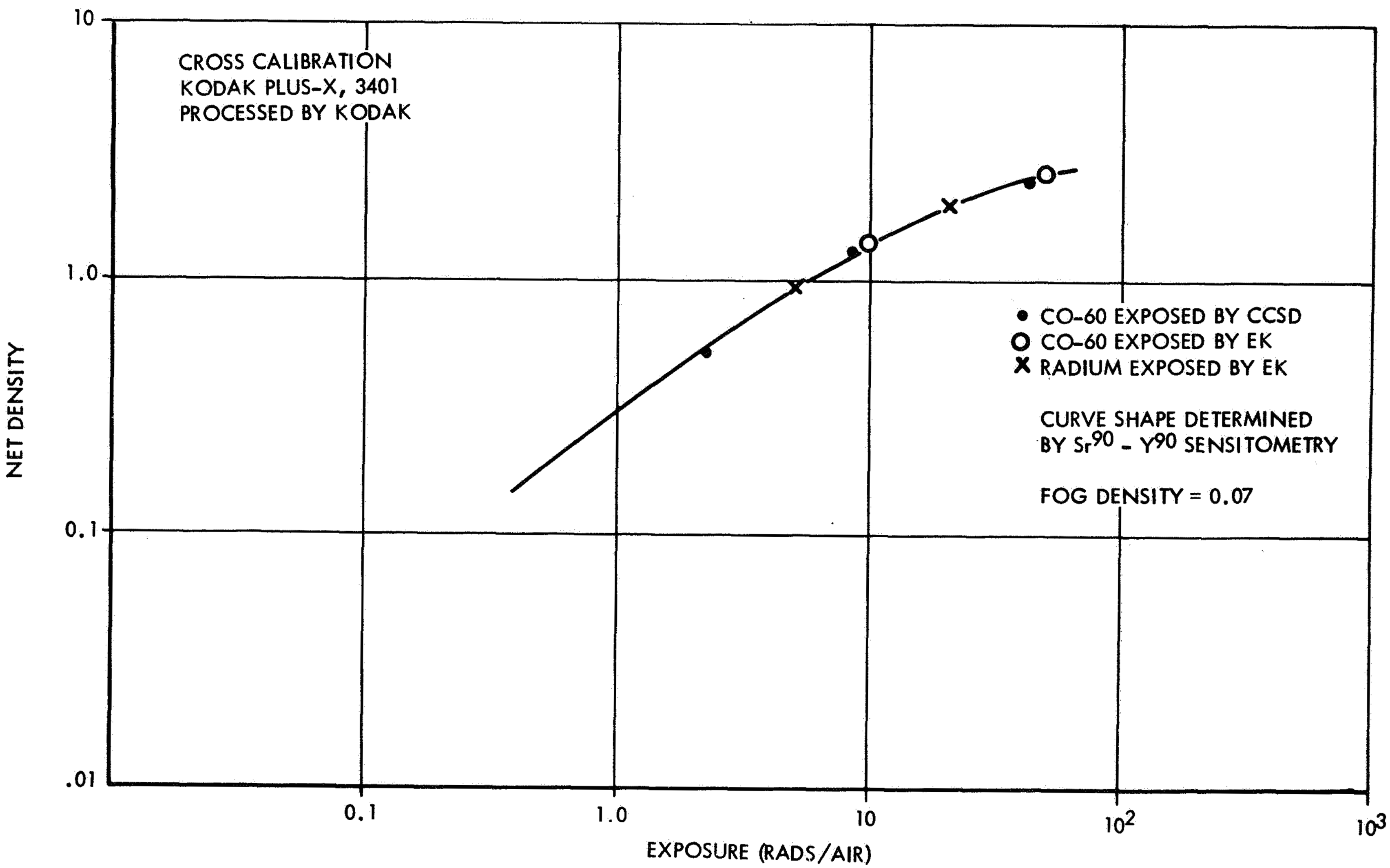

Figure III. A. 2

Table III.A.1

TYPE AND PROCESSING CONDITIONS FOR GAMMA IRRADIATED FILMS

Film	Kodak Developer	Time	Temp.
1. Kodak Spectroscopic Film, Type 103-0	D-19	4 min.	68°F
2. Kodak Spectroscopic Film, Type 103-a0	D-19	4 min.	68°F
3. Kodak Plus-X Aerial Film, Type 3401 (Estar Thin Base)	D-19	8 min.	68°F
4. Kodak Panatomic-X Aerial Film, Type 3400	D-19	8 min.	68°F
5. Kodak Special Film, Type 101-01	D-19 (1:1)	2 min.	68°F
6. Kodak SWR Film	D-19	2 min.	68°F
7. Kodak SC-5 Film	D-19	2 min.	68°F
8. Kodak SO-392 Film	D-19	8 min.	68°F
9. Kodak Special Solar Recording Film Type SO-375	D-19	8 min.	68°F

3). Characteristic Curves for Gamma Radiation

The responses of the film types listed in Table III.A.1 to Cobalt-60 gamma radiation are shown in Figures III.A.3 and III.A.4, where net density is plotted as a function of absorbed dose in rads/air.

A comparison of these results with those of the Eastman study[2] shows excellent agreement for all film types except the ultraviolet. CCSD density values are generally lower for this type of film, ranging from approximately 10 percent for the SWR and SC-5 film to a factor of 2 or 3 for the 101-01. The reason for this discrepancy is not known, but should be investigated further since it could lead to the use of the more sensitive 101-01 film on flights such as ATM.

It can be seen that the characteristic curves for all but the three last sensitive films (SWR, SO-392 and SO-375) have approximately the same shape, but their position along the dose axis is dependent on the film type.

The most sensitive films to gamma radiation are the 103-0 and 3401 types. The least sensitive film is the SO-375. There is approximately two orders of magnitude difference between the gamma dose required to produce a net density of 0.2 in the least sensitive film (SO-375) and the most sensitive film (103-0).

The SO-392 film, which is Eastman Kodak's replacement for their SO-375 film, was approximately 2.3 times more sensitive to gamma radiation than the SO-375 for a net density of 0.2, when both are processed as described in Table III. A.1.

4.) Proton-Gamma Correlation

We wish to compare the gamma response of the film listed in Table No. III.A.1 to the proton response. The purpose is to determine if the radiation fogging produced by a proton beam can be simulated by exposure to gamma rays. Cobalt-60 radiation with its reproducibility and its defined spectrum provide an appropriate reference standard.

Eastman Kodak Company supplied us with the proton irradiation data[2], which were obtained during the summer of 1967. Our correlation study is based upon these proton experiments, in which Cobalt-60 gamma exposures served as a film control. The film used by us for the Cobalt-60 exposures are the same film type but different emulsion batches.

In studying the effects of various types of nuclear radiation on photographic film, it has generally been found that an absorbed dose of one type of radiation (e.g., gamma) will not produce the same effect as the same absorbed dose

