

FINAL REPORT

Passive Particle Dosimetry

Charles B. Childs

Department of Physics and Astronomy

University of North Carolina at Chapel Hill

Chapel Hill, North Carolina 27514

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ABSTRACT

A brief review of the present methods of dosimetry is given, followed by a review of the processes operating which enable silver chloride crystals to be used for ionizing particle dosimetry. There were differences between various crystals ability to record ionizing particle paths, with the differences being directly related to impurities in the range of a few ppm (parts per million). To understand the roles of these impurities in the process, it was necessary to develop a method for consistent production of high purity silver chloride, as well as silver bromide. The developed method yields silver halides with detectable impurity content less than 1 ppm, and this high purity silver chloride was used in growing crystals with controlled doping. Crystals were grown by both the Czochalski method and the Bridgman method, and the Bridgman grown crystals were used for the experiments discussed in this report. In addition, the distribution coefficients of ten divalent cations were determined for the Bridgman crystals. It was found that the best dosimeters were made with silver chloride crystals containing 5 to 10 ppm of lead; other impurities tested did not produce proper dosimeters. Since this research was concerned with making tracks observable by sweeping in photoelectrons, a comparison was made of this decoration method with that of decoration by actinic light, a method developed by Schopper and Haase. A table is included which includes the unique properties of each of these methods.

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Part One

Silver Chloride Dosimetry and Experiments Prior to 1970

This research was performed to advance knowledge of silver chloride crystals as a new type of dosimeter by decoration of radiation induced imperfections. Since these decorated imperfections are similar to tracks in nuclear emulsions, the following are comparisons which summarize the unique features of silver chloride crystals for radiation detection:

1. Thickness of crystals can be greater than for emulsions, a characteristic important for hypernuclei studies;
2. Decoration of 5000 μ thick crystals requires less than two hours at room temperature whereas development of 600 μ emulsions requires more than 12 days of careful development at different temperatures;
3. There is saturation of track decoration in crystals so that excessive decoration does not increase decoration of tracks; continual development of emulsions produces profound changes in track structure.
4. Multiple scattering measurements can be made with high accuracy in crystals because no distortion is introduced by decoration whereas emulsions undergo various distortions during and after development;
5. Absence of hygroscopic sensitivity enables crystals to be exposed to various environments without fear of density or thickness changes;

6. Crystals should be of particular use in studying heavy primaries in high flux of singly charged, minimum ionizing particles, (i.e., relativistic protons whose low energy loss does not produce tracks which can be decorated);
7. Silver chloride refractive index of 2.07 (5890A) increases microscope working distances by 35% over than in G-5 emulsions with index of 1.53;
8. Silver chloride stopping power is almost 100% greater than standard G-5 emulsions.

Among present detectors, that closest related to silver chloride is the nuclear emulsion; consequently, this report contains a short survey of emulsion composition, development, limitations, and radiation properties. In addition, to explain how the crystals function as detectors, we have included a review of radiation effects in solids. Latest results from experiments on detection of radiation by silver chloride crystals are discussed, as well as many possible experiments with potentially important ramifications.

* * *

Although it is impossible to predict what benefits will result from the previously proposed experiments, several potential benefits are readily visualized. For example, there is an interest in studying the heavy primary component of cosmic rays during solar flares but nuclear emulsions "blacken" during such flares. Silver chloride crystals, however, are "insensitive" to the low energy loss protons from such flares but they do record the heavy primaries. In addition, exposures of crystals in recoverable satellites could be for longer periods than for emulsions.

I. INTRODUCTION

The detection of ionizing radiation is of major importance because it provides vital information in both the biological and physical sciences. The methods used to study radiation have employed two general types of detectors. In the first type are those instruments which require auxiliary electronic equipment for registration of radiation whereas the second type does not need any auxiliary equipment for recording particles. Among the first type are gas-containing instruments such as ionization chambers, geiger counters, and proportional chambers. The cloud chambers and "light chambers" are also filled with gases which permit determination of trajectories of ionizing particles. Cerenkov counters use both gases and liquids whereas scintillation detectors primarily utilize solids. The bubble chamber is already well established, and the spark chamber is rapidly establishing itself as a reliable and unique detector. Solid state detectors, in comparison, can be very small, thereby giving them many advantages under certain conditions.

Detectors of the second type consist of materials in which particle trajectories are permanently recorded. Included in this type are electron microscope specimens, nuclear emulsions, and silver chloride crystals. The electron microscope technique possesses such limitations as being restricted to samples less than about 1000A thick^{1,2}, detection variation with incident angle³, and inability to utilize conventional optical microscopes for examination of large areas and volumes. In contrast with this technique are nuclear emulsions and silver chloride crystals which do enable microscope studies of radiation tracks over extensive areas and volumes.

II. NUCLEAR EMULSIONS

One of the most widely used methods of particle detection is that in which trajectories are made visible by development of nuclear emulsions.

Some of the many important discoveries which were made in emulsions have been reviewed by Shapiro⁴, Yagoda⁵, Goldschmidt-Clermont⁶, Demers⁷, Blau⁸, Powell, Fowler and Perkins⁹, and Barkas¹⁰. Because of the relationships between certain aspects of emulsions and silver chloride crystals, we will briefly summarize several emulsion properties such as composition, formation and trajectories (tracks), and development. Also, certain measurements performed will be mentioned because of the potential extension of silver chloride crystals.

A. Composition, Track Formation, Developing

Nuclear emulsions consist of a suspension of silver bromide-iodide crystals in gelatine. The silver bromide crystals range from about 0.07 μ diameter in Gavaert NUC 3.07 to 0.28 μ in the Rian P-R NIKfi Hr, R and MP emulsions.¹¹ Various additions, such as silver iodide and sulfide, are required to make the emulsions sensitive, and included among the sensitizers is the complex organic gelatine itself. Penetration of an emulsion grain by a charged particle causes positive hole and electron formation within the grain. The hole, with the lower diffusion range, can diffuse to the surface where it may be trapped by halide ions which, when at the surface, are deep hole-traps. This results in an excess of positive charge at the surface, with the excess being in the form of a silver ion. The positive silver ion can migrate along the surface until it becomes trapped by a sensitizing center composed of two or three surface silver atoms. This resulting positively-charged cluster may then capture the electron which was placed in the conduction band at the same time that the hole was created, thereby adding a silver atom to the cluster. After this process has added one or two silver atoms to the surface clusters of silver, the whole grain can be reduced to silver by a chemical developer.¹² The path of the particle is thus delineated by microscopic silver specks.

The most important step in making tracks visible in emulsions is the chemical development. Since the "degree" of development is a function of both temperature and duration of development, the grain density of tracks depends upon the processing. To utilize "thick" emulsions, various temperature-cycle methods have been devised but in all these methods there must be careful control of developing temperature, solution pH, and drying. Barkas^{13,14} gives a complete review of these factors. As examples of time required for processing emulsions, the commercial 600 μ thick Ilford G5 requires about 12 days⁶ while the Yagoda 1500 μ thick emulsions require more than 15 days.

It is well to point out here that the emulsion gelatine makes emulsions sensitive to relative humidity (RH).^{15,16} The emulsions will change dimensions and density with changes in relative humidity, necessitating a "standard" RH for emulsions.^{17,18} This is usually taken as 58% RH although experiments have been performed at other RH.¹⁹ Variation in density can be "considerable" and has been reported to be present even when stored at constant RH. The important thing is to know the density at time of exposure, and such accurate data is sometimes difficult to obtain. For example, in going from an environment of 58% to "vacuum," about a month is required for the "density" to stabilize whereas less than a day is required when going to 85% RH.²⁰ Because of the humidity effect on emulsions, it is essential to have precise measurements of density for every "batch" at various RH to determine the emulsion stopping power.

B. Particle Properties Obtained From Emulsions

The primary purpose of nuclear emulsions is to enable measurement of the charge, velocity, and mass of the particle. These properties are generally

obtained from grain density, delta-ray distribution, and scattering. And all are related to the energy loss per centimeter of the particle. The energy loss, therefore, is an important characteristic of any particle.

Energy loss is basically determined by either grain or blob density (B), depending upon the rate of energy loss. For low energy losses, the grain density is sufficient but for high losses, accurate measurement of grain density becomes difficult because of overlapping of adjacent grains, unresolvable adjacent grains, and saturation for highly ionizing particles. The blob density (i.e., the density of clusters consisting of one or more developed grains) overcomes these difficulties of grain density. Barkas showed that the grain density consists of a primary part and a secondary part,²¹ the latter being grains not penetrated by the particle but made developable by secondary processes such as delta rays. From established energy loss theories, it is possible to determine charge and velocity from blob densities, but highly ionizing, multiply-charged particles present some difficulties because of saturation of grain density. In those situations, measurement of the delta rays leads to determination of the charge of particles with known velocities. Various methods have been advised to measure the density of these delta rays, despite the difficulties when the charge is greater than about eight.

Charge determination for non-relativistic particles is more difficult than for relativistic particles; however, with the non-relativistic particles, Kristiansson, Mathiesen and Waldeskog²² reported that mean track width (MTW) and slope of (MTW) vs residual range could provide sufficient data for charge identification, provided that the track length be greater than 25 μ and momentum less than a certain value. The "little delta rays" technique of Alvial and associates²³ and studies of range-energy and effective charge

may also be employed in charge determination. O'Brien and Noon²⁴ made a comparison of four independent methods for charge determination and found that they gave comparable results.

The particle velocity can be derived from small angle scattering. This scattering is explained by several theories, the most widely employed being those by Williams,²⁵ Goudsmit and Saunderson,²⁶ and Moliere.²⁷ The important conclusion of these theories is that the arithmetic mean deflection $\bar{a}$ of a particle from a straight trajectory is

$$\bar{a} = \frac{KZt}{pB}^{1/2}$$

Z = particle charge

p = momentum

B = ratio particle velocity to light velocity

t = thickness of material penetrated in scattering $\bar{a}$

where K is a "constant" which varies slightly with velocity and scattering medium thickness. For multiply-charged particles, Z is replaced by $Z(Z + 1)$. Fechtel and Friedlander²⁸ found that for $Z \geq 4$, K does not change with Z and velocity effect on K is negligible.

C. Emulsion Limitations

While emulsions are a reliable experimental medium, they do possess characteristics which complicate and limit their utilization:

1. density and thickness vary with changes in relative humidity;
2. background accumulation restricts storage time;
3. sensitivity at various temperatures depends upon type of emulsion and irradiating particles;

4. fading of tracks is related to sensitizing agent, relative humidity, temperature, and time [i.e., prior to development, G-5 emulsion can undergo changes which result in an apparent particle mass of 0.54 of its true value when stored for two months at 20°C and 75% relative humidity];
5. spurious scattering²⁹ restricts usefulness of scattering measurements to a few BeV per nucleon.³⁰⁻³⁴

We have listed some of the particle parameters, such as charge and velocity, which are obtained from nuclear emulsions, along with certain difficulties and limitations of emulsions. We will now proceed to discuss briefly radiation effects in solids and to illustrate how decoration of such damage in silver chloride results in a new radiation detector which may be considered the complement of nuclear emulsions.

III. RADIATION EFFECTS IN SOLIDS

Since the 1949 review of radiation effects in solids by Seitz,³⁵ there have been a number of similar papers which serve as both reviews and sources of basic data.³⁶⁻⁴⁸ We will refer only to those which are pertinent to this report.

When a material is penetrated by a heavy charged particle, the particle loses most of its energy by two methods. First, it may lose energy by coulombic interaction (elastic collision) of its nucleus with that of nuclei in the material. In such elastic collisions, the electrons play no role other than partial screening of the nuclear charge. Second, the particle may lose energy by direct interaction with electrons which results in their being placed in excited states (inelastic collisions). Of these two mechanisms, the dominant one is loss by inelastic collision, with the rate of loss being a function of the charge, velocity, mass of the particle, and of density of the penetrated material.

The electronic excitation, resulting from direct interaction between the atomic electrons with either the incident particle or from primary recoil atoms, may produce what Seitz and Koehler have termed "thermal spikes."⁴³ These spikes are regions in which the localized energy loss is mainly converted into heat.

In these thermal-spike regions, material is heated to several hundred degrees and then thermally quenched. This heating process takes place in less than 10^{-10} seconds and forms "large" concentrations of point defects which may form stable clusters during the subsequent rapid cooling. In addition, these defects can produce a disordering which causes a volume change ΔV . The volume change depends upon the number of defects, their volume, and atomic rearrangement resulting from thermal quenching. This ΔV plus the intense temperature gradients produce a stress field which results in plastic flow near the spike and thereby forms permanent imperfections (dislocations) at distances much greater than the radius of the molten core of the spike.

Another type of spike concept has been formulated by Brinkman.⁴⁹ He proposed that since the time of the molten state is greater than relaxation time, there is sufficient strain energy, released after density fluctuations have relaxed, to raise the temperature even higher and thereby extend the period of existence of the liquid state. This resulting temperature extension produces turbulent motion so that most of the atoms will occupy new lattice sites. Such a region which has undergone melting and resolidification is a "displacement spike."

As recently surveyed by Seitz,⁵⁰ the present explanations of radiation effects in terms of either the thermal or displacement spikes are far from

satisfactory. It is not known whether a possible combination of these spikes exists or whether an entirely new description of radiation displacement by heavy particles must be formulated. However, some experimental results have been interpreted in terms of thermal spikes such as that by Piercy,⁵¹ and in terms of displacements spikes such as that by Beevers and Nelson.⁵² These latter results differed from the displacement theory prediction by a factor of 10,000. Despite the obvious lack of agreement between the various spike models and experimental results, the work of Chadderton⁵³ appears to be leading toward an answer to the basic question as to whether spikes are composed mainly of vacancies or interstitials. In any event, regardless of the radiation damage process, one expects the track of a heavy charged particle to be surrounded by a cylindrical region of intense thermal strain. In a soft material such as silver chloride, such a region would be highly dislocated, and hence contain a high density of electron traps. It is this circumstance which provides the basis of track decoration by the method to be employed in the proposed experiments.

IV. DECORATION OF IMPERFECTIONS

The essential act in using crystals for radiation detection is making microscopically visible the radiation-induced imperfections. The method used to achieve this is called "decoration."

Decoration is a term which refers to making crystal imperfections microscopically visible by deposition of material at the imperfections. The decoration can then be observed when the deposited material is sufficiently opaque to illumination transmitted by the crystal. For practical studies of radiation-induced imperfections, the decoration should be observable with a conventional microscope using visible-light illumination. Such conditions have been employed in studying decoration of inherent imperfections in two groups of crystals: alkali halides and silver halides.

Since this report concerns the use of silver chloride for radiation studies, we will review decoration in both groups and explain why silver halides are preferred for radiation detection.

A. Alkali Halides

Methods for decorating alkali halides⁵⁴ have been developed by Amelinckx, van der Vorst, Geners and Dekeyser,⁵⁵ by van der Vorst and Dekeyser,⁵⁶ and by Barber, Harvey and Mitchell.⁵⁷ Each of these methods requires that the crystal be first doped with either Na, Ag, or Au and then annealed for several hours at temperatures greater than 500°C. A rapid cooling to room temperature from these high temperatures produces a supersaturation of the doping material which condenses at the imperfections. In the first method, decoration is obtained only within a region 1/2 mm thick. A disadvantage of such methods is that the necessary anneal may modify or remove imperfections such as radiation damage tracks.

B. Silver Halides

There have been three techniques reported for decoration of silver halides (AgBr and AgCl). One of these techniques was developed by Bartlett and Mitchell⁵⁸ who exposed doped crystals to ultraviolet light and then annealed them to obtain decoration. Since here, as with the alkali halide decoration, there is an anneal which would affect radiation-induced imperfections, we will consider this technique as improper for radiation detection. The two remaining methods produce imperfection decoration at room temperature, thereby being of primary interest for radiation studies by decoration.

Another procedure for decoration of a silver halide was formulated by Hedges and Mitchell.⁵⁹ Their initial decoration was obtained by merely exposing AgBr to light in its long-wave edge absorption band at room temperature. They

also decorated within 10 microns of the surface with exposure to tungsten filament lamps and mercury lamps. One interesting observation was that the silver specks (decoration) did not grow indefinitely but reached a maximum size.

In pursuing this technique, Jones and Mitchell found decoration dependent upon individual properties of the crystals⁶⁰ while depth of decoration varied with exposure duration.⁶¹ Parasnis and Mitchell⁶² studied copper-doped silver chloride but their results did not settle the problem of impurity effects on decoration, as illustrated by silver formation in silver bromide.

While the Hedges-Mitchell method does produce decoration to a depth of $\sim 30\mu$, it is far from providing decoration for depths approaching the thickness of the widely used 600 μ G-5 emulsions. This limitation of decoration depth at room temperature was overcome in a manner recently reported.

In 1960, it was found that room temperature decoration in silver chloride crystals could be obtained when photoelectrons were swept into the crystals.⁶³ The interesting factors here were the low temperature of decoration and large thickness (5000 μ) of crystals; thus were solved the problems of high temperature anneals for decoration in alkali halides and the shallow decoration depths of the Hedges-Mitchell technique for silver halides.

The technique of sweeping photoelectrons into silver chloride crystals was first developed by Haynes and Shockley in 1948.⁶⁴ Later modifications were made by Hedges and Mitchell^{59-b} in 1953, Brown in 1955,⁶⁵ Webb in 1955,⁶⁶ Hamilton, Hamm and Brady⁶⁷ in 1956, Süptitz⁶⁸ in 1958. It was not until 1960, however, that this technique was found capable of producing decoration of imperfections within the volume of large crystals.⁶³ The reason for failure to observe decoration in the earlier studies is two-fold: first, the earlier investigators had considerable difficulties in producing consistent data from

adjacent crystals from the same boule; second, no consideration was given to minute amounts of impurities present in several parts per million. It was the difficulty in producing consistent data which caused the general abandonment of this technique before any consideration was given to the second factor of impurities. Whereas earlier investigators had obtained inconsistent results for crystals from the same ingot, Childs⁶⁹ reported that those failures were caused by a severely deformed surface region containing an extremely high density of electron traps. He showed that careful removal of this deformed region resulted in good penetration of photoelectrons in all crystals. He later showed that ability to decorate depended upon the impurity content of the crystals and that crystals with greater than about 10 ppm of Al, Fe, and Ni would not decorate.⁷⁰ These elements, notably nickel, were assumed to be very good electron traps, an assumption in agreement with earlier studies by Michel and Sliker (to be discussed below).

Paths of charged particles have been made microscopically visible by sweeping photoelectrons into silver chloride at room temperature and we will present a brief description of that method and some of its results.

V. DECORATION OF RADIATION INDUCED IMPERFECTIONS OF CHARGED PARTICLES IN SILVER CHLORIDE

A. Experimental Methods

In 1948, Haynes and Shockley performed unique experiments in which they measured various electronic properties of silver chloride.⁶⁴ In these experiments, the crystal was placed between two quartz plates with outer surface of an electrically conducting, ultraviolet transmitting thin film. A potential of several kilovolts charged the resulting capacitor, the crystal being the major dielectric. At the end of the charging cycle, the internal field

was zero, having been cancelled by ionic charge migration within the crystal. This external field was removed and an ultraviolet light flashed. The internal field immediately became several kilovolts/cm by virtue of the surface charges, with a time constant for decay of a few hundred microseconds. The light duration of one microsecond and electron lifetime of several microseconds were much less than this time constant; therefore, with the light being absorbed within one micron of the negatively charged surface, the photoelectrons were swept toward the opposite positively charged surface. Since the original work of Haynes and Shockley, various forms of apparatus to achieve the same effect have been constructed by Hedges and Mitchell,^{59-b} Brown,⁶⁵ Webb,⁶⁶ Hamilton, Hamm and Brady,⁶⁷ Süptitz,⁶⁸ and Poubeau and Schmitt.⁷¹

The decoration mechanism is as follows. A quantum forms a chlorine atom and an electron near the surface, from which the chlorine escapes. The electron, driven into the crystal by the polarization field, becomes trapped at an imperfection. An interstitial ion, with a jump frequency of about 10^{10} per second at 300°K,⁷² joins with the trapped electron, thereby forming a silver atom. This atom may trap another electron, with the process repeating to form microscopic coagulation of silver atoms. An explanation for continuation of this process was reported by Seitz. He showed that dislocation jogs can have a net charge $|e/2|$. Hence, if the initial charge is $+e/2$, the imperfection may trap an electron and thereby have a charge $-e/2$. This $-e/2$ charge may in turn trap an interstitial silver ion. This leaves the imperfection site with a silver atom of charge $+e/2$, the initial charge of the imperfection, so the process of silver coagulation continues through charge cycles of $e/2$.