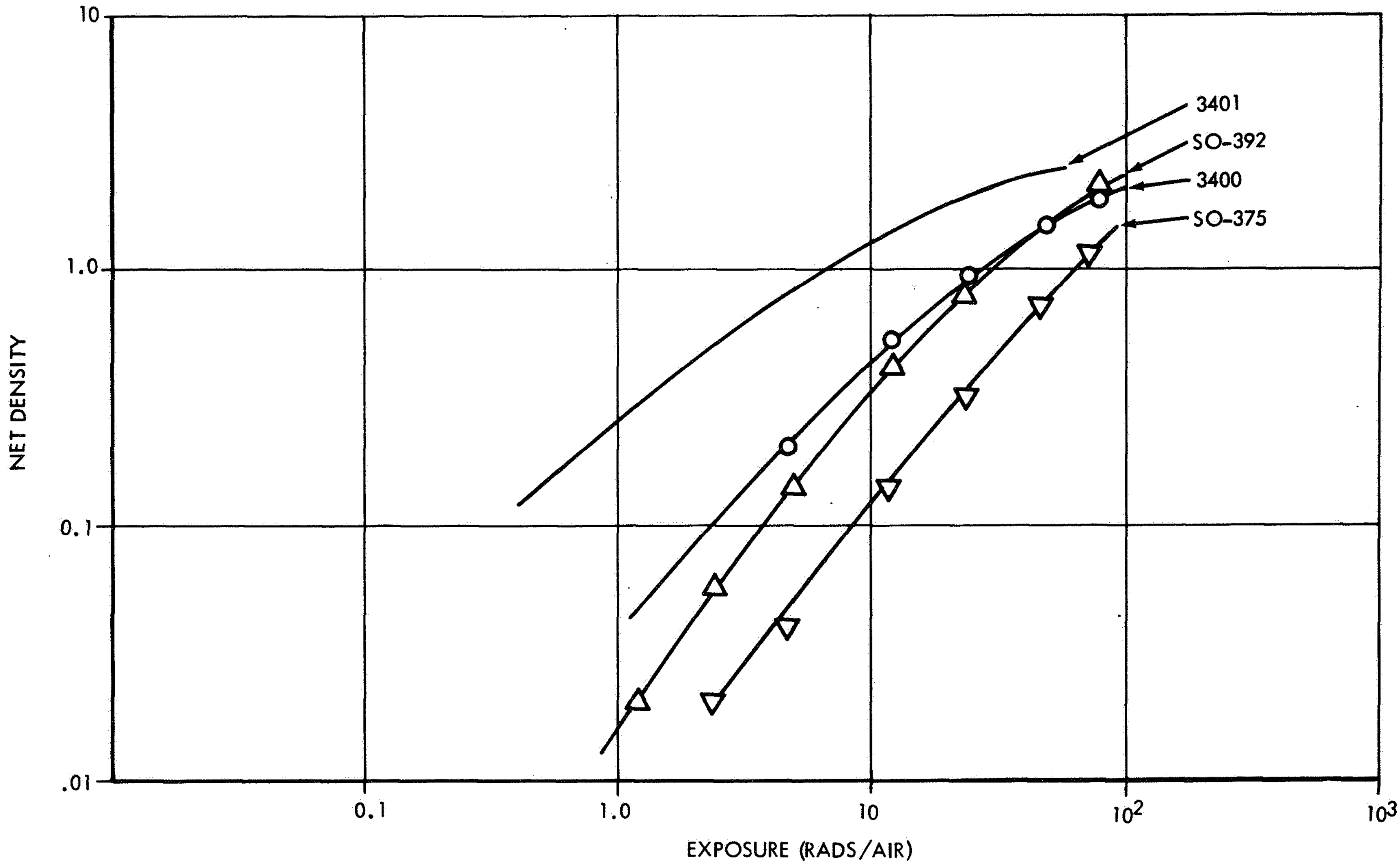

Figure III. A. 3

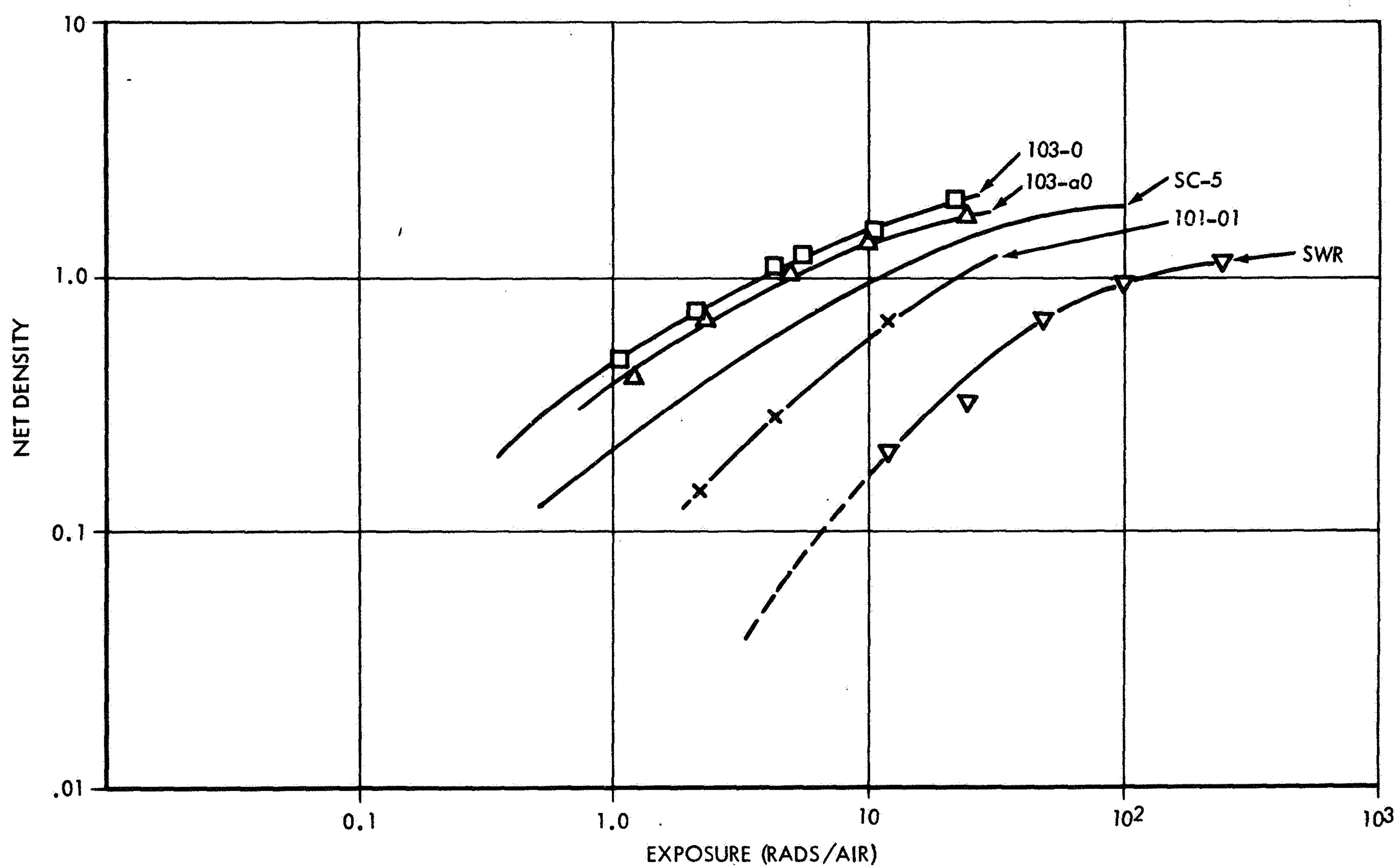

Figure III. A. 4

of a different type of radiation (e.g., protons). In order to compare the effects between different types of radiations, the concept of relative effect is used as a conversion factor. In this study, we have correlated the results obtained from 1.25 MeV gamma rays with those from varying energies of proton radiation. Again, absorbed dose is measured in terms of rads in air for easy cross reference.

The term relative photographic sensitivity (RPS) is defined for this study as the following ratio:

$$\text{RPS} = \frac{\text{Absorbed dose of 1.25 MeV gamma radiation required to produce a given density for a specified film.}}{\text{Absorbed dose of specified proton energy radiation to give the same density.}}$$

For example, if D rads/air of Cobalt-60 are required to produce a density of 0.5 for Type 3401 film, and if (7.1 x D) rads/air of 10 MeV protons are required to produce this same density for 3401 film, then

$$\text{RPS} = \frac{D}{7.1D} = 0.14$$

It can be seen that the RPS for any film is a function of the proton energy and the reference density. For this report a reference density of 0.2 has been selected, and RPS has been investigated as a function of proton energy. The base density of 0.2 was selected because several of the experiments planned for ATM stipulate 0.2 as the maximum allowable space radiation fog density. No common standard can be established for allowable radiation damage to photographic film. The limits will have to be determined according to the specific application of the film, i.e., upon the information that is desired from the recorded image.

The RPS results obtained in this study are shown in Figures III.A.5 and III.A.6.

Since the Eastman proton data did not include either 103a-0 or SO-392 films, no correlation could be performed for these types of film. For both the 101-01 and 103-0 films, there was sufficient discrepancy at 0.2 density between Eastman and CCSD gamma data to make suspect any associated proton correlation, and these two films were also omitted from this phase of the study.

The data in Figures III.A.5 and III.A.6 show that protons in the energy range investigated are generally less effective than Cobalt-60 gamma radiation in producing photographic film fogging. It can also be seen that the relative photographic sensitivity is dependent on film quality and that for all films the RPS decreases as the proton energy decreases. This can be restated by saying that low linear energy transfer (LET) particles are more efficient than those of high LET at inducing film density.

Although the described evaluating correlation is not rigorous, it is evident that the sensitivity of photographic films to protons of energy greater than 10 MeV can be determined by using Cobalt-60 gamma radiation.

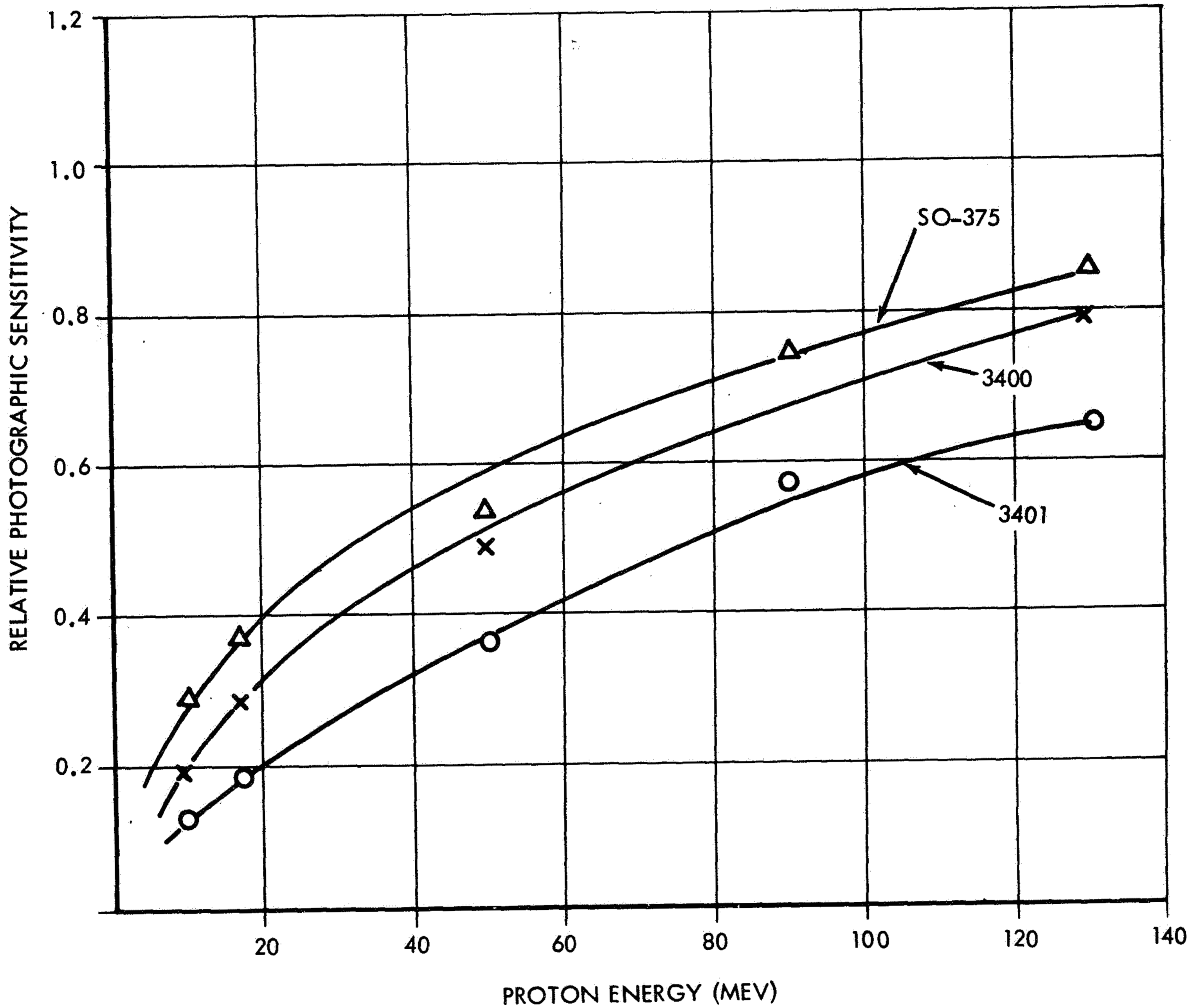

Figure III. A. 5

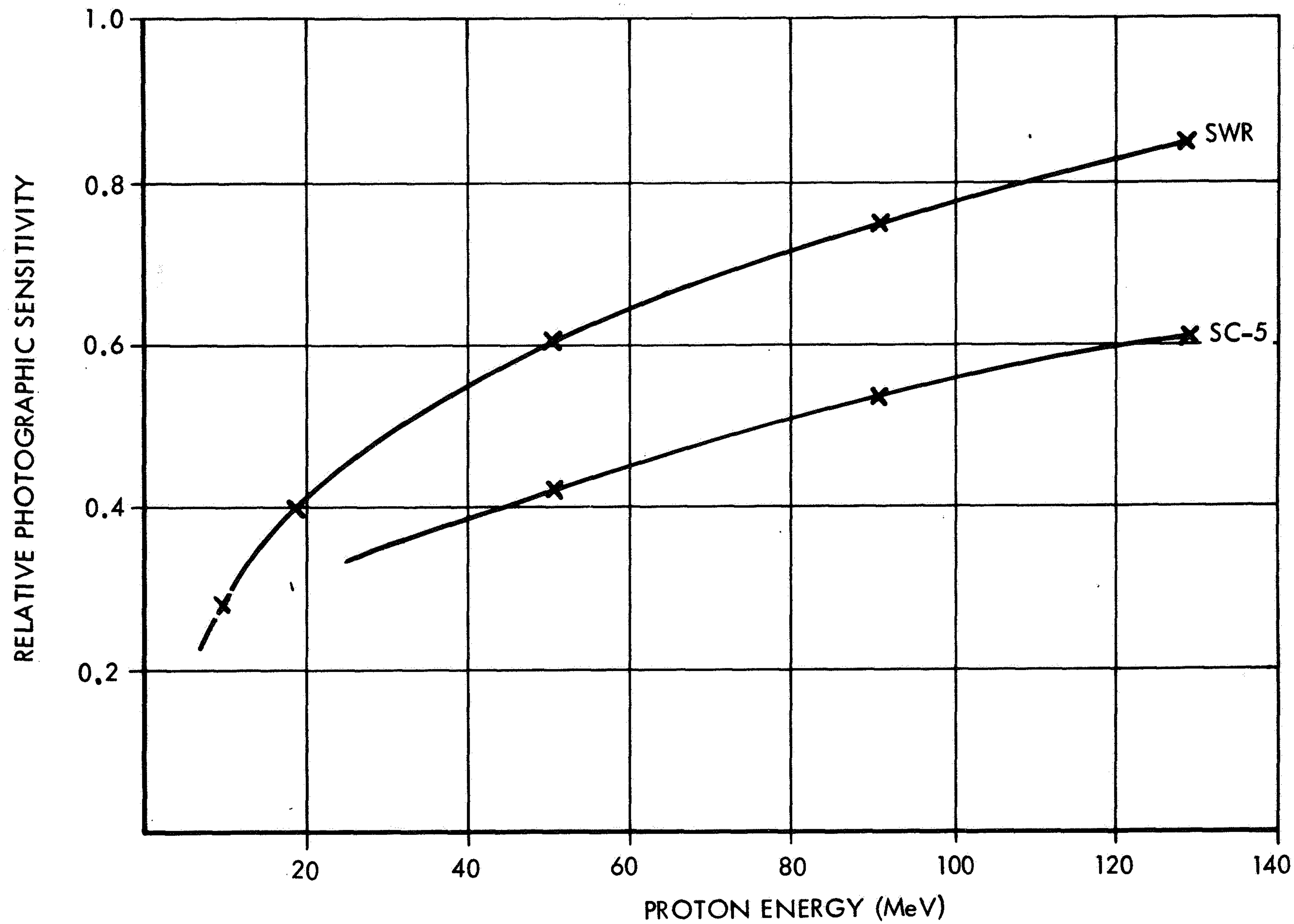

Figure III. A. 6

III.B. VARIATION OF DEVELOPMENT PROCESS PARAMETERS

Tests were performed to determine the effects of development process changes on the density of film which had been exposed to Co-60 gamma radiation. The following parameters were varied:

(a) Processing temperature

(b) Processing time

(c) Age of developer

All parameters were investigated independently, two of the parameters being maintained at their nominal values while the third was varied.