B. Decoration of Imperfections Induced by Primary Cosmic Rays, Proton Collisions, and Alpha Particles

As mentioned, in 1960 it was shown that photoelectrons which were swept into silver chloride could result in dislocation decoration. The apparatus⁶⁹

used for these experiments provided potentials of two kilovolts pulsed at a rate of 1000 per second. The light flash provided 10^9 photons per cm^2 at the crystal. With this apparatus, the first decoration of radiation tracks in silver chloride was obtained in 1961,⁷³ followed by additional data in 1962⁷⁴ and 1963.⁷⁵ Particles produced by 1.55 BeV proton collisions with Ag and Cl nuclei have also been detected.⁷⁶ The decoration of these tracks was obtained in less than two hours in crystals up to 5 mm thick. Schmitt recently succeeded in decorating tracks of 5.3 MeV alpha particles.⁷⁷

The appearance of tracks in silver chloride are similar to those in Ilford G-5 emulsions. Fortunately, tracks are preferential electron traps in well-annealed crystals and consequently decorate in preference to random dislocations. This observation is in agreement with the conclusion of Seitz that dislocations are not good electron traps in well-annealed silver chloride crystals.⁷²

By comparisons of heavy primary cosmic ray tracks in Ilford G-5 emulsions with the same particle tracks in adjacent silver chloride crystals, it was found that apparent relative track widths were preserved, along with definite differences in granularity. There is a distinct increase in both track width and granularity for particles stopping in the crystals.

Because of the well-known photosensitivity of silver chloride, we should mention that decorated tracks are stable to illumination. There was no qualitative change when decorated tracks were exposed to Wratten K2 filtered microscope illumination for 40 hours and to the noon-day sun for 30 minutes. Even in the self-decorating crystals to be discussed, there is expected to be a saturation of decoration.

C. Self-decorating Crystals

Whereas the technique of decoration by sweeping photoelectrons into silver chloride crystals is very simple, the ideal decoration method would be merely to expose the crystal to light, similar to the Hedges-Mitchell method but with decoration at depths greater than a few tens of microns. Speculation on the composition and decoration of such self-decorating crystals was published in 1963.

The advantages of such self-decorating crystals are obvious, and we find encouraging the unpublished observation that dislocations have been decorated 500 microns in a crystal containing about two ppm Ni. That some decoration was obtained in the presence of this Ni is not surprising because Moser has pointed out that the Ni traps are first filled, leaving further electrons to be trapped by the imperfections which are not as good electron traps as nickel.⁷⁸ It was estimated that properly doped silver chloride crystals will decorate within 10 minutes of actinic illumination.

Prior to the beginning of the research under this contract, there was considerable lack of knowledge about why some silver chloride crystals could be used for track decoration whereas in some other crystals only dislocations and precipitates could be decorated. While it was apparent that those differences might be due to impurity differences, high purity silver chloride with less than tens of ppm of cations was not available from any source. The first step in the research was, therefore, preparation of silver chloride of a purity which had not been previously produced, followed by using it in growing both pure and doped crystals, characterizing the impurities in the crystals, and studying the impurities' effects on the track decoration.

Part Two

Experimental Procedures and Results

I. Production of High Purity Silver Halides

Earlier research had shown that the ability to decorate tracks of ionizing particles in the crystals depended on both a minimum energy loss per unit length and a particular impurity. The first part of the research was, therefore, directed towards producing the high purity silver chloride in order to grow crystals in which there could be valid comparisons of track decoration in pure crystals with those containing controlled dopants.

A. Silver Halide Precipitate Production

The first stage of this program of obtaining high purity silver halides was the design and construction of equipment to produce high purity silver halide precipitates to be used in growing the crystals. The precipitates produced by this method and equipment do not contain any detectable cationic impurities within the detection limit of less than 1 ppm (parts per million mole fraction) for the 54 cations sought. These silver halide precipitates are the purest produced and not available from any other source. Although high purity silver chloride and silver bromide are produced in this first stage of crystal growth, the second state of crystal growth concentrated on only the silver chloride.

B. Silver Chloride Crystal Growth

Silver chloride crystals were grown by both the Czochralski methods and the Bridgman method, and details of the results with both methods follow.

The Czochralski method employed a stainless steel pulling furnace with a graphite (ultra high purity) heater with the charge in a platinum crucible. The temperature of the charge is controlled by a Pt-Pt-13% Rh thermocouple (between the heater and heat shield) connected to a proportional controller

(Model 624-A-RC10: Research, Inc.) providing the control signal for a SCR power controller (Model SPR 240/140: Research, Inc.). The output of the SCR was connected to the primary of a low voltage-high current transformer (Model 56-100: Signal Transformer Co., Inc.) whose secondary was connected to the graphite heater. After various positioning of the thermocouple, a location was found which enables all of the charge to be pulled in a crystal, with a maximum crystal volume of approximately 70 ml. The best crystals were grown with a seed rotation of 1 rev/minute and a pulling rate of one centimeter per hour. Figure 1 shows a mixed AgBr:AgCl crystal grown with these conditions.

The Bridgman method employs a large Bridgman furnace with the charge having been treated in a quartz crucible prior to its being sealed and placed into the Bridgman furnace. Growth took place by lowering the sealed crucible at 1 mm per hour. (Details on the procedures and the crystals are given in the reprint "High Purity Silver Bromide Crystals Containing Less than Several Parts per Billion of Iodide" attached to this report as Appendix 1.)

While the Czochralski method did produce oriented crystals with a growth rate ten times that of the Bridgman method, the Czochralski method did not produce crystals with uniform cross sections. Such crystals probably could have been produced with more elaborate and expensive equipment, but the additional cost, as well as other factors, did not justify such modifications in light of the Bridgman grown crystals. The Bridgman grown crystals had uniform cross sections of 2 cm by 2 cm, excluding the initial 3-4 cm of the tip where solidification of the melt began, with lengths up to approximately 18 cm. Since these Bridgman grown crystals had a square cross section and since added dopants were in the melt, these crystals were easily cut into specimens for the study of both dopant distribution in the crystals and dopant effect on track decoration.

All results in this report were obtained with Bridgman grown silver chloride crystals.

II. Methods of Chemical Analyses

The most fundamental aspect of the research was to determine the effects of various dopants and their concentration on track decoration. Such research required that accurate methods be employed for determination of both the impurities present in the starting material used to grow the crystals, the amount of added dopant actually in the crystal, its distribution in the crystals, and the valance state of the dopant. The analytical methods used to determine these crystal characteristics were: mass spectrograph, emission spectrography, atomic absorption spectrography, and electrical conductivity.

A. Mass Spectrographic Analyses

The mass spectrographic analyses turned out to be the most unreliable because of the unreproducibility of the analyses and because of lack of even qualitative agreement with other analytical methods. For example, mass spectrographic analyses at both the University of North Carolina and the Eastman Kodak Company differed by factors of tens, and, even within the same laboratory analyses sometimes differed by several factors. This lack of agreement and lack of reproducibility is attributed to contamination from several sources. Among those sources may be specimen preparation, small mass consumed in the analyses, and contamination from the spectrographs' pumping and electrode systems.

B. Atomic Absorption Analyses

The atomic absorption method requires a vapor lamp for the element being sought and a liquid in which the silver chloride has a "high" solubility at room temperature. Since a different lamp is required for each element being sought in the silver halide, this method would take considerable time and equipment to check for the 54 elements covered by the emission spectrographic analyses discussed later. In addition, the atomic absorption technique is also limited by the required standards and solvents for the silver chloride; however,

this method is useful when seeking small amounts of only a few impurities.

C. Emission Spectrographic Analyses

This method is the fastest, easiest, and least expensive of any of the present analytical methods to determine the presence of 54 cations. The typical detection limit for each of the cations is 1 ppm (one part per million weight fraction) and has an accuracy of approximately ± 1 ppm in the range of 1 ppm. The general policy for the commercial analytical laboratory is to state that some elements not detected are "less than 1 ppm." This policy does not interfere with obtaining reliable data for the presence of most divalent cation impurities which might be present in amounts less than 1 ppm.

D. Electrical Conductivity

The electrical conductivity of a silver halide crystal at room temperature may be represented as

$$\sigma = N (x_v \mu_v + X_i \mu_i) e \quad (1)$$

where

N = number of ion pairs of the silver halide
per unit volume

X_v = mole fraction of vacancies

X_i = mole fraction of interstitials

μ_v = room temperature mobility of a vacancy

μ_i = room temperature mobility of an interstitial

e = electron charge.

At room temperature, the intrinsic concentration of vacancies and interstitials are equal so the electrical conductivity becomes

$$\sigma = n_v \mu_v e \quad (2)$$

with n_v = number of vacancies per unit volume.

A divalent cation impurity, when present in low concentrations C introduces one free vacancy, to maintain electrical charge neutrality, so the electrical conductivity at room temperature becomes

$$\sigma = C\mu_v e \quad (3)$$

Equation 3 can therefore be used to measure low concentrations of divalent impurities since μ_v is a well known parameter of a silver halides:

It should be noted that σ can give reliable data on the variation of small amounts of divalent impurities whereas the previously described analytical methods have up to at least a 50% error when measuring the divalent impurities when at the few ppm levels. At the same time, it must be kept in mind that σ does not give any data on monovalent cation impurities, nor does it give reliable data on a divalent impurity content when that impurity has a high association with a vacancy, such as observed when cadmium is present above approximately 200-300 ppm.

III. Analyses of Precipitates

Each lot of silver halide precipitate was analysed for 54 cation impurities, and the first lots were also analysed at the Eastman Kodak Research Laboratories for eleven impurities by a special spectrochemical method. In the latter analyses, three lots of silver chloride were analysed, and the results are given in the following table.

Eastman Kodak Research Laboratories Analyses of Three
Lots of Silver Chloride

U. N. C. Lots	Impurity	Detection Limit (ppm)	Observed (ppm)
2, 3, 4	Cadmium	0.5	Not Detected
	Chromium	.03	Not Detected
	Cobalt	0.1	Not Detected
	Copper	.03	Not Detected
	Gold	1.0	Not Detected
	Iron	.07	Less than .07
	Lead	0.2	Not Detected
	Manganese	0.03	Not Detected
	Nickel	0.1	Not Detected
	Tin	0.1	Not Detected
	Zinc	4.0	Not Detected

Identical specimens of these three lots were analysed by Johnson Matthey Chemicals for 54 cations of which only iron was detected at 1 ppm. This iron content, while higher than that found by Kodak, is in agreement with the Kodak analyses when the Johnson Matthey error is included.

Johnson Matthey made emission spectrographic analyses of all lots of silver chloride precipitate made for this research, and no more than one impurity was ever observed at its detection limit in any lot of silver chloride.

IV. Analyses of Crystals

A. Pure Crystals

The purpose of having analyses made of the pure crystals was to determine if any detectable impurities were being introduced during the melt treatment and during the crystals growth. For example, the silver chloride melt is treated with chlorine which is supplied in a metal cylinder which reacts with slightly moist chlorine. That reaction produces such compounds as ferric chloride which can be carried by the chlorine as it leaves the cylinder. To help eliminate such particles and other impurities, the halogen is filtered and is given a special treatment (discussed in Appendix 1) but the evaluation of these methods, designed to decrease possibility of impurity introduction into the crystals, can only be done by both emission spectrographic analyses and electrical conductivity measurements on specimens from the pure crystals.

The emission analyses found that no detectable impurities are introduced at any stage of the pure crystals preparation and growth. In addition, electrical conductivity measurements gave a vacancy concentration equivalent to approximately less than 1 ppm divalent cation impurity. With these analyses, it is seen that any differences in track decoration in pure crystals and those crystals containing dopants of several ppm will be attributable to the differences in the impurities (dopants).

B. Doped Crystals

The doped crystals are analyzed primarily by electrical conductivity measurements at room temperature after the effective distribution coefficients have been determined for the impurities. Since there is careful control of the

average concentration of dopants in the crystals, the exact dopant content of any specimen can be calculated, as well as measured by the conductivity. The latter calculations are reliable for divalent impurity concentrations below the room temperature solubility limits.

V. Distribution of Dopants in Bridgman Grown Crystals

A crystal grown by the Bridgman method may have a variation of dopant concentration along its growth direction. This variation depends upon the dopant, growth rate, temperature gradient across the liquid-solid interface, and diffusion within the melt and solid. While complete knowledge of these parameters would be informative, they are not necessary to determine the effective distribution coefficients of different dopants in crystals grown under the same conditions. Accurate values of k enables selection of the region of a crystal containing the exact desired amount of dopant, prior to and without the need to cut or analyze the crystal.

In all crystals grown for this study, we assumed that the crystals were formed by normal freezing which requires that there is no diffusion of the dopant in the solid and that there is uniform dopant in the melt by constant mixing of the melt.⁷⁹ While these two conditions might not have existed in our growth method, the assumption that they are grown under normal freezing does enable us to obtain the effective distribution coefficient k . With this assumption, the distribution of a dopant in a crystal would be

$$C_g = k C_o (1 - g)^{k - 1}$$

where

- C_g = concentration of dopant at g in the crystal;
- g = fraction of total melt solidified at g in the crystal;
- k = effective distribution coefficient;
- C_o = average dopant concentration throughout the crystal
(amount of dopant added to the melt).

For a specimen of a few millimeters thick from a crystal 18 cm long, the g value is the average of the g value at the top of the specimen and the bottom of the specimen. This averaging of g will have a negligible effect on determination of k , and, once k has been determined, it will be a reliable value to use in selecting specimens with the desired amount of dopant.

VI. Use of Room Temperature Electrical Conductivity Measurements to

Determine Effective Distribution Coefficient, Dopant Solubility Limit, and Charge Carrier Mobility.

For a divalent cation in silver chloride, the ion's effective distribution coefficient k , room temperature solubility limit, and the mobility of the defect which it introduces can be determined from the room temperature electrical conductivity of specimens from the crystal containing the cation as an added dopant.⁸⁰ This method assumes normal freezing and that the dopant is a divalent cation whose addition to the crystal introduces one free vacancy which contributes to the electrical conductivity. For example, in silver chloride containing cadmium, the addition of divalent cadmium ions introduces an equal number of silver ion vacancies. At room temperature and at low concentrations of cadmium, virtually all of these vacancies are free to carry electric current. Thus, with 1 ppm of cadmium, the conductivity is approximately 5×10^{-9} mho/cm at room temperature, a value sufficiently large that the intrinsic conductivity may be neglected.

Taking the dopant concentration to be proportional to the measured conductivity, one has for n_g vacancy concentration

$$\sigma_g = n_g \mu_v e = \mu_v e C_o k (1 - g)^{k-1}$$

where μ_v is the vacancy mobility, e is the electron charge, g is the fraction of total melt solidified in the specimen with electrical conductivity σ_g , and k is the dopant's effective distribution coefficient. Since the initial dopant average concentration C_o is known and the value of g can be measured, a plot of $\log_e \sigma$ as a function of $\log_e (1 - g)$ can be used to determine the following:

1. slope gives k through the relationship

$$\text{slope} = k - 1;$$

2. intercept at $g = 0$ yields μ_v by the relationship

$$\mu_v = \frac{\sigma_g = 0}{e C_o k}$$

3. saturation of σ with g gives the equivalent solubility limit when it has been exceeded, via the value of 5×10^{-9} mho/cm being approximately 1 ppm of free vacancies at room temperature.

As an example of the application of the described relationships, the following parameters were obtained from specimens of a silver chloride crystal containing an average concentration of 200 ppm of cadmium:

1. effective distribution coefficient: $1.94 \pm .05$;
2. $\mu_v = 1.9 \times 10^{-6} \frac{\text{cm}^2}{\text{sec-volt}}$ at 24.5°C ;
3. solubility limit ~ 250 ppm at 24.5°C .

These values agree with those obtained by different and more elaborate techniques.

VII. Preparation of Specimens for Electrical Conductivity Measurements and Exposure to Ionizing Particles

A. Cutting Crystal

Each crystal was weighed and then cut with a specially designed stainless steel cutter to introduce as few strains and impurities as possible. Immediately after being cut, the specimen and the section from which it was cut were washed with isopropoal alcohol to remove any lubricant used during cutting. This procedure measures the exact mass removed during cutting so that the exact g values (top and bottom) can be calculated for each specimen. Since the crystal had an uncut mass greater than the upper limit of the Mettler, the first cut of the crystal divided it into two sections of approximately equal mass, with the mass removed during this first cut being correct for later when the mass per unit length for that region was determined experimentally from later specimens cut from that region. The crystal is cut into 15-20 specimens.

Since the electrical conductivity data is plotted as a function of $\log_e (1 - g)$, a small error in the cumulative mass for specimens near the top will have a large effect on the value of $\log_e (1 - g)$; therefore, it is essential to have accurate measurements of the uncut crystal, of the material removed during each cut, and of each specimen. Slight variation in the crystal cross section along its length required that the material removed by each cut be determined for each cut, with exception of the initial division of the crystal. A direct comparison of the uncut crystal with the mass obtained by summation of the specimens and the material removed from the cuts never differed by more than 0.2 gms. This difference of 0.2 gms is negligible when plotting the data for a 350 gms crystal because the $\log_e (1 - g)$ value would be $-4.035 \pm .015$ when $g = .982$, the approximate upper experimental

limit of g. (This limit was set because the top, uneven surface has to be removed to a dept of 2-3 mm.)

B. Electrical Conductivity Measurements

After being weighed, each specimen's thickness at its four corners and its center were measured. The average of these values were then used with the mass to obtain the ratio

$$\frac{t}{A} = \frac{\text{thickness}}{\text{Area}} = \frac{(\text{density}) t^2}{\text{mass}} = \frac{(5.56) t^2}{\text{mass}} \frac{\text{gm}}{\text{ml}}$$

to be used later to calculate the electrical conductivity σ from the conductance G.

Electrodes were applied by painting the two largest surfaces with either silver paint or Dag.

The conductance G was measured with a General Radio Type 1615A bridge. For pure specimens, the G values were measured from 200 Hz to 50 KHz whereas for doped specimens the frequency used was 10 KHz. The reason for using a single frequency for doped specimens will be explained in the next section.

The bridge was in the three terminal mode for G measurements, with the power being provided by a General Radio Type oscillator and bridge balance being monitored on a General Radio Type amplifier and null detector. The electrical conductivity σ was thereby calculated by

$$\sigma = G \frac{t}{a} = G \frac{(5.56) t^2}{\text{mass}}$$

where the units of G are nhos and σ is expressed in mho/cm. All G values were corrected for bridge error and were made at room temperature, approximately 24°C. A calibrated mercury thermometer was used to measure the specimen temperature during measurements and corrections were made to the σ values so that they could be expressed as being made at a particular temperature. This correction

is that σ changes $\pm 3.35\%$ per $\pm 1^\circ\text{C}$ and is based on the vacancy mobility change per degree celcius at room temperatures. To decrease the influence of handling on the specimen's temperature, measurements were made immediately after the specimen had been quickly placed into the conductivity apparatus and were repeated until the G value varied no more than 0.5% over a five minute period, the apparent time required for the specimens to reach thermal equilibrium after handling.

C. Preparation of Specimens for Exposure to Ionizing Particles

The specimens, after being cut from the crystals, were strained, so it was necessary to remove as much of this strain as possible prior to the exposure to ionizing particles. This strain, if permitted to remain in the specimens, would increase the electron trap density and thereby decrease the photoelectron mean free path during the decoration of the regions through which the ionizing particles passed.