1. Time and Temperature Variation

Five types of space photographic film were investigated. Each sample of a given type was exposed to the same level of radiation, but, in general, different levels were applied to the different types. Table III.B.1 lists the pertinent data for each type of film, including the absorbed radiation dose.

In investigating the effects of variations in processing temperature, all elements in the development process, i.e., developer, stop bath and fixer, were maintained at the same temperature. Since all of these solutions are in containers immersed in a common water bath, the bath was used as the medium to set and maintain the temperature at the desired level. Bath temperature was raised by adding hot water and lowered by adding dry ice. For all data points, the temperature was allowed to stabilize for several minutes before processing and was monitored throughout the development process. Maximum error at any temperature value was $\pm \frac{1}{2}$°F.

Development time was measured with a Picker X-Ray timer for all tests. In determining the effects of variations in development time, maximum error was estimated to be $\pm$ 2 sec., including operator action in transferring films. The only segment of the developing process which was varied during the time studies was the length of time in the developer, i.e., the rinse, fix and wash times were maintained at the nominal recommended values.

The results of these investigations are presented in Figures III.B.1 through III.B.5. In these curves, temperatures and time have been normalized to their recommended values and the densities have been normalized to the densities obtained at these recommended values. By definition, then, the value along the density scale must be 1.0 for a

Table III.B.1

Film Type	Recommended Development Time (Mins.)	Recommended Development Temperature (°F)	Absorbed Test Dost (Rads/Air)	Resultant Gross Density for Normal Development	Control Density For Normal Development
SWR	2	68	96	1.02	.03
3400	8	68	14	.74	.07
SC-392	8	68	16	.73	.14
103a-0	4	68	2.4	.76	.14
3401	8	68	3.2	.78	.08