The strains were considerably decreased by two operations. Firstly, the cut surfaces were removed to a depth of 2-3 mm by careful polishing on three different sizes of silicon carbide polishing papers and three different grit sizes on special polishing cloths. (At this stage of the preparation, no surface strains were observed in back reflection Laue x-ray photographs.) Secondly, the specimens were etched with a 3% (by weight) KCN solution in deionized distilled water. Following this etch, the specimens were washed in a 50% nitric acid-50% deionized distilled water solution and then washed with DDW.

The final procedure was an anneal in various atmospheres, with the anneal consisting of increasing the temperature at $\sim 9^\circ\text{C}/\text{hour}$, a soaking of ~ 12 hours at $\sim 425^\circ\text{C}$, and cooling to room temperature at a rate not greater

than ~5%/hour. The specimens were then exposed to ionizing particles at times and places described later in this report.

VIII. Experimental Effective Distribution Coefficient

The effective distribution coefficient k was measured for each of ten cations in silver chloride crystals grown in the chlorine-nitrogen mixture by the Bridgman method. Each crystal was cut into ten to twenty separate specimens whose σ and g values were used in the plots of $\log \sigma$ as a function of $\log_e (1 - g)$. Figure 2 contains data for four of the cations, along with the data for a pure crystal. These values are reproducible and have an accuracy of $\pm 5\%$.

Table 2
Effective Distribution Coefficients of
Ten Cations in Bridgman Silver Chloride Crystals

DOPANT	k
Cadmium	1.94
Calcium	1.39
Chromium	.58
Cobalt	.91
Iron	.82
Lead	.70
Manganese	1.50
Strontium	1.35
Tin	.70
Vanadium	.80

These distribution coefficients should be related to the energy of solid solution of the divalent cations, and details of this relationship will be reported in a later publication.

IX. Decoration of Tracks Produced by Ionizing Particles

Specimens were exposed to ionizing particles by four methods:

1. high altitude balloon flights; 2. machine accelerated non-relativistic carbon ions; 3. machine accelerated relativistic singly charged particles;
4. machine accelerated multiply charged particles.

A. High Altitude Balloon Flights

Specimens were sealed with Type G-5 nuclear emulsions in light-tight plastic holders, with the specimens positions being indexed on the emulsions. The flights were at ~120,000 feet for ~8 hours. After the packages were recovered, the emulsions were processed and the silver chloride specimens were decorated for two hours at room temperature.

The emulsions were examined for "high Z" tracks whose coordinates were recorded. Then, the decorated silver chloride specimens were scanned at a magnification of 200X; all observed track coordinates and directions were recorded.

All of the "heavy" emulsion tracks were observed in the specimens; however, these observations depended upon both the specimens impurity and its quantity. The best decoration of tracks was obtained in specimens which contained from 4 to 8 ppm of lead and which had been annealed in air. The inability to decorate tracks in specimens containing different dopants and having different treatment might have been caused by a high electron trap density within a few hundred micrometers of the surface where the photoelectrons were formed during decoration.

B. Machine Accelerated Non-relativistic Carbon Ions

Specimens were exposed to the highest energy carbon ions available at the Yale University accelerator. Those ions had an energy of 10 Mev/nucleon, giving a total energy of 120 Mev per carbon ion. Tracks of these particles were decorated in lead doped specimens, and typical decorated tracks are shown in Figure 3.

C. Machine Accelerated Relativistic Singly Charged Particles

Specimens were exposed to relativistic machine accelerated protons and mesons at the Brookhaven accelerator and at the Bevalac at the Lawrence Laboratory at the University of California at Berkeley. The purpose of these exposures was to determine if these minimum ionizing particles produced decoratable tracks and to observe any nuclear reactions these particles might produce within the silver chloride.

The results of these exposures were that the relativistic singly charged particles did not produce decoratable tracks. These particles, however, did cause some nuclear reactions in which some secondary particles loss sufficient energy to produce a decoratable track. As found in the previously described exposures, such tracks were observed only in lead doped silver chloride specimens annealed in air.

D. Machine Accelerated Relativistic Multiply Charged Particles

Specimens were exposed to 2.5 Gev nitrogen and 12 Gev carbon ions at the Bevalac at the Lawrence Radiation Laboratory at the University of California at Berkely. Tracks of these particles were decorated only in specimens of silver chloride containing 4-8 ppm of lead. These particles had sufficient energy to pass through the specimens with lengths of 2 cm. An example of a 12 BeV carbon track is shown in Figure 4.

X. Self-decorating Silver Halide Crystals

It was proposed in 1963 that it might be possible to produce silver halide crystals in which tracks could be decorated by merely exposing the crystal to actinic light in the visible region. That proposal was based on the observation that cuprous ions in silver chloride could be changed to the cupric ions by high intensity light at 430 mμ. Moser, Nail, and Urbach explained this change in terms of absorbed quanta producing holes and electrons, with the holes being trapped by cuprous ions to form some cupric ions and the electrons combining with silver ions to form silver atoms. Sliker showed that their observations of volume darkening and saturation were consistent with a mathematical analyses of their model in which there is a consumption of hole traps. Sliker also found that for less than 0.2 ppm Cu^+ , the recombination of holes and electrons become important. The self-decorating crystals, therefore, would require a sufficient quantity of cuprons ions which could provide free electrons, the same as during the synchronous pulses of UV-electric field, for forming silver atoms at the imperfections produced by the energy loss of the ionizing particles.

Crystals containing approximately 10 ppm of Cu^+ were exposed to continuous UV light passed through a 1 mm thick filter of pure silver chloride. This AgCl filter removed the light strongly absorbed and thereby enable the approximate uniform production of electron and holes throughout the copper doped specimens. The results were that decoration was obtained at dislocations but that decoration had two problems: 1. the decorated dislocations were "extremely fine" and thereby difficult to observe; 2. the decoration was unstable to microscope's strong illumination and disappeared within a few minutes after the illumination began.

The most successful self-decorating crystal yet devised was developed by the group of Schopper in cooperation with the group of Haase. Those crystals contained several thousand parts per million of cadmium. The variation of decoration with such variables as cadmium content, decoration duration, decoration temperature, track latent image stability, and decorated track stability have all been investigated. Equally important, from the standpoint of practical applications, Wendnagel has developed a process for growing these cadmium doped 0.6 mm thick silver chloride crystals with cross sections of 4 cm by 50 cm.

Two of the Schopper-Haase self-decorating cadmium doped AgCl specimens were made available by Professor Haase. The specimens were exposed to 1.05 GeV/nucleon carbon ions, and, 60 seconds later, they were exposed to actinic illumination. Tracks were observed decorated in one of the specimens but not in the other specimen. This lack of reproducibility to decorate tracks in "properly" prepared specimens is the major problem with the self-decorating crystals.

XI. Comparisons of Dosimetry by Swept-in Photoelectrons and by Actinic Illumination

There are differences in the basic characteristics of the decorated tracks by swept-in electrons and by exposure to actinic light (self-decorating crystals). These differences are tables below, and it is obvious that each method has its unique properties which might be best for certain types of experiments.

Comparison of Track Decoration in Silver Chloride Crystals

Property	Swept-in Electrons Method	Schopper-Haase U. V. Method
Dopant	Lead (8 ppm)	Cadmium (Thousands ppm)
Track Latent Image Stability (25°C)	greater than 6 months	less than seconds
Decoration Stability Under Microscope Illumination (25°C)	greater than ten hours, longest exposure given	a few minutes, depending upon intensity
Maximum specimen thickness	5000 microns; set by objective lenses	200 microns; set by growth method
Maximum Area per Specimen		200 cm ²
Long Term Decoration Stability at Room Temperature	Greater than 10 years	

There are also obvious differences in the information content of the tracks decoration, with the swept-in electrons producing a wider track with larger silver specks, as seen in Figure 4.

XII. Proposed Future Research on Silver Chloride for Dosimetry

The research performed under this contract showed that it is possible to prepared both silver precipitate and to use that precipitate in growing the highest purity silver chloride crystals yet obtained. In additional, similar results have been obtained with silver bromide.

Controlled doping of the silver chloride crystals have produced specimens in which swept-in photoelectrons results in docoration of the tracks of highly ionizing particles. Such tracks at room temperature have a latent image stability greater than six months and have a decorated lifetime greater than ten years. Although these are reproducible, the self-decorating crystals would probably be the most fruitful from the standpoint of producing a simple and easily utilized dosimeter. This prediction is based on two observations: 1. the crystals can be grown in large thin sheets up to approximately 4 cm by 50 cm;

2. decoration is performed by merely exposing the crystals to the proper illumination. In contrast, the decoration produced by sweeping in photoelectrons requires specimens which have to be prepared after growth of the crystal, is sensitive to surface deformation, and must be done with special illumination in the form of a few microseconds flash synchronized with an electric field.

As the first step in investigating the self-decorating specimens, the growth methods of Wendnagel should be produced and further studied as to the effective distribution coefficients of various dopants. Then, there should be growth of specimens containing more than one dopant because it might be possible to grow specimens in which there may be long term track latent image stability as well as long term decoration stability. By doing such research on both silver chloride and silver bromide, the stopping power could be either 5.56gm/cm or 6/47 gm/cm. Also, since we have grown mixed AgBr:AgCl crystals, it would be possible to grow a crystal in which the stopping power is anywhere between that of the two silver halides. The starting point of any such study should be with crystals containing lead and crystals containing cadmium, the latter having been developed by the Schopper group at Frankfurt and the Haase group at Munich. These two groups have reported some difficulty with their self-decorating cadmium doped AgCl so it would be helpful if the source of the problem could be identified and eliminated.