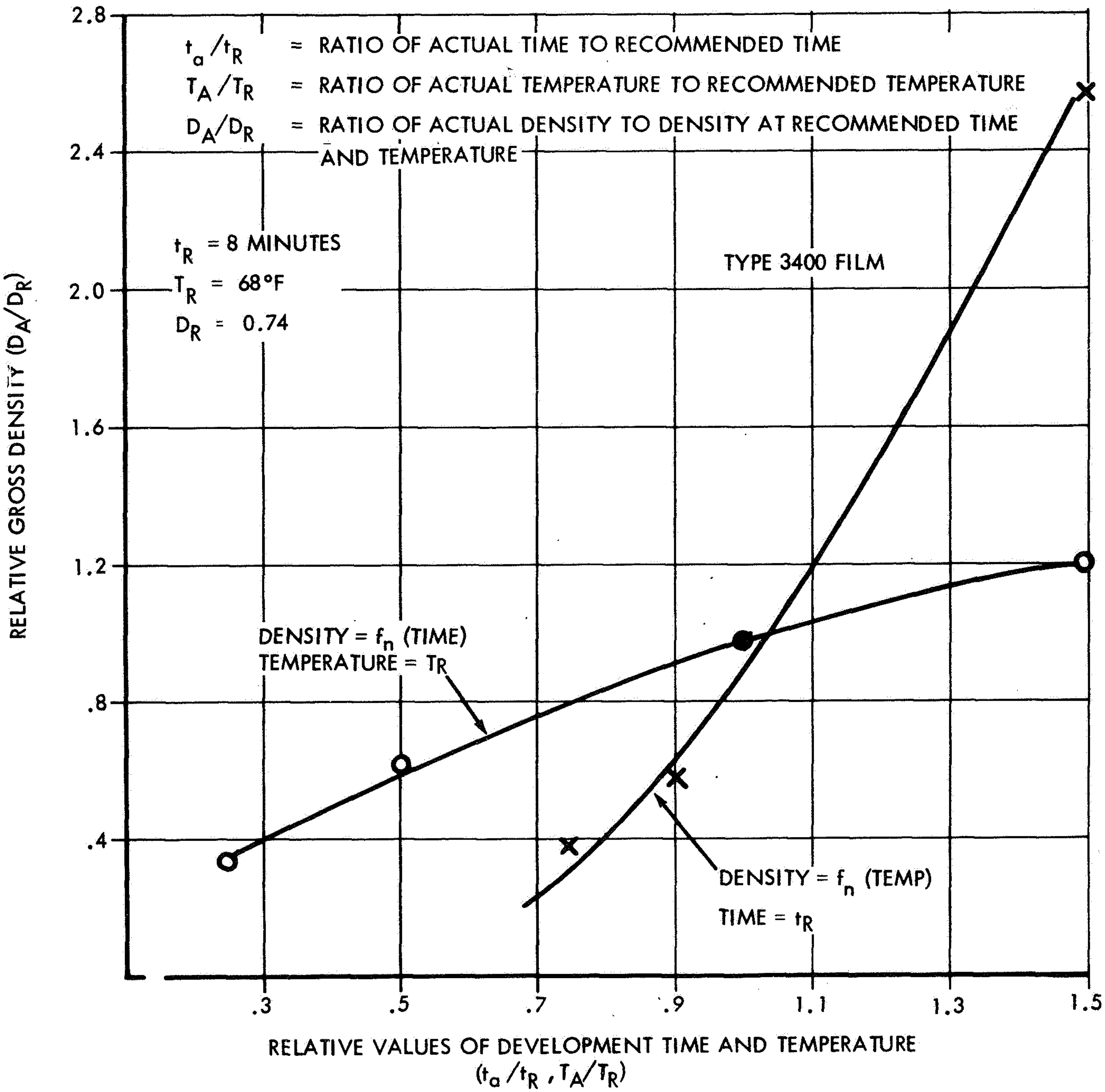

Figure III. B. 1

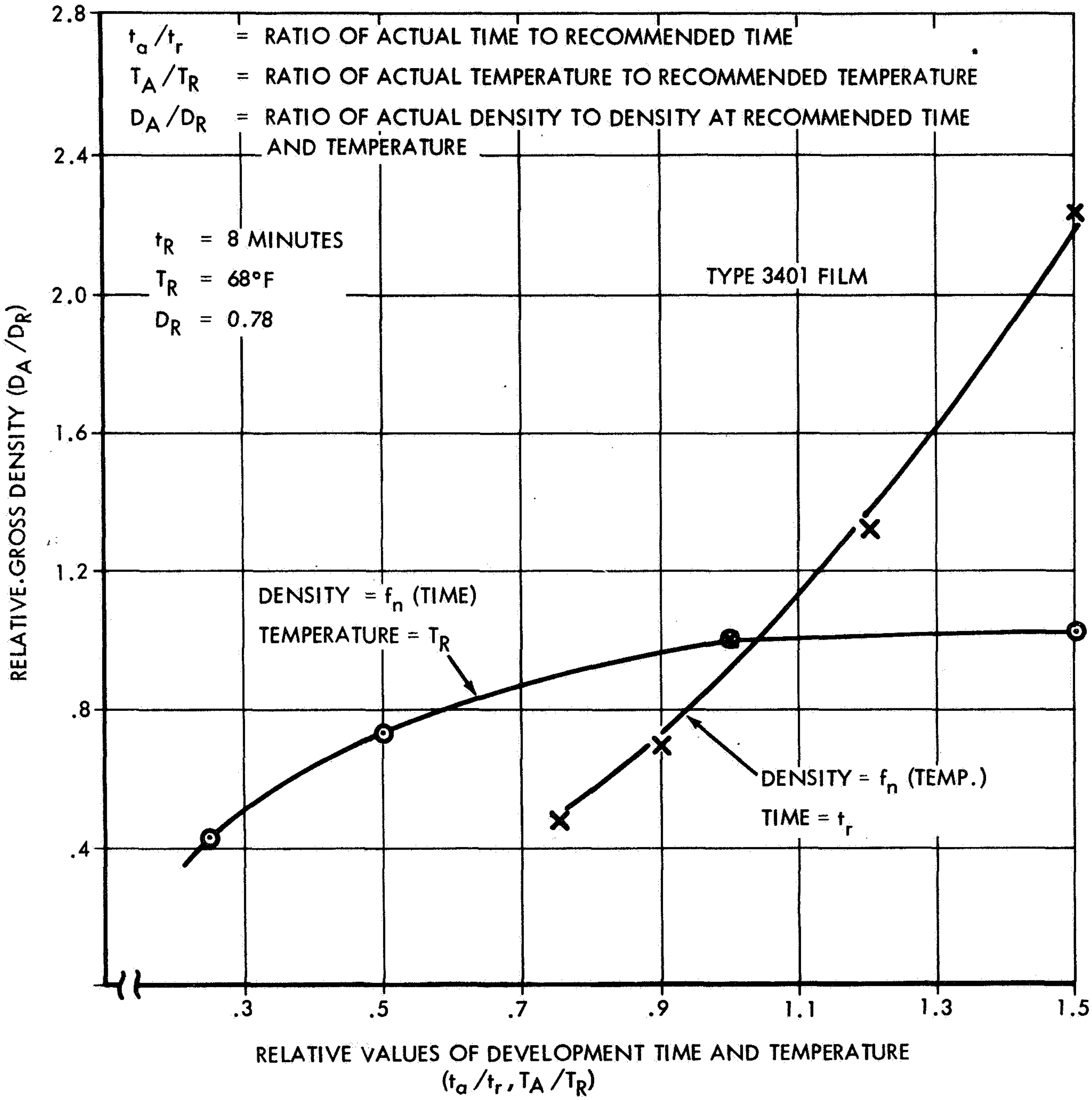

Figure III. B. 2

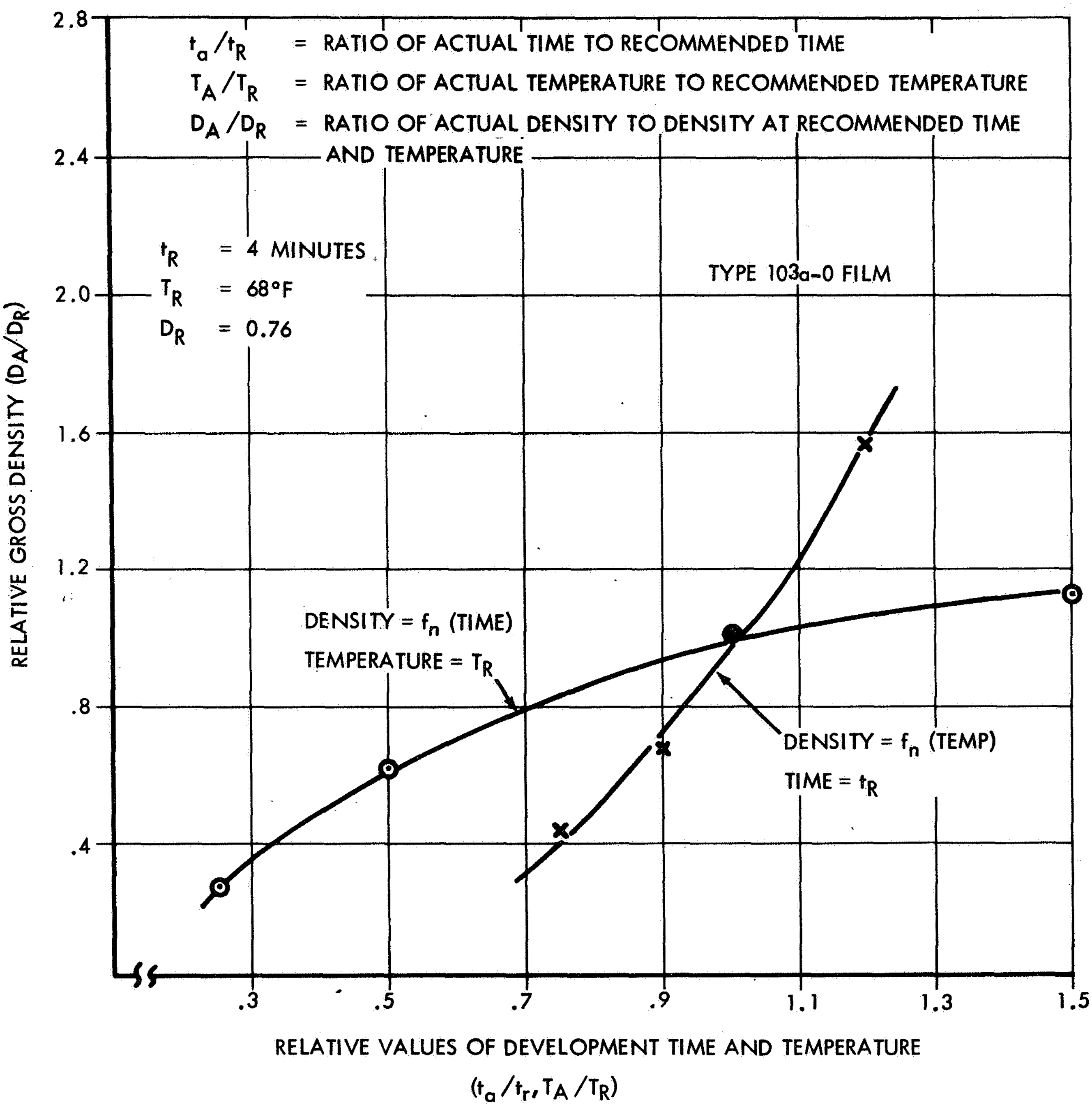

Figure III. B. 3

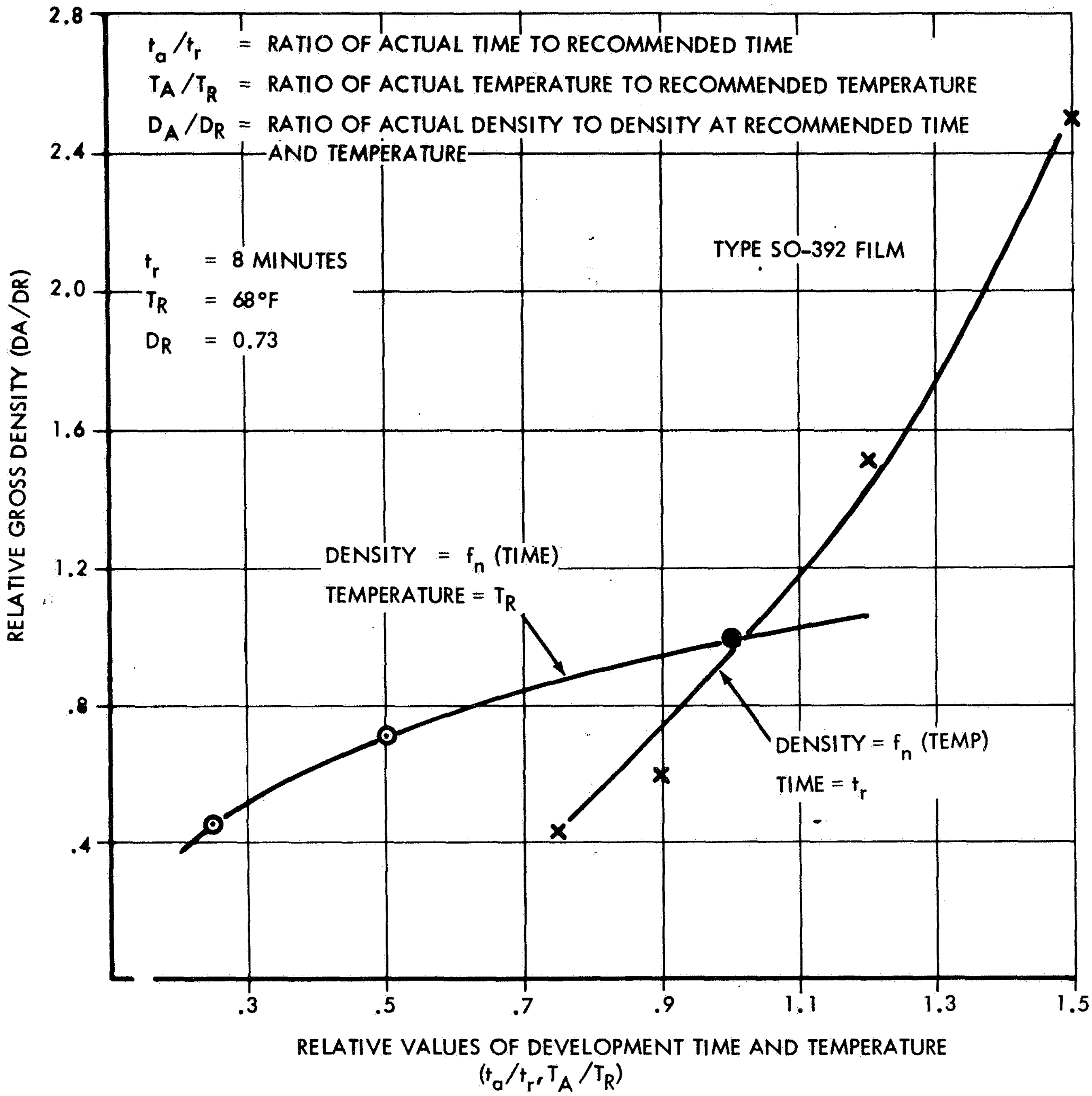

Figure III. B. 4

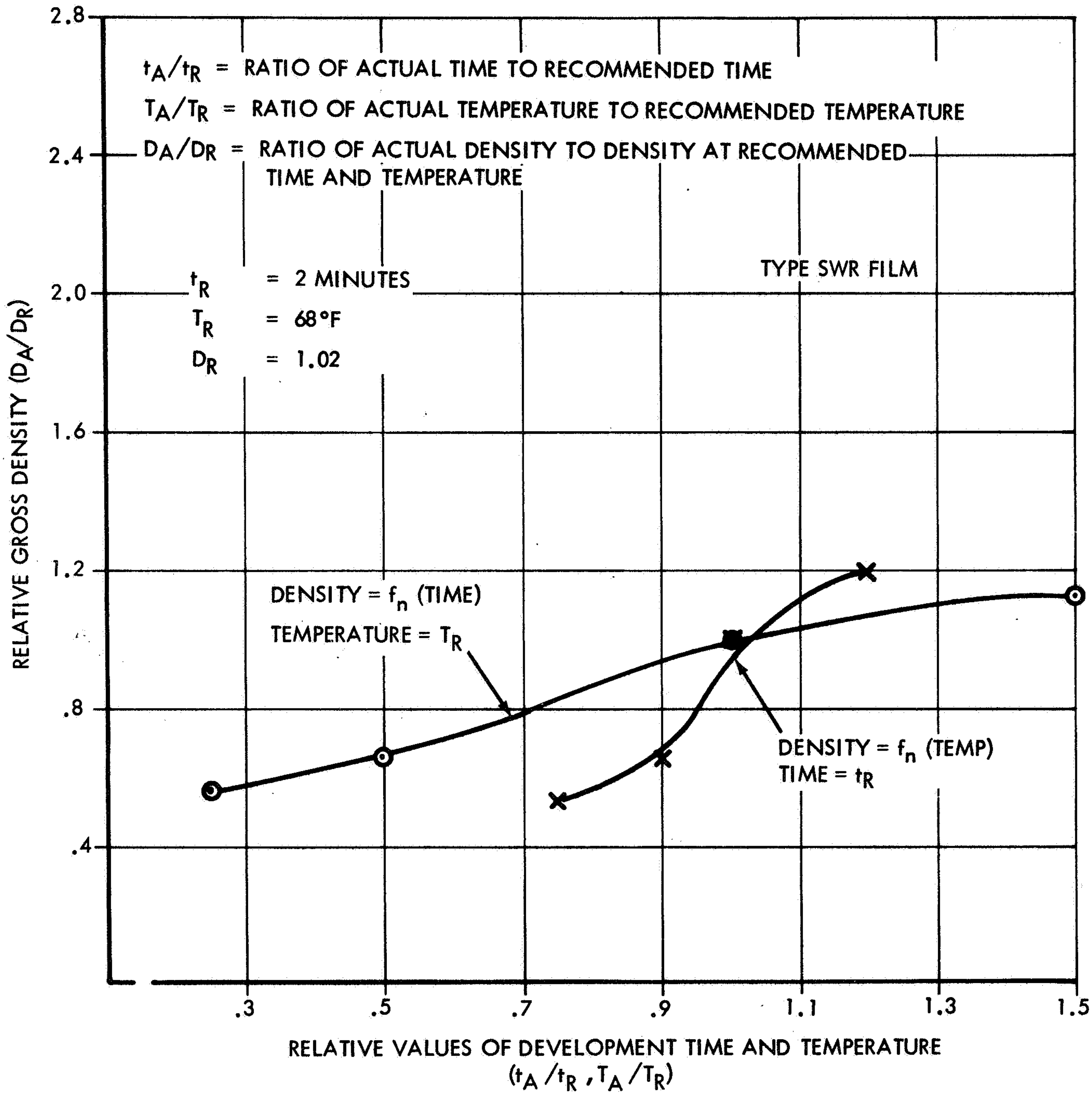

Figure III. B. 5

time or temperature value of 1.0.

The data represented in Figures III.B.1 through III.B.5 are preliminary in nature and are only intended to indicate the trend of density variation to be expected as a function of the specified parameters. Further work originally planned in this area was diverted to an investigation of the low temperature protection discussed in Section III.C.

The following effects should however be noted:

(1) The change in film density is much greater for a temperature variation than it is for an equivalent time variation, particularly for values greater than nominal.

(2) The density-time relationship is a much 'smoother' function than is the density-temperature relationship.

For both the time and temperature variations study, the results show the variation in density to be insignificant within the processing controls available in a professional darkroom.

2. Aging of Developer Solution

In order to assess the effect of developer solution aging on film density, a number of strips of Eastman 3401 film were exposed to the Co-60 source on the same day on which a new batch of developer (Eastman Kodak D-19) was prepared. Half of these strips were exposed for 15 seconds and the other half for 30 seconds so that two density values could be obtained. The exposed strips were then stored under refrigeration. Each week, one strip from each exposure level was developed in the original developer, so that a plot of density as a function of developer age was determined for 1-week aging intervals. No films other than those used in this test were processed in this particular developer so that the effects of chemical exhaustion were negligible. The developer was covered but not air tight during the period when not in use. The effect of aging of the developer solution is shown in Figure III.B.6.

From Figure III.B.6 it can be seen that the effect of developer aging alone on density is minimal. Although there appears to be a gradual dimunition with time, the increases appearing for both densities at the end of the first and ninth weeks show that other factors can mask out the effect of aging.

The effect of developer aging alone on density is not nearly so great as the effect due to large numbers of films being processed in the same developer solution. The latter effect was not investigated during this study, but has been noted previously by the experimenters and is reported on in the literature.