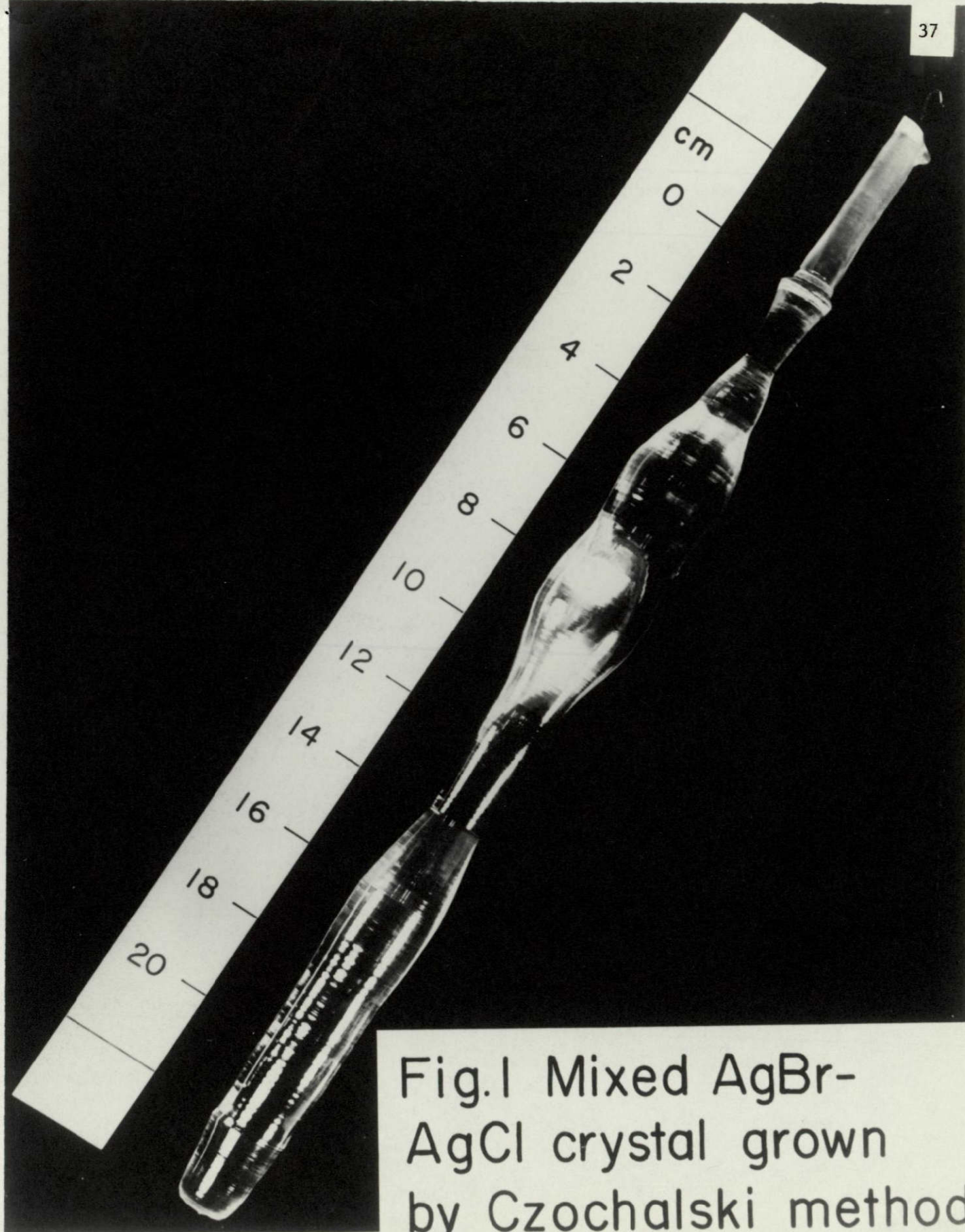


Fig.1 Mixed AgBr-AgCl crystal grown by Czochalski method.

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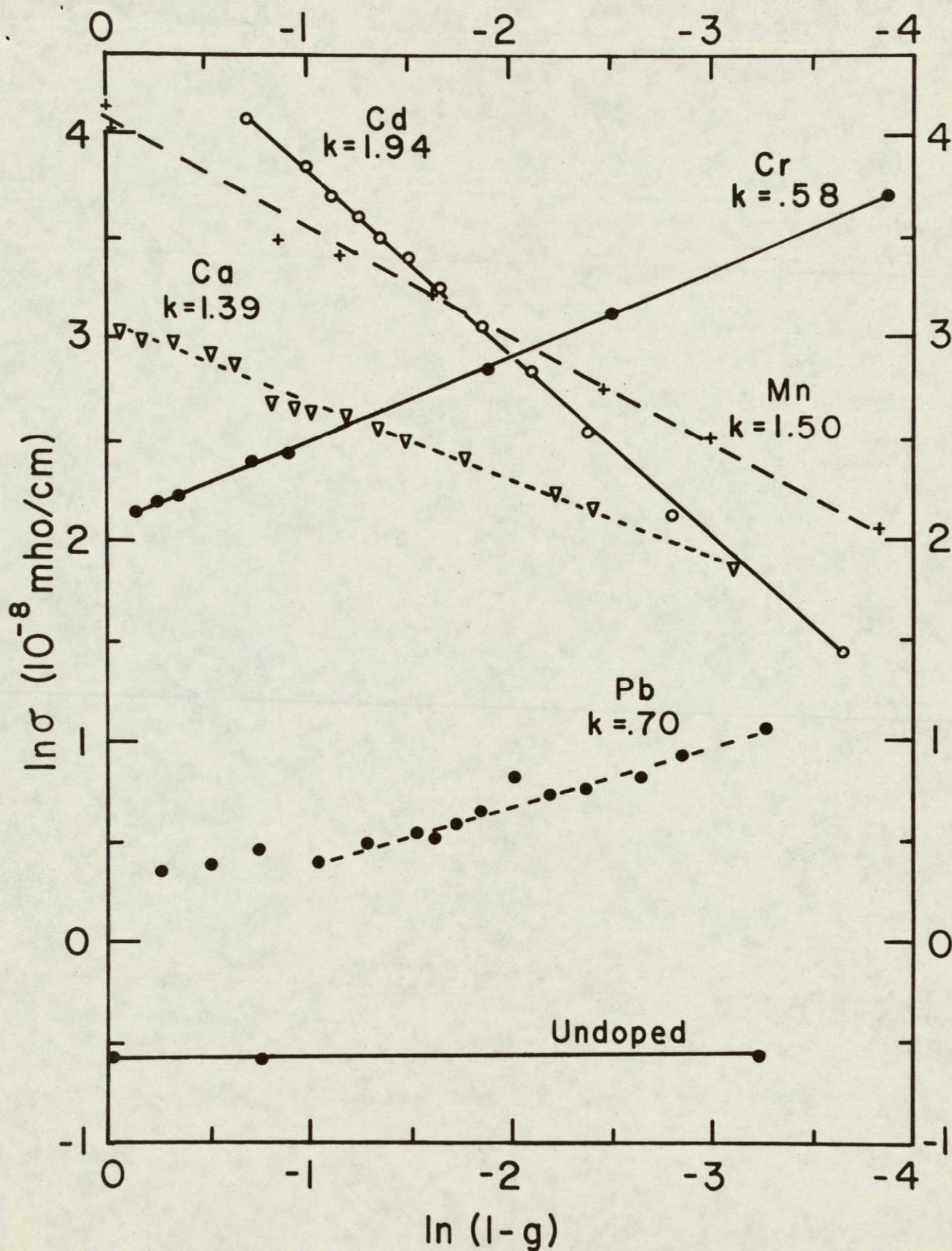
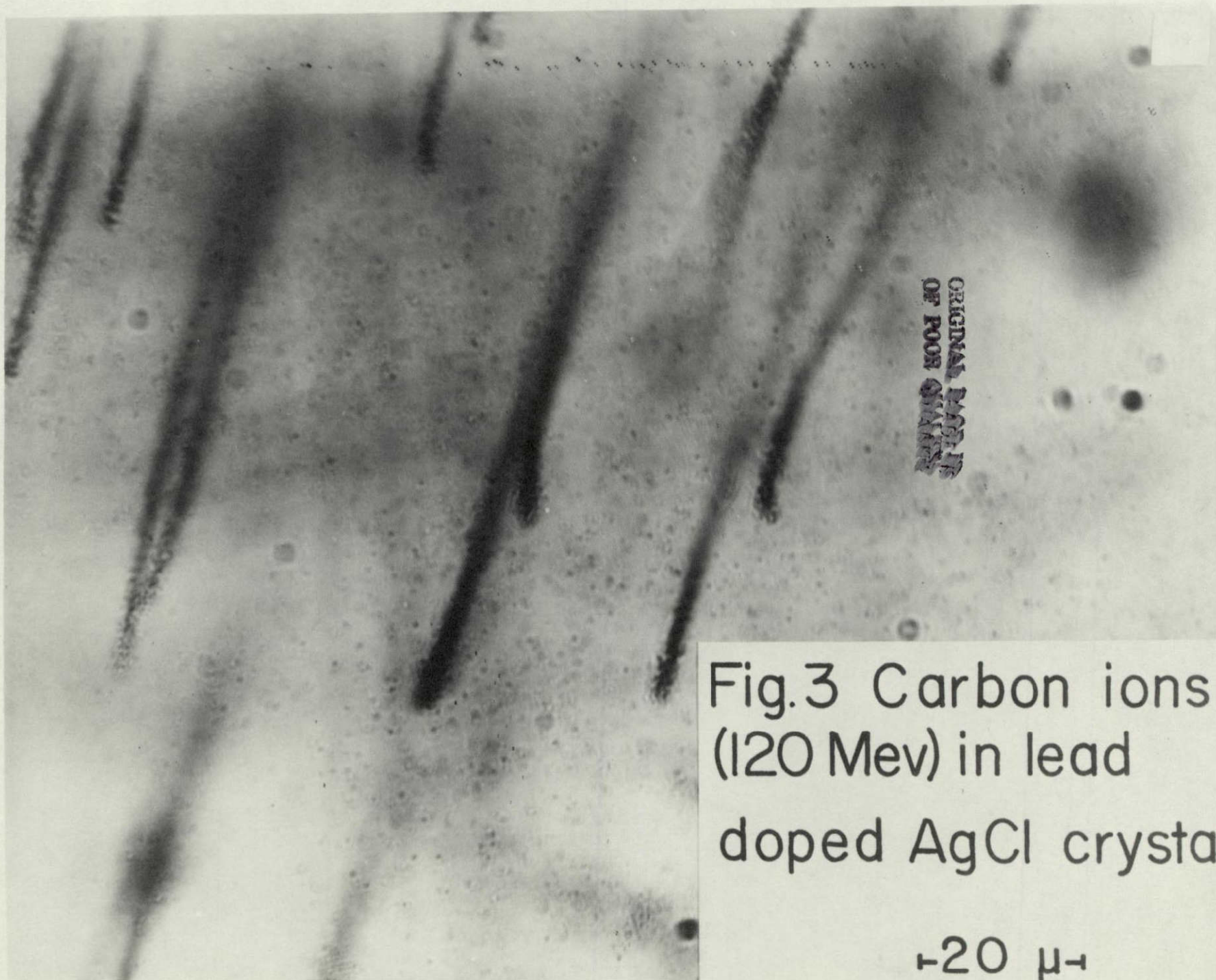


FIG 2. Normal freezing graphs of $\ln \sigma$ vs $\ln (1-g)$ for five doped and one undoped AgCl crystals (k = eff. distribution coefficient).