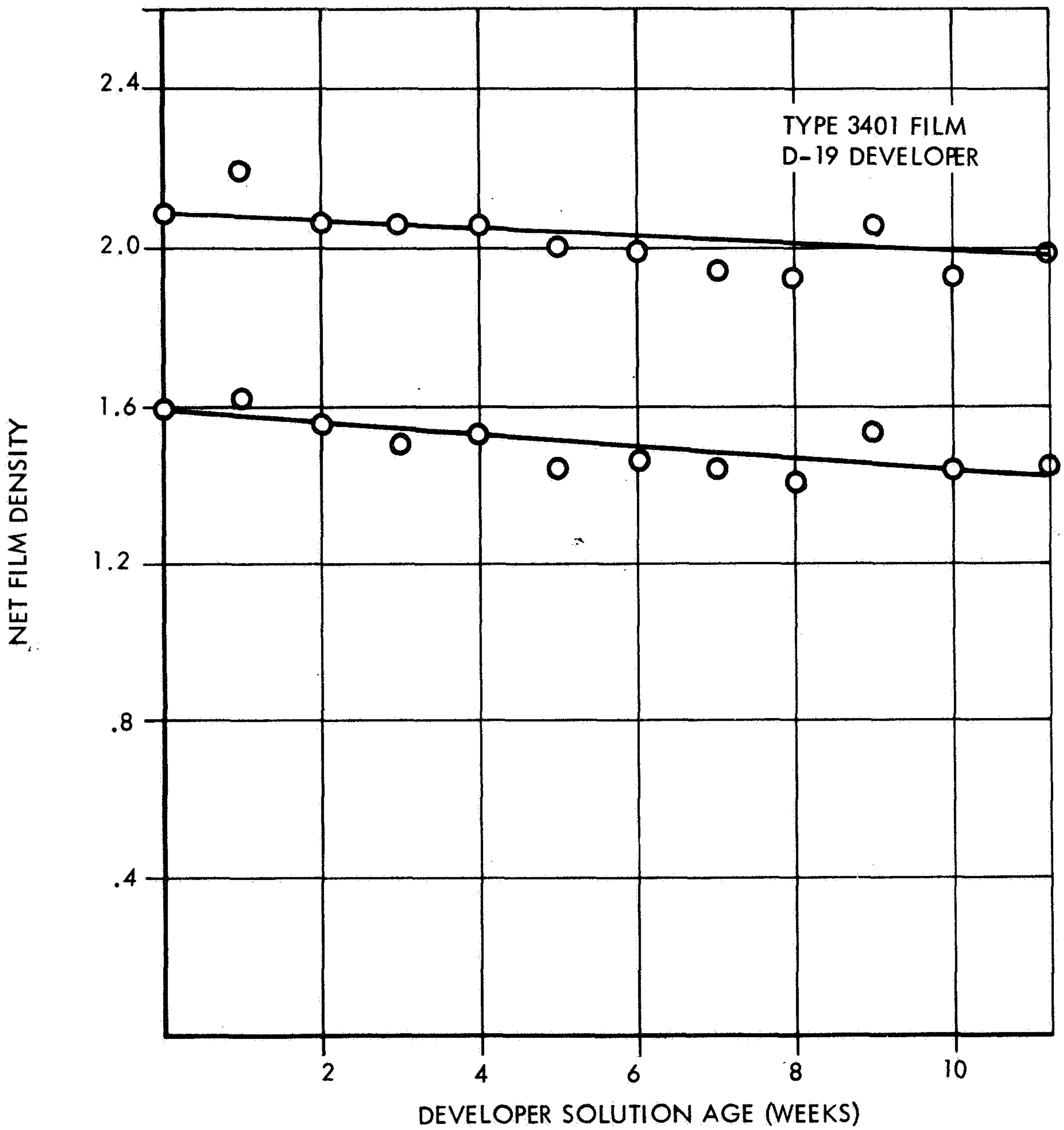

Figure III. B. 6

III.C. LOW TEMPERATURE FILM STORAGE

The terms of the original contract NASW-1741 did not include an investigation of low temperature storage. However, an in-house study by CCSD (See Appendix A) of methods to reduce background fogging of photographic film indicated the high potential of such a technique. It was therefore decided, with NASA approval, to redirect a portion of the original work scope into this area. As a result, the investigation of the effects of variations in processing parameters was not completed to the extent originally planned.

Considerable data exists in the literature to verify the fact that film sensitivity is affected by the exposure temperature. Previous theoretical and experimental studies are discussed in Section III.C.2. For purposes of reducing background fogging, the 'exposure time' in this study corresponds to the time during which the film will be stored awaiting exposure to light for a space mission.

1. Rationale for Study

There are several characteristics of the low temperature storage method which made it particularly suitable for study under this contract. From an appraisal of the various protective methods we have examined, low temperature storage appeared to be the most feasible protective method for space. In addition, meaningful data could be obtained in the relatively short time span (less than two months) remaining in the program at the time of go-ahead for this portion of the study. The particular points which contributed to the decision to proceed with this investigation are listed below;

(1) It has previously been demonstrated that film sensitivity is reduced at low temperature. No proof-of-principle testing is required, only the extent of the effect must be determined.

(2) The testing apparatus required is relatively simple. This was necessary because of the limited time remaining under the contract.

(3) Only two parameters need to be investigated -- temperature and sensitivity.

(4) The simplicity of the method makes it the most feasible for space application in the near future, such as ATM.

(5) The possibility of a degrading effect on the film due to the protective measure is slight, and the extent of degradation can be easily determined.

A requirement for the application of low temperature storage in space is the availability of a lightweight, low volume, low power refrigerant system. Such a requirement can be satisfied by a system consisting of a solid state cryogen and a heat pipe, or pipes, as shown in Figure III.C.1. Once the film has been cooled to a temperature approximating that of the cryogen (this can be done prior to launch), the only refrigerating components required for space flight are the cryogen, the insulated chamber in which the cryogen is stored, and the heat pipe (or pipes, if separated packages are to be cooled). The system shown in Figure III.C.1 has the additional advantage that it can go around corners or be inserted between pieces of equipment or structural members.

2. Previous Studies

The explanation for reduced silver halide sensitivity at low temperatures is apparent from an examination of the basic two-step process involved in the formation of the latent image. In step one, an electron is released by the energy of the incident photon and is "trapped" at some location in the crystal. In step two, an interstitial silver ion moves toward, and combines with, this trapped electron. Therefore, the mobility of the silver ions is an essential requirement for latent image formation. This mobility is a direct function of the temperature of the crystal. Therefore, as temperature decreases, the mobility of the silver ion decreases and the probability of silver ion-electron combination is reduced.

Although considerable work has been done in the past to investigate the effects of reduced film temperature on film sensitivity, there has been little effort aimed specifically at using this technique to reduce background nuclear radiation fogging in photographic film. A brief review of the more pertinent work appearing in the literature is presented in the following paragraphs.

- Webb and Evans[3] performed an experimental study of latent-image formations by means of interrupted light exposures at low temperature. They found that the higher the flashing frequency, the greater was the film density, provided the emulsion was warmed between flashes. When the film was maintained at low temperature during the time between flashes, the resultant density was very much reduced over that when the film was warmed between flashes. They interpreted this as evidence that the loss in emulsion sensitivity with lowered exposure temperature is due to a decrease in ionic conductivity.

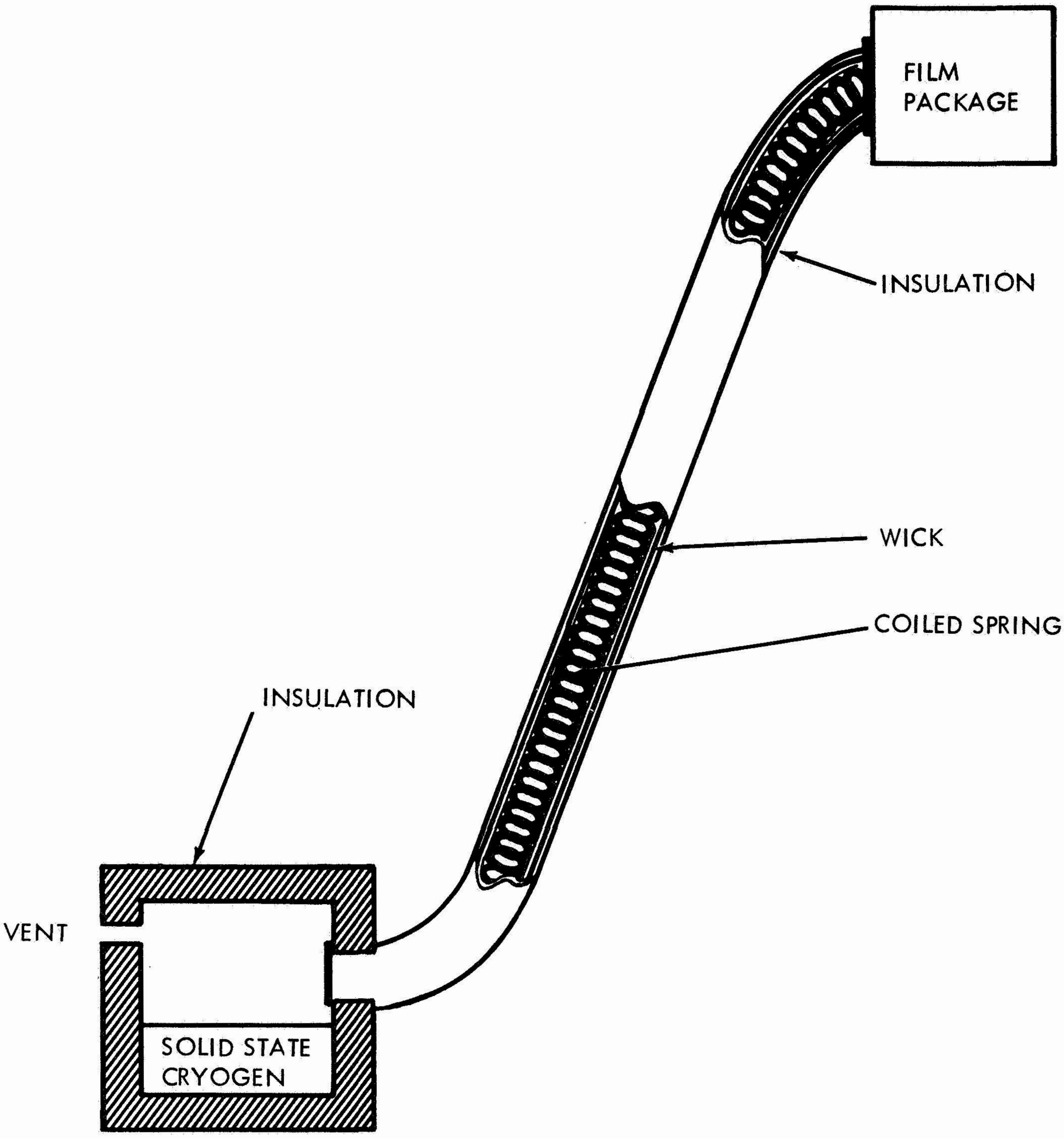

Figure III. C. 1

- Berg and Mendelssohn[4] investigated photographic sensitivity and the reciprocity law at low temperatures. A principal conclusion of this study was that at the temperature of liquid helium (4°K), the ratio of X-ray sensitivity to light sensitivity is much higher than that ratio at room temperature.

- J. Reekie[5], in studying the dependence of photographic sensitivity on temperature, found a linear relationship between sensitivity and temperature down to approximately -80°C.

- H. Baas[6] also investigated the variation of film response with temperature. He found that the temperature of the film before and after exposure has no effect on the film density.

- In 1952 Beltz[7] investigated photographic sensitivity as a function of exposure time and temperature, particularly in the region between -150°C and -195°C. In this region he observed a large decrease in film sensitivity and also a strongly temperature-dependent fluorescence. He believed these two phenomena to be related.

- Wanick[8] in 1956, published an extremely important paper describing his investigation of the sensitivity variation of nuclear emulsions with temperature. He reported the following observations:

    1. A fluctuating density-temperature response for temperatures in the region 10° to 65°C.

    2. A peak in sensitivity in the region near -40°C.

    3. A decrease in sensitivity from -40°C to -180°C.

    4. Sensitivity at -196°C was 75% of that at 20°C.

- Confirmatory observations of Biltz's work were obtained in 1957 by G. C. Farnell[9].

Two major reporting areas are not covered here. These are the studies which are either deemed company proprietary or have been classified for national security reasons.

3. CCSD Tests

The fact that the gamma exposures were performed at a local medical clinic necessarily restricted the complexity of the apparatus which could be used to obtain and monitor low temperatures during exposure.

In line with this restriction, a passive method was used to obtain the desired range of low film temperatures. Several film strips, each sandwiched between the plexiglass layers required for electron equilibrium, were placed in a polystyrene container which was filled with liquid nitrogen (LN_2) to a level sufficient to cover all the films. The LN_2 was then drained from the container and the films were allowed to passively rise to room temperature while in the container. The temperature of these films was a function of elapsed time during the warm-up process. Figure III.C.2 shows the elapsed time-temperature relationship for the system used in the tests.

Initially the data of Figure III.C.2 was used as a direct measurement of the exposure temperature, i.e., the films were removed from the container for irradiation after an elapsed time value corresponding to the desired temperature as determined from Figure III.C.2, and this temperature was recorded as the exposure temperature. In later tests a thermocouple was inserted between the plexiglass slabs during irradiation, and the recorded temperature determined from the thermocouple reading. However, some difficulty was experienced in positioning and maintaining the thermocouple on the film during exposure.

Results of these tests are shown in Figure III.C.3 for film type 3401. For all tests, exposures were performed at 150 cm SFD. amd for 3 second and 6 second intervals, corresponding to absorbed doses in air of approximately 0.6 and 1.2 rads, respectively. The plexiglass slabs are sufficiently good thermal insulators to maintain the film at relatively constant temperature during these exposure periods.

Although the data for two separate test runs at 3-second exposures is quite consistent (density values within 10 per cent of one another), the 6-second data is erratic with poorer repeatability between the two tests. We are not certain at this time as to how much of the fluctuations in the 6-second curves is due to experimental error and how much is true data. Previous experiments in the literature show similar fluctuations for low temperature tests using light as the irradiation source. There is obviously some discrepancy between the separate test runs and this is almost certainly due to errors in measurement of the temperature during exposure.

All of the curves of Figure III.C.3 show a relatively sharp increase in film sensitivity in the region above -30°C. Below this temperature, the sensitivity remains relatively constant for the low density measurements, corresponding to 3-second exposures, but appears to continue to slowly decrease, with possible fluctuations, for the higher density measurements, corresponding to the 6-second exposures.

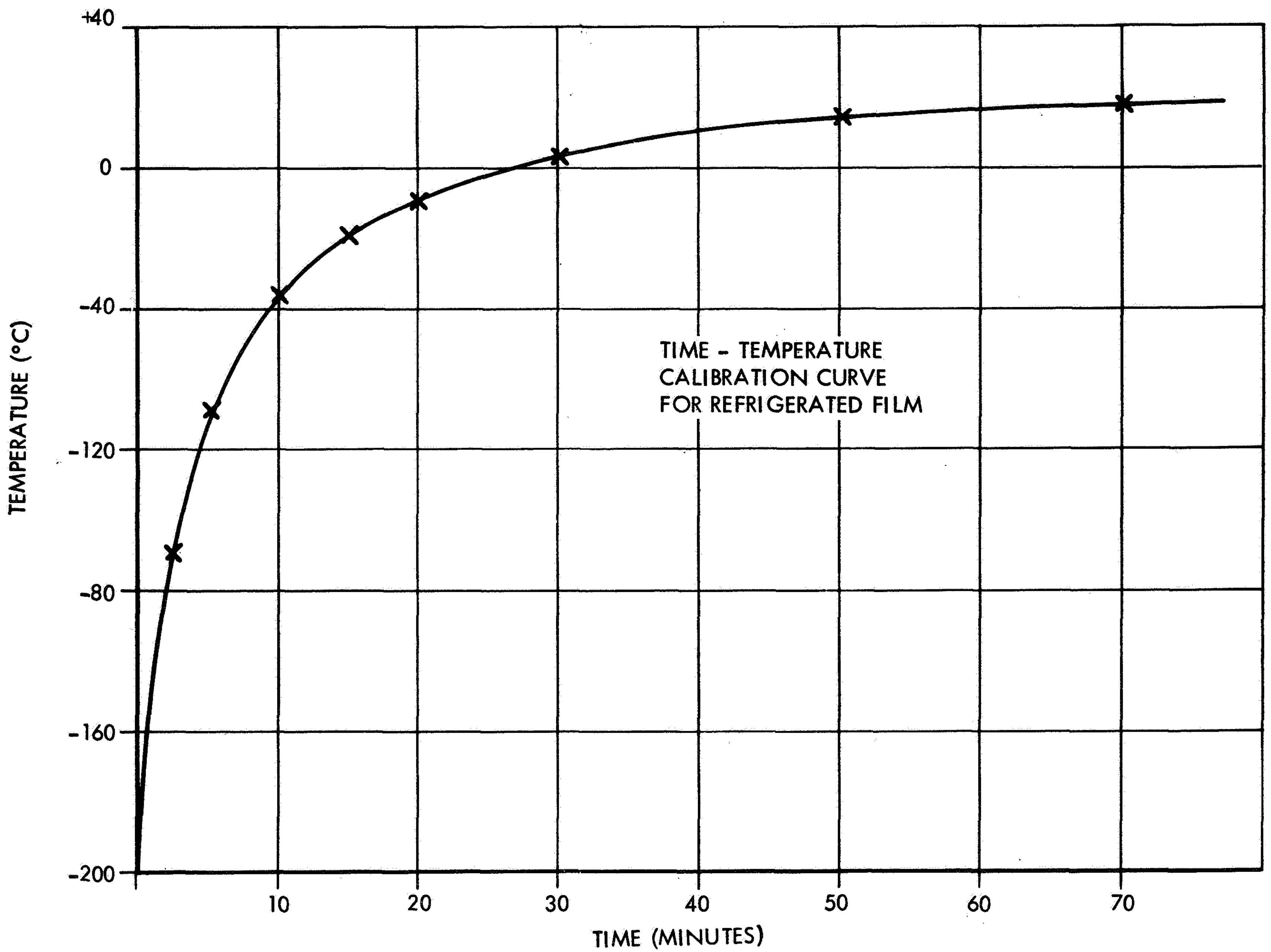

Figure III. C. 2

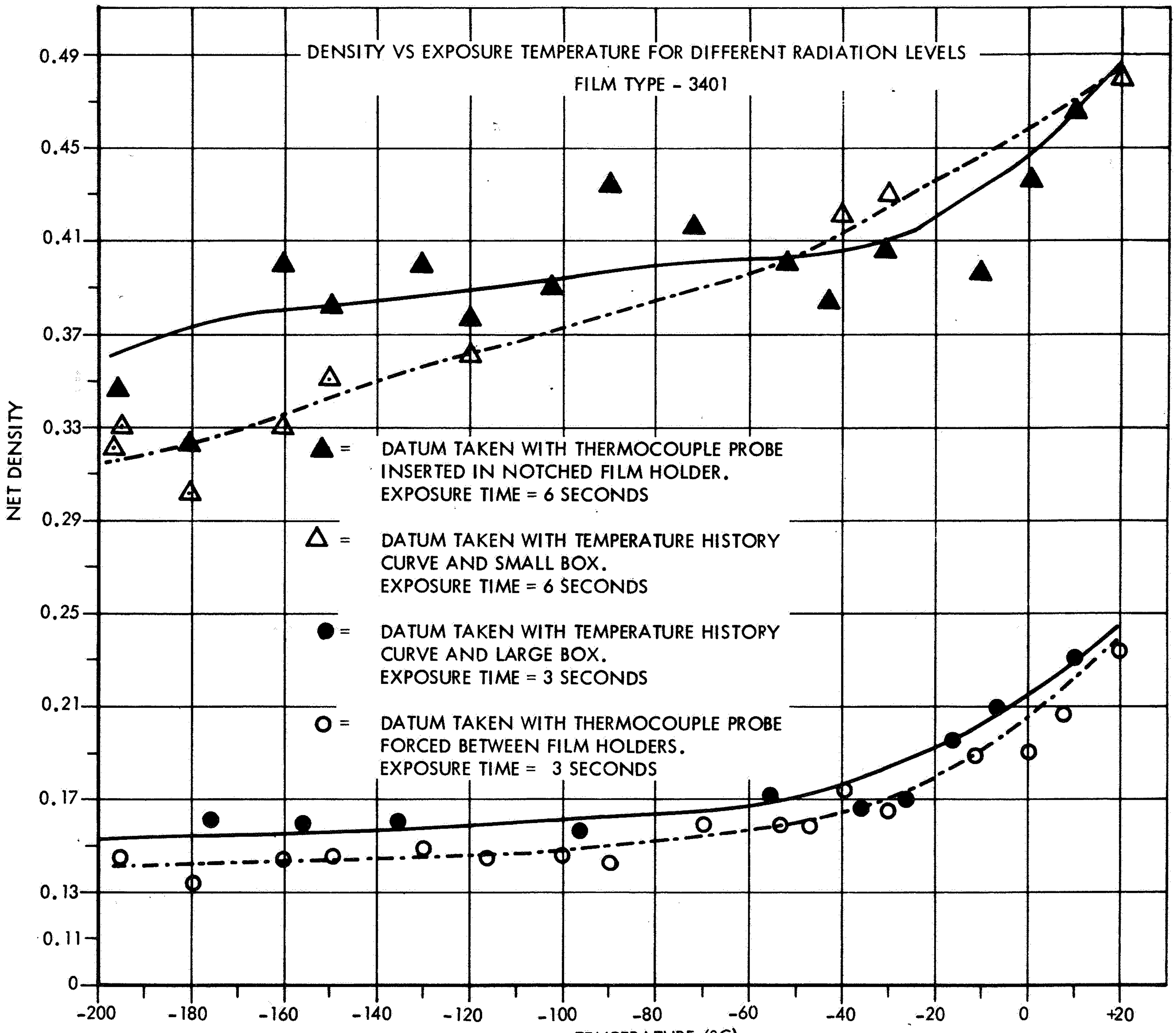

Figure III. C. 3

Table III.C.1 lists the reduction in density obtained at - 96°C exposure temperature for 3401 film at three gamma exposure levels.

Exposure Level	Density		Percent Density Reduction
	Exposure Temp. = 22°C	Exposure Temp.= -196°C	
1	0.24	0.15	37
2	0.49	0.35	29
3	0.90	0.65	28

Table III.C.1

These tests show the feasibility of this method as a fog retardant and provide an indication of the magnitude of the effect to be obtained.

IV. RECOMMENDATIONS FOR FUTURE STUDIES

Of the various study areas investigated and described in this report, the most promising is that of low temperature film storage.

The preliminary results obtained under this contract show it to be a feasible protection method, and there is a definite requirement for some form of film protection in addition to mass shielding. The necessity for some form of additional protection is apparent in current plans for the ATM-A mission, where, in order to reduce the background fog from space nuclear radiation to an acceptable level, photographic film has been selected which downgrades the overall optical system of the experiments.

The low temperature study should be divided into three separate phases. Phase I will consist of a determination of film density as a function of exposure temperature for a given radiation dose. This should be done for various amounts of absorbed dose and for all film which is currently being planned for space use. The simplest technique would be to irradiate with a gamma source in the manner performed under this contract, and extrapolate the results to an equivalent proton dose.

Phase II will consist of a study of low weight refrigerant systems designed for this particular application - that is, to refrigerate a small package (or packages) to extremely low, possibly cryogenic, temperatures.

Phase III will combine the results of Phase I and Phase II in parametric and trade-off studies to determine the weight, power, etc. penalties. In particular, the relative effectiveness and penalties associated with mass shielding and low temperature storage will be investigated.

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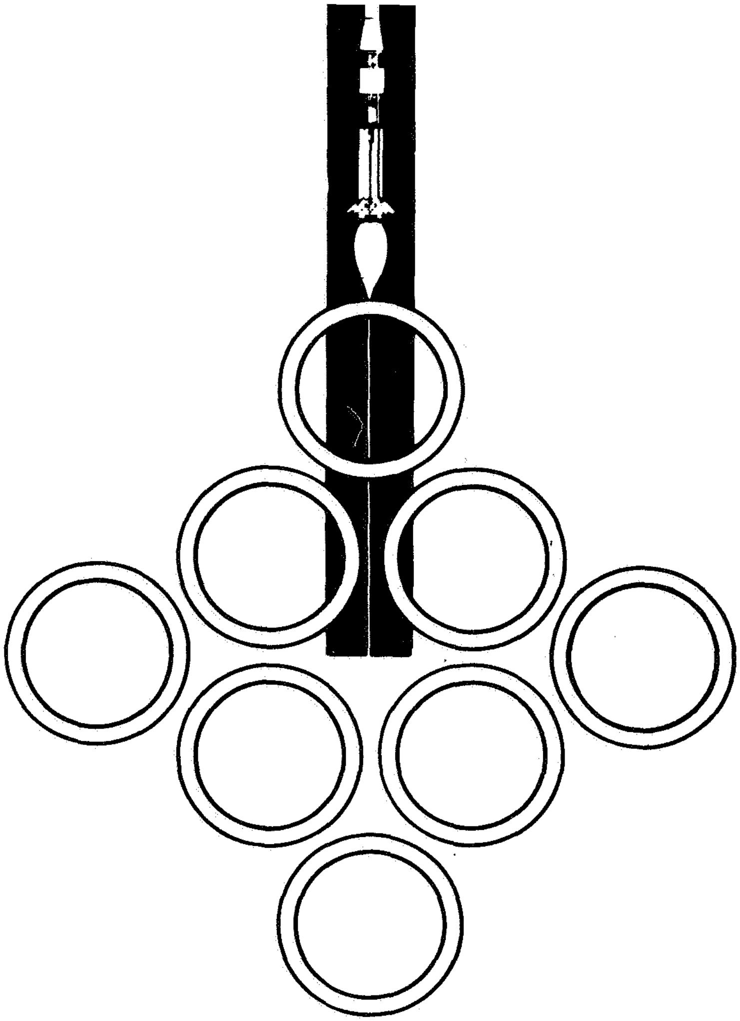
APPENDIX A



ENGINEERING DEPARTMENT

TECHNICAL NOTE

TN-AE-68-183

PROPOSED METHODS TO REDUCE BACKGROUND RADIATION IN PHOTOGRAPHIC FILMS

SATURN S-IB STAGE AND SATURN IB PROGRAM

SPACE DIVISION CHRYSLER CORPORATION

TECHNICAL NOTE TN - AE - 68 - 183

PROPOSED METHODS TO REDUCE BACKGROUND RADIATION IN PHOTOGRAPHIC FILMS

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ABSTRACT

The reduction of radiation background can be effected in two different ways: by inhibiting the formation of the undesired latent image, or, once it is formed, by its erasure. Five methods are discussed here under the first category and three under the second.