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Fig.3 Carbon ions
(120 Mev) in lead
doped AgCl crystal.

$\sim 20 \mu$

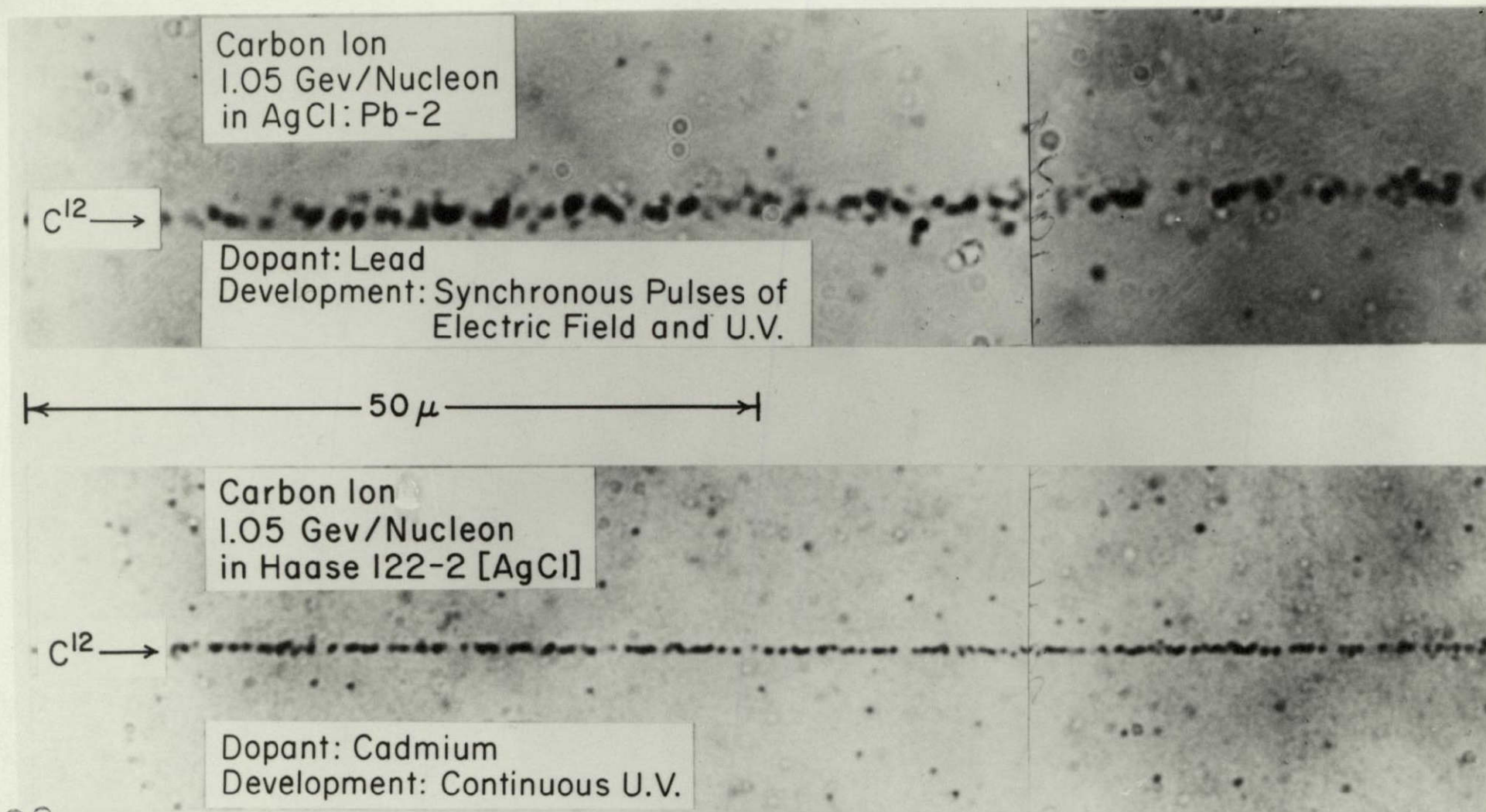


Fig.4 Carbon ions in AgCl crystals.

REFERENCES

1. R. W. Walker, "Characteristics and Applications of Solid State Track Detectors," Reunion de Travail sur l'enregistrement des traces de particules chargees dan les cristaux, CNRS Strasbourg (1963).
2. P. B. Hirsch, Proceedings of the International School of Physics, 18th Course, Academic Press, p. 39 (1962).
3. R. L. Fleischer, P. B. Price, R. W. Walker, and E. L. Hubbard, Phys. Rev. 133, 1443 (1964).
4. M. M. Shapiro, Rev. Mod. Phys. 13, 58 (1941); Handbuch der Physik 45, 342 (1958).
5. H. Yagoda, Radioactive Measurements with Nuclear Emulsions, John Wiley and Sons, Inc., New York (1949).
6. Y. Goldschmidt-Clermont, Ann. Rev. Nucl. Sci. 3, 141 (1953).
7. P. Demers, Ionographie. Les Presses Universitaires de Montreal (1955).
8. M. Blau, Methods of Exp. Phys. 5-A, 208 (1961).
9. C. F. Powell, P. H. Fowler, and D. H. Perkins. The Study of Elementary Particles by the Photographic Method, Pergamon Press, New York (1959).
10. W. H. Barkas, Nuclear Research Emulsions; Techniques and Theory; Vol. 1, Academic Press, New York (1963).
11. W. H. Barkas, ibid., p. 14.
12. J. W. Mitchell, Reports on Prog. Phys. 20, 433 (1957).
13. W. H. Barkas, op. cit., pp. 153-179.
14. W. H. Barkas, op. cit., p. 172.
15. C. F. Powell, P. H. Fowler, and D. H. Perkins, op. cit., p. 42.
16. W. H. Barkas, op. cit., pp. 78, 123-124, 186-188, 191.
17. P. J. Duke, W. O. Lock, P. V. March, and B. A. Munir, J. Sci. Instr. 32, 365 (1955).
18. W. M. Gibson and J. G. McEwen, Nuovo Cimento 7, 67 (1958).
19. J. K. Boggild, K. H. Hansesn, J. E. Hooper, and M. Scharff, Suppl. Nuovo Cimento 26, 305 (1962).
20. E. Dahl-Jensen. 1962 Easter School, CERN, Geneva (1963), p. 129.
21. W. H. Barkas, op. cit., pp. 387-388; Phys. Rev. 124, 897 (1961).
22. K. Kristiannson, O. Mathiesen, and B. Waldeskog, Arkiv Fysik 17, 485 (1960).

23. G. Alvial, L. Grimaldi, J. Riquelme, E. Silva, and S. Stantic, *Nuovo Cimento* 15, 25 (1960).
24. B. J. O'Brien and J. H. Noon, *Nuovo Cimento* 8, 807 (1958).
25. E. J. Williams, *Proc. Roy. Soc. A* 169, 531 (1939); *Phys. Rev.* 58, 292 (1940).
26. S. A. Goudsmit and J. L. Saunderson, *Phys. Rev.* 57, 24 (1940); *Phys. Rev.* 58, 36 (1940).
27. G. Molliere, *Z. Naturforschg* 2a, 133 (1947); *Mehrfach-und Vielfachstreuung* 3a, 78 (1948).
28. C. Fichtel and M. W. Friedlander, *Nuovo Cimento* 10, 1032 (1958).
29. S. Biswas, B. Peters, and B. Rama, *Suppl. Proc. Indian Academy of Sciences* A41, 154 (1955).
30. F. A. Brisbout, C. Dahanayaki, A. Engler, P. H. Fowler, and P. B. Jones, *Nuovo Cimento* 3, 1400 (1956).
31. A. J. Apostolaki, J. O. Clarke, and J. V. Major, *Nuovo Cimento* 5, 337 (1957).
32. A. Engler, M. F. Kaplon, A. Kernan, J. Klarmann, C. E. Fichtel, and M. W. Friedlander, *Nuovo Cimento* 19, 1090 (1961).
33. E. Dahl-Jensen, M. Gailloud, W. O. Lock, and G. Vanderhaeghe, *Nuovo Cimento* 22, 850 (1961).
34. P. J. Aditya, V. S. Bhatia, and P. M. Sood, *Nuovo Cimento* 29, 577 (1963).
35. F. Seitz, *Disc. Faraday Soc.* 5, 271 (1949).
36. J. S. Koehler and F. Seitz, *Zeits. f. Physik* 138, 238 (1954).
37. G. J. Dienes, *Ann. Rev. Nucl. Sci.* 2, 187 (1953).
38. J. S. Koehler and F. Seitz, Defects in Crystalline Solids; Report of 1954 Bristol Conference, pp. 222-231.
39. E. A. Uehling, *Ann. Rev. Nucl. Sci.* 4, 315 (1954).
40. B. T. Price, *Reports on Prog. in Physics* 18, 52 (1955).
41. G. H. Kinchin and R. S. Pease, *Reports on Prog. in Physics* 18, 1 (1955).
42. H. Brooks, *Ann. Rev. Nucl. Sci.* 6, 215 (1956).
43. F. Seitz and J. S. Koehler, Solid State Physics 2, 377 (1956).
44. G. J. Dienes and G. H. Vineyard, Radiation Effects in Solids, Interscience Publishers, Inc., New York (1957).
45. R. M. Sternheimer, *Methods Exp. Phys.* 5-A, 1 (1961).