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INTRODUCTION

Radiation which produces background fog in photographic emulsions is unavoidably present in space missions.

Several methods have been investigated to reduce this background fog without appreciably affecting the desired image.

The purpose of this paper is to survey these methods and to present any pertinent experimental data available. Indeed, the object is not the solution of the problem but a clear specification of the possible solutions.

DISCUSSION

The methods proposed to reduce background radiation in photographic films can be divided into two categories:

I. ERASURE OF LATENT-IMAGE

a. Herschel Effect
b. Oxidizing Agents
c. High Humidity

II. PREVENTION OF LATENT-IMAGE FORMATION

a. High Humidity Storage
b. Manufacturing the Films in Space
c. Developing the Films in Space
d. Low Temperature Storage
e. Storage Under the Influence of an Electric Field

Each method will be discussed separately.

I. ERASURE METHODS

a. THE HERSCHEL EFFECT

If an emulsion which has been exposed to blue light is subsequently exposed to red light before it is developed, some of the effect of the original exposure may be destroyed. This change in the latent image is known as the Herschel effect, since observations of a similar kind were published by Sir John Herschel in 1840.

The mechanism for this bleaching action which is generally accepted at present was proposed by Gurney and Mott.[1] In order to understand this mechanism a clear picture of the formation process of the latent image must be borne in mind. This is essentially the same for heavy charged particles and for the photons of visible light. By action of the incident radiation, electrons are raised from their normal crystal lattice positions in the silver halide grain to the conduction band of the silver halide. These electrons move through the conduction band until they fall back into their normal positions or become trapped at impurity centers where their potential energy is lower than in the conduction band. The negatively charged center will then attract a silver ion, thus forming an atom of metallic silver. When this process has been repeated a sufficient number of times a speck of metallic silver (the latent image) will have been formed which will serve as a nucleus for the reduction of the entire grain to metallic silver in the developing process.

In the first step of the Herschel effect an electron is ejected from the latent image by the absorption of an infrared photon. The energy required for this is small compared to the energy which is necessary to raise an electron to the conduction band of the silver halide. In the second step the silver ion left by the removal of the electron moves away leaving the latent image smaller by one atom. That the migration of the silver ion is a necessary part of the process has been demonstrated by Webb[2] by conducting the infrared exposure at the temperature of liquid air (87°K). At this temperature the silver ions cannot migrate, and consequently, no Herschel effect was obtained.[3]

The Herschel effect has been used as a means of obtaining selective erasing of nuclear emulsions. Experiments performed by Goldstein and Sherman[3] have proven that it is possible to erase background darkening which would have completely obscured particle tracks without erasing any of the tracks, thus extending useful exposure periods. Background darkening of Eastman Kodak nuclear track plates (types NTA) was produced by visible light, x-rays, Co^{60} gamma radiation, and beta particles of Sr^{90} and Y^{90}. The 5.3 Mev α- particles of Po^{210} were used to obtain the individual tracks. The source of infrared radiation, in conjunction with a Wratten gelatin filter, No. 88, transmitted at wavelengths greater than 7000 Å. By use of this exposure the permissible background exposure intensities were increased by factors of 3 to 7, without destroying any of the alpha tracks. A reproduction of their results is given in Table I.

Table I

INCREASE IN MAXIMUM ALLOWABLE BACKGROUND EXPOSURE WHEN USING THE HERSCHEL EFFECT AS A MEANS OF SELECTIVE ERASING

Type of Radiation	Maximum Normal Background Density for Seeing all the Tracks	Maximum Normal Allowable Exposure	Maximum Background Density Which can be Reduced to Point Where all Tracks can be Seen	Maximum Allowable Exposure Using Herschel Effect for Selective Erasing
Light	0.3	5 sec	2.2	15 sec
X-Ray	0.8	0.25 sec	1.8	0.75 sec
Co^{60} γ	0.8	4×10^9 γ/cm^2	3.3	3×10^{10} γ/cm^2
Sr^{90}- Y^{90} β	0.8	6×10^8 β/cm^2	3	2×10^9 β/cm^2

The effects of the infrared radiation were no different in the cases where the plates were exposed to the alpha-radiation before or after the background exposure, from which it was concluded that the results would be no different if the tracks were recorded simultaneously with the background darkening. Unexposed NTA plates were exposed to the infrared source for periods up to 90 minutes and showed no significant increase in density upon development.

To insure that alpha-particle tracks were not being erased by the infrared, two areas on a plate were given equal exposures to the polonium source: one area was darkened with x-rays and then erased by infrared exposure, the other area had no further exposure. The plate was developed, and the tracks in each case counted at high magnification. The number of tracks in each area was the same within the limits of experimental error.

It is worth noting that the maximum background density for seeing all the tracks was lower for background produced by visible light, because most of the developed grains are near the surface of the emulsion in this case, and the tracks are also near the surface. In backgrounds produced by the other types of radiation the developed grains are uniformly distributed through the thickness of the emulsion.

The results of the work described above constitute sufficient evidence for the efficiency of the Herschel effect as a means of obtaining selective erasing in nuclear emulsions.

As the authors point out, however, several aspects remain to be further investigated:

(a) Variation of selectivity with wavelengths of infrared radiation.

(b) Variation of selectivity with energy of the radiation producing the background.

(c) Limits on selectivity imposed by the statistical nature of energy absorbed per grain.

(d) Limits on selectivity imposed by grain size and distribution of latent image silver atoms in a grain.

b. <u>OXIDIZING AGENTS</u>

In his book, "Photography", C. D. Neblette makes the following observation: "A latent image may be produced in a number of ways other than by exposure to light or other forms of radiant energy. Toward the end of the last century, W. J. Russell found that many substances, including freshly scratched metals, many fats and volatile oils and numerous organic bodies, such as wood, straw, and resin, would produce developability in a photographic emulsion without exposure to light. The activity of all of these materials was traced to the formation of hydrogen peroxide as a result of the oxidation of the substances in moist air. Hydrogen peroxide itself exhibits the phenomenon in a much more marked degree.

The action of hydrogen peroxide on a photographic emulsion shows an extraordinary parallelism with the action of light and other forms of radiant energy. It increases with time, or with concentration, giving rise on development to a characteristic curve similar to that produced on exposure to light. In both cases, there is a well-defined reversal portion and the sensitivity to hydrogen peroxide parallels that of light".[4]

Experiments in relation to this phenomenon have been performed by Yagoda and Kaplan.[5] They were able to diminish the latent image by the process of accelerated fading caused by augmenting the H_2O_2 concentration of the atmosphere in which the plates were stored. Their experiment consisted essentially on the following: After exposure to 10^7 α- particles from RaF, the latent image was completely obliterated by keeping the emulsion over 3 percent H_2O_2 at 25°C for 4 hours. After 1-2 hour drying of the eradicated emulsion over $CaCl_2$, sensitivity is restored and tracks develop from new exposures with normal mean grain spacings.

The authors point out that this method of latent-image eradication has been applied only to fine-grained nuclear type emulsions as Eastman NTA and Ilford Conc. With coarse-grained, light-sensitive emulsions as X-rays films and process plates there is a tendency for the H_2O_2 vapor to produce pseudophotographic effects. Loading fine-grained emulsions with borax and lithium sulfate inhibits the destructive action of H_2O_2 on the latent image, and background eradication must therefore precede loading.

The mechanism is essentially an oxidation-reduction process. It consists of electron capture by the oxidizing agent and the consequent oxidation of metallic silver.

In the case of hydrogen peroxide, the reactions are the following:

a) In weak alkaline medium

REACTION POTENTIAL

(1) $H_2O_2 \longrightarrow H^+ + HO_2^-$

(2) $HO_2^- + H_2O + 2e^- \longrightarrow 3OH^-$ .... + 0.87 v.

$\underline{Ag} \longrightarrow Ag^+ + e^-$ .... – 0.79 v.

+ 0.08 v.

b) In acidic medium

REACTION POTENTIAL

$H_2O_2 + 2H^+ + 2e^- \longrightarrow 2H_2O$ .... +1.77 v.

$\underline{Ag} \longrightarrow Ag^+ + e^-$ .... –0.79 v.

+0.98 v.

It is seen that in both cases the total reaction potential is positive; therefore the reactions proceed spontaneously, The hydrogen peroxide (i.e. oxidizing agent) has sufficient oxidation potential to oxidize the metallic silver which constitutes the latent image.

Several experimenters have investigated this effect. Lüppo-Cramer[6] noticed the effect of chromic acid in depressing sensitivity when it was added to the liquid emulsion during manufacture. Sheppard and Mees[7] showed that sensitivity is greatly reduced by immersion after exposure in N/50 chromic acid solution. Perfilov[8] used the mild oxidizing action of chromic acid to obtain fission tracks in the presence of a high alpha-particle track background. Clark[9] found that action of chromic acid on silver bromide emulsion grains causes a reduction in sensitivity.

Another effect should be mentioned in relation with oxidizing agents. If an emulsion which shows a normal Herschel effect is treated with chromic acid after the actinic exposure, the surface latent image on the grains is reduced, and subsequent exposure causes an increase in developed density. This is called the Debot effect.

We have discussed that when a latent image speck absorbs red or infrared radiation, it loses an electron, which is then trapped at another site. There it reacts with a silver ion to form a fresh silver center. If the new center is also on the surface of the grain, there may be no change in developability, but if it is in the interior of the grain, it cannot be developed in a normal developer and a photographic reversal is obtained. This is the normal Herschel effect.

However, latent image specks in the interior of the grain also absorb light and are redistributed. If they are redistributed to the surface of the grain, the developed density may be increased (a positive Herschel effect). By using chromic acid to destroy the surface latent image, the normal reversal is eliminated and the positive Herschel effect is observed as the Debot effect.[8] This is the most convincing evidence of redistribution.

c. HIGH HUMIDITY

A major function of the gelatin in which the silver halide particles are suspended is to afford colloidal protection by insuring the stability of the latent image and the crystal.

At high humidities, however, the water in the atmosphere eventually forms a complex structure with the gelatin, and reduces its ability to provide this colloidal protection.

Water, by itself, is a relatively good acceptor of electrons; the gelatin, on the contrary, is a very poor conductor, is not ionized and does not become ionized with facility.

Therefore, it seems reasonable to attribute to the water, rather than to the gelatin, the role of electron acceptor when electrons are ejected in the presence of high humidity.

$$e^- + H_2O \longrightarrow H + OH^-$$

Electrons trapped in this reaction will not contribute to the photographic process. Obviously, this reaction increases with increasing humidity.

This explanation holds for the prevention of latent image formation, i.e., where the emulsion is always stored at high humidity, both during light exposure and after it.

In the case of a latent image which has already been formed, its fading or complete erasure is due to the formation of a complex chemical structure between the water and the gelatin, depriving the latter of its protective quality towards latent image stability.

The above criterion is generally agreed upon.[10]

The two reactions taking place are:

$$Ag \longrightarrow Ag^+ + e^-$$

$$2\,H_2O + 2e^- \longrightarrow H_2 + 2OH^-$$

Experiments have been performed by Wiener and Yagoda[11] in which they used air saturated with water vapor as a means for eradication of accumulated background due to incident cosmic radiation or internal radioactive impurities. The same authors demonstrated that the background could be destroyed by exposing the plates above 3% H_2O_2 at 25°C. With plates having coatings 25 - 50 μ thick, the process was completed in about 4 hours. However, when the method was applied to thicker emulsions (100-200 μ), the oxidation process had to be extended in order that the vapor had the opportunity to diffuse through the entire layer. The prolonged oxidation ($\approx$16 hr.) caused a reduction in sensitivity as evidenced by tracks of alpha-particles subsequently produced in the dry emulsion which developed with reduced grain density. This partial desensitization was avoided by eradicating the plates with a milder oxidant (i.e., moist air), conducting the operation at 35 $\pm$ 2°C to accelerate the process. No noticeable variation in sensitivity between eradicated and unradicated plates over the first 200 μ range was reported. Beyond 200 μ, the radicated plate exhibited a slight desensitization.

Experiments have been conducted at CCSD on the effect of high humidity on the erasure of the latent-image.[12] Selected for the experiment were Kodak film types Plus-X and Tri-X. The effect of humidity after irradiation was tested by developing film samples at regular intervals for subsequent densitometric readings of the density. The relative humidity was 70%. The temperatures used were 55°C and room temperature (~25°C). The data obtained is tabulated in Table II.

Table II

EFFECT OF HIGH HUMIDITY ON LATENT IMAGE DENSITY

Kodak Plux-X Film
Storage Temperature = 55°C, Relative Humidity = 70%

Time (Hours)	Optical Density
12	0.98
36	0.86
58	0.93
82	0.83

Kodak Tri-X Film
Storage Temperature = 25°C, Relative Humidity = 70%

Time	Optical Density
12	1.0
58	0.89
105	0.91
130	0.84

It can be seen that the experimental results are in accord with the theory. However the fading effect is also dependent upon other parameters such as emulsion composition and type of source radiation, which must be investigated in combination with environmental conditions after exposure.

II. STORAGE METHODS

a. HIGH HUMIDITY STORAGE

This is actually a corollary to the high humidity erasure method described previously. In the former case, the already formed latent image is erased; in this case the image is prevented from forming. Experiments on humidity storage of photographic films have been performed by M. Ehrlich.[13] She irradiated films with a Co^{60} source, each film being at a different relative humidity in a sealed glass jar. Four different relative humidities were obtained by using a small amount of saturated salt solution that produced a known relative humidity within the jar. At high relative humidities and low temperature she found that excessive fading of the latent image was produced, while at higher temperatures emulsion fogging was the result.

b. MANUFACTURING THE FILMS IN SPACE

Manufacturing films in space would be the best method, theoretically, to avoid film degradation. The film would be manufactured just before the desired light exposure and developed immediately after.

However, the steps for film manufacturing include:

1. Precipitation of the silver halide in a dilute gelatin solution.

2. Ripening of the precipitate.

3. Addition of fresh gelatin to allow it to set to a firm gel upon cooling and standing.

4. Washing the emulsion, shredding it into small pieces and rewashing to free the emulsion from soluble salts.

5. Melting the emulsion and keeping it for some time at controlled temperature. This is called the after-ripening period.

6. Coating the emulsion on the support.

This procedure seems to be too complicated and detailed to be carried out in space. In-house studies at CCSD[14] concluded that a complete manufacturing process in space would be impractical. However, the study also pointed out that if the initial stages of the manufacturing up to the after-ripening period were performed in the earth laboratory, it should be possible to perform the after-ripening and coating steps in space.

It is during the after-ripening stage that sensitizing substances are added to the emulsion. This has little effect on the size of the crystals of silver halide but results in a marked increase in their sensitivity to light. The absorption of sensitizing substances by the silver halide crystals brings about the formation of a few "sensitivity centers". The mechanism by which these centers increase the sensitivity of emulsions has been explained by several authors.[15]

Consequently, CCSD has proposed[14] to obtain several types of emulsion suitable for space application at the after-ripening stage of manufacturing and store them under various experimental conditions and storage times. After storage, the manufacturing process will be completed. The film will then be exposed and examined to detect any differences, such as fogging,inhomogeneity or non-uniformity in the image, etc. due to the break in the manufacturing process.

In order to test the efficiency of this method it has been proposed to expose both partially and fully manufactured films to the same dose of gamma radiation, complete the manufacturing process for the partially manufactured film and expose both films to the same light source. Upon development any differences would be noted.

c. DEVELOPING FILM IN SPACE

There are two basic types of development: chemical and physical.

Physical Development

A physical developer is a solution composed of a weak reducing agent, a pH regulator for the acid region, and a source of silver ions, usually silver nitrate. The reducing agent is not powerful enough to reduce the grains of the emulsion but can slowly reduce the silver salt in solution. Eventually the emulsion grains are also deposited as metallic silver. Studies have been conducted to determine the size of the silver specks which are required to initiate the deposition of silver from the physical developer; the conclusion was that four atoms were sufficient.

One of the theories proposed to explain the mechanism is Ostwald's supersaturation theory. "It is supposed that the developer reduces some of the silver ions to metallic silver; the metallic silver remains in solution in the supersaturated state. This supersaturation prevents the futher reduction of silver ions. In the present of metallic silver specks the supersaturated solution of silver deposits metallic silver, which relieves the saturation and allows more silver to be reduced, etc."[16]

Other theories explain the mechanism of physical development as an adsorption of the silver ions to the specks.[16]

Chemical Development

A chemical developer is a solution containing a developing agent and a buffer to establish an alkaline pH. Chemical development is usually carried out at a pH in the 8 to 13 region, where the reducing agents are most powerful. In chemical development the silver which forms the developed image comes from a reduction of the individual silver halide grains.

This is the conventional way to develop a photographic film or plate; physical development is very seldom used. The chemical developer solution reduces the silver ions to silver in a selective way; that is, the exposed silver halide grains are reduced at a greater rate than the unexposed. If development is extended over a long period of time, however, all grains are developed. After development, the film is rinsed, fixed, washed and dried.

A method of efficient agitation is required, since during development, products are formed which retard the developing process. Only by proper agitation can uniform development be assured. One method used has been nitrogen-burst agitation; for small systems a flat tray has been used which is manually rocked; brushing of the emulsion, slowly and continuously during development with a soft camel's hairbrush of greater width than the plate has produced uniform results. The best method for space application remains to be selected.

After developing, a number of silver salts remain which, due to their slight solubility in water, cannot be removed by simple washing. Fixation consists in addition of a fixing agent which forms complexes with these insoluble salts rendering them water-soluble. Another possibility consists in the conversion of the insoluble salts to colorless non-light sensitive compounds.

Very recently an entirely different process of development and fixing has been developed by Kodak Laboratories: the Bimat Transfer Processing System. The process is based on the diffusion transfer mechanism. The Bimat Transfer Film is soaked in a special imbibant which is absorbed by the gelatin layer on the Transfer Film. When the exposed negative is placed in intimate contact with the presoaked Bimat Transfer Film, the solution begins to diffuse into the emulsion of the negative film. Exposed negative grains begin to develop, and both unexposed and exposed negative grains begin to dissolve. Some of the dissolved silver halide diffuses into the Bimat Transfer Film where it is reduced to silver on the nuclei present and forms a positive image. This positive can be ejected from the camera/processor mechanism without damaging the negative.

All the possibilities mentioned should be investigated carefully; any advantages or disadvantages noted and the optimum method for space applications selected.

d. LOW TEMPERATURE STORAGE

At low temperatures the mobility of electrons is affected very little, but that of ions is reduced considerably. This phenomenon affects photographic process in the following way: at low temperatures the electrons produced by light exposure move until they are trapped; the silver ions, however, remain relatively immobile until the temperature of the emulsion is increased. Latent image formation, therefore, does not occur until the temperature of the emulsion is increased.

Exposure at low temperatures produces a very dispersed latent image since many photoelectrons are liberated and none is trapped by silver ions. Upon warming up the emulsion only those electrons trapped in the grains contribute to latent-image formation.

Investigations performed on the temperature-dependence of the photographic process lead to the following conclusions:

1. The sensitivity of photographic emulsions to x- or gamma rays is a linear function of the temperature during exposure.[17]

2. In general, films increase in speed with increasing temperature.

3. The effect is independent of the temperature of the film before and after exposure.[18]

4. The linear relationship between sensitivity and temperature holds only down to temperatures of approximately -80°C.[19]

5. Below −253°C (liquid hydrogen temperature) the sensitivities of photographic materials to x-radiation are essentially independent of temperature.[19]

6. At the temperature of liquid helium, (4°K), the ratio of x-ray sensitivity to light sensitivity becomes much higher than the same ratio at room temperature.[20]

From these results it seems possible to prolong film life in space missions by storing them at low temperature, letting them warm up just prior to the desired exposure; and re-storing them at low temperature after the light exposure. Variations in optical characteristics such as sensitivity and resolution associated with this method remain to be investigated.

The following data on the temperature dependence of photographic films have been obtained at CCSD.

Two groups of Kodak 103A-O film were exposed to the same amount of radiation -- one group at dry ice temperature and the other at 74°F. They were then kept for a period of time and afterwards developed. The optical density was noted in each case.

The data is tabulated in Table III.

Table III

CHANGE IN OPTICAL DENSITY WITH CHANGE IN TEMPERATURE

Exposure (Rads/Air)	Density		% Change in Density with Temp. Increase
	Film at Temp. of Dry Ice (−78°C)	Film at 74°F (23°C)	
1.37	0.33	0.36	+ 9.0
2.73	0.60	0.58	− 3.3
5.47	0.83	0.94	+13.2
13.66	1.32	1.42	+ 7.5
27.33	1.74	1.86	+ 6.8
54.66	2.06	2.16	+ 4.8
82.0	2.26	2.31	+ 2.2

e. STORAGE UNDER THE INFLUENCE OF AN ELECTRIC FIELD

Pulsed electric fields have been used to study the motion and trapping of photoelectrons. Hamilton, Hamm and Brady[21] have used simultaneous pulses of light and voltage to study the motion of electrons and holes in photographic grains. The latent image centers were detected by giving the grains a brief development, which enlarged them, and then examining them under the electron microscope.

Exposures made with no applied field showed either a completely random distribution of photolytic silver or some pattern of orientation corresponding to the crystalline grain geometry. In the presence of an applied field the photolytic silver was asymmetrically distributed toward the side of the grain to which the electrons should have been displaced, i.e., the direction opposite to that of the field or positive electrode. Concluding, both photolytic silver and developable centers of the latent image are asymmetrically located in the direction of displacement of negative carriers by a pulsed electric field during exposure. The mechanism is essentially the same for latent-image formation as for the print-out effect, namely, the liberation of an electron from a bromide ion into the conduction band of the crystal, its subsequent trapping, and finally its neutralization by an interstitial silver ion to form a silver atom. The chemical reactions involved are the following:

$$2Br^- \longrightarrow Br_2 + 2e^-$$

$$Ag^+ + e^- \longrightarrow Ag$$

Based on the above experiment, and several other with similar results, the possibility has been considered of applying an alternating field to sweep the electrons back and forth in the crystal, therefore reducing the probability of their combination with a silver ion to form a silver atom. This method could reduce or completely prevent the formation of an undesired latent image.

Another method to be considered is the inhibition of mobility of interstitial silver ions by the application of a constant electric field.

In accordance with the Gurney-Mott Principle, mobile interstitial silver ions are attracted to electrons which have been trapped about sensitivity specks. It is the formation of silver atoms by this process which produces the image. A constant electric field applied to the film would sweep and hold these mobile interstitial silver ions to one side of the crystal in each grain and sweep any free electrons in the opposite direction (and presumably prevent collecting about a sensitivity speck). Since confinement restrictions require that silver atoms be formed only at sensitivity specks, the formation of any image would be inhibited as long as the constant electric field exists.

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