46. A. N. Goland, *Ann. Rev. Sci.* 12, 243 (1962).
47. D. S. Billington and J. H. Crawford, Radiation Damage in Solids, Princeton University Press (1961).
48. Proceedings of the International School of Physics; Enrico Fermi; 18th Course, Editor: D. S. Billington; Academic Press (1962).
49. J. A. Brinkman, *J. Appl. Phys.* 25, 961 (1954); *Am. J. Phys.* 24, 246 (1956).
50. F. Seitz, *Rev. Modern Phys.* 34, 656 (1962).
51. C. J. Beevers and R. S. Nelson, *Phil. Mag.* 8, 1189 (1963).
52. G. R. Piercy, *J. Phys. Soc. Japan* 18, Suppl. III, 169 (1963).
53. L. T. Chadderton, *Phil. Mag.* 8, 2085 (1963).
54. S. Amelinckx, Proc. International School of Physics; Enrico Fermi; 18th Course, pp. 422-517.
55. S. Amelinckx, W. Van der Vorst, R. Gevers, and W. Dekeyser, *Phil. Mag.* 46, 450 (1955).
56. W. Van der Vorst and W. Dekeyser, *Phil. Mag.* 1, 882 (1956).
57. D. J. Barber, K. B. Harvey, and J. W. Mitchell, *Phil. Mag.* 2, 704 (1957).
58. J. T. Bartlett and J. W. Mitchell, *Phil. Mag.* 3, 334 (1958).
- 59-a. J. M. Hedges and J. W. Mitchell, *Phil. Mag.* 44, 223 (1953).
- 59-b. J. M. Hedges and J. W. Mitchell, *Phil. Mag.* 44, 357 (1953).
60. D. A. Jones and J. W. Mitchell, *Phil. Mag.* 2, 1047 (1957).
61. D. A. Jones and J. W. Mitchell, *Phil. Mag.* 3, 1 (1958).
62. A. S. Parasnis and J. W. Mitchell, *Phil. Mag.* 4, 171 (1959).
63. C. B. Childs and L. Slifkin, *Phys. Rev. Letters* 5, 502 (1960).
64. J. R. Haynes and W. Shockley, Report of a Conference on Strength of Solids, Physical Society London (1948), p. 151.
65. F. C. Brown, *Phys. Rev.* 97, 335 (1955).
66. J. H. Webb, *J. Appl. Phys.* 26, 1309 (1955).
67. J. F. Hamilton, F. A. Hamm, and L. E. Brady, *J. Appl. Phys.* 27, 874 (1956).
68. P. Süptitz, *Z. f. Physik* 153, 174 (1958).
69. C. Childs, M. S. Thesis, University of North Carolina (1959).

70. C. Childs, "Impurity Effects on Decoration and Speculation on Self-Decorating Silver Chloride Crystals," Reunion de Travail sur l'enregistrement des traces de particules chargees dans les cristaux, CNRS Strasbourg (1963).
71. P. Poubeau and R. Schmitt, "Quelques Etudes et Realisation en cours a Strasbourg dans le cadre de la detection ionographique," Reunion de Travail sur l'enregistrement des traces de particules chargees dan les cristaux, CNRS Strasbourg (1963).
72. F. Seitz, Rev. Mod. Phys. 23, 328 (1951).
73. C. Childs and L. Slifkin, Bull. Am. Phys. Soc. 6, 52 (1961).
74. C. Childs and L. Slifkin, I.R.E. Trans. Nucl. Sci. NS-9, 413 (1962).
75. C. Childs and L. Slifkin, Rev. Sci. Instr. 34, 101 (1963).
76. C. Childs and L. Slifkin, Phys. Rev. Letters 9, 354 (1962).
77. R. Schmitt, "Detection des Particules Ionisantes dans les Monocristaux de Chlorure d'Argent," Reunion de travail sur l'enregistrement des traces de particules chargees dan les cristaux, CNRS Strasbourg (1963).
78. F. Moser, N. R. Nail, and F. Urbach, Phys. Chem. Solids 9, 217 (1959).
79. William G. Pfann, Zone Melting, John Wiley and Sons, Inc., New York, 1958, Chapter 2.
80. C. Childs, ACCG Conference on Crystal Growth, National Bureau of Standards, 1969, page 19.

Appendix 1

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LETTERS TO THE EDITOR

HIGH PURITY SILVER BROMIDE CRYSTALS CONTAINING LESS THAN SEVERAL PARTS PER BILLION OF IODIDE

Charles B. CHILDS

Materials Research Center, University of North Carolina, Chapel Hill, North Carolina 27514, USA

Received 16 February 1977

High purity, nearly iodide-free silver bromide crystals of 360 g are routinely grown from specially prepared silver bromide precipitate. The iodide content is decreased from 13–30 ppb in the precipitate to 3–6 ppb in the crystals by proper bromine treatment of the melts. Although zone refining is not used at any stage in preparing these crystals, their ionic conductivity at 25°C is comparable to that of zone refined silver bromide. A proposal is made for growing silver bromide crystals which should have iodide contents less than 3 ppb and ionic conductivity at low temperatures nearing that expected for pure silver bromide.

High purity silver bromide crystals are required for studies of intrinsic excitons and as a standard for the identification of various photoexcited centers in both the pure crystals and those doped in known ways. To be certain that the optical features being studied are those of "pure" silver bromide and not those of impurities, many different methods have been used to decrease the impurity content of silver bromide. Among those methods have been fractional distillation [1], Bridgman growth [2], zone refining [3], and dropping growth [4]. While all of these methods decrease the cation impurity content of the "pure" silver bromide crystals, none of them has produced silver bromide crystals with less than 200 ppb (ppb = 10^{-9} mole fraction) of iodide [5–8]. Although one might expect that an iodide content as small as this would be tolerable, in fact, the iodide acts as a very potent trap for photoproduced holes. Thus, even at this low level, iodide is found to produce considerable luminescence at low temperatures [6]. It is therefore difficult to prepare crystals whose behavior is that of the intrinsic AgBr, in large part because the distribution coefficient of iodide is near unity in silver bromide [9].

The present paper reports the routine growth of large silver bromide crystals (~360 g) containing less than a few ppb iodide and having, at the same time, room temperature ionic conductivity (a measure of

divalent cation content) comparable to that of zone refined silver bromide. The growth of these high purity crystals does not employ either zone refining or fractional distillation of the silver bromide but it does include careful choice of starting materials and a subsequent bromine treatment of the molten AgBr, as well as treatment of the apparatus.

(1) Preparation of silver bromide precipitate. The silver bromide precipitate is made with only slight modifications of the method devised by Nail, Moser, Goddard, and Urbach [2] who employed high purity silver nitrate*, distilled hydrobromic acid, and distilled water. Our modifications include changes in apparatus, use of commercially available high purity hydrobromic acid**, and use of filtered† deionized distilled water with a resistivity of 17–18 megohm-cm at 25°C. Each AgBr lot has a theoretical yield of 8 moles (1.5 kg) but the losses in the various washings of the precipitate cause the useful yield to be about 7.6 moles (1.43 kg).

(2) Cleaning of apparatus. Before the glass and quartz components of the apparatus are used, they

* Silver nitrate, Special Product X491, Eastman Kodak Co., Rochester, NY, USA.

** "Suprapur" HBr acid, Product No. 306, EM Laboratories, Inc., Elmsford, NY, USA.

† Pore sizes of filters 0.45 and 0.22 μm .

are cleaned with a cleaning solution (5% HF acid, 35% nitric acid, 60% deionized distilled water), thoroughly rinsed with deionized distilled water, and dried at 40°C.

(3) Preparation of the bromine. Analytical reagent bromine is fractionally distilled three times in a closed glass manifold. The central two-thirds of each distillate is used for the next distillation in order to discard possible fractions containing iodine and chlorine. The final distillate is collected in a vacuum-tight stop-cock fitted container, which is then attached to the melt-treating manifold. (Dow Corning High Vacuum Silicone Grease is used on all ground joints.)

(4) Bromine treatment of crucible and envelope. Each crystal is grown by the Bridgman method. The crucible is made of high purity quartz (2 cm × 2 cm × 23 cm; wall thickness 1.5 mm) ^{††}. This crucible is placed in a standard taper-fitted Pyrex envelope which is then inserted into a furnace at ~475°C. The envelope is evacuated to 2×10^{-6} Torr, filled to ~1 bar with a gas mixture composed of 25% of the distilled bromine and 75% of dried, high purity nitrogen. After 20 minutes, the gas mixture is evacuated from the envelope which is then refilled with a fresh mixture. This process is repeated twice, with the third and final gas filling being at a pressure slightly higher than atmospheric. This gas treatment decreases the possibility of later crystal contamination by surface impurities on both the crucible and the envelope.

(5) Placement of silver bromide charge into crucible. The silver bromide precipitate is melted in a quartz tube fitted with a 1.5 mm capillary [10]. A hand torch is used to melt the precipitate, which then flows through the capillary directly into the crucible, with some heavy metal impurities being removed as a slag by their adhering to the quartz capillary.

(6) Melt treatment in crucible. A 4 mm quartz tube is inserted through the melt and extends to within 5 mm of the crucible's tip. The same Br₂-N₂ mixture is flowed through this tube and the melt at a rate of ~35 ml/min at a pressure of ~1.3 bar. After 1 h of treatment, the gas flow is ended, the tube is removed from the melt, and the envelope is sealed with a hand torch.

(7) Crystal growth. The sealed envelope is placed into a Bridgman furnace [2] whose upper section is at a temperature near to 490°C and whose lower section is ~300°C. The crystal growth is started several hours later by lowering the envelope at a rate of 1.0 mm/h. When the complete envelope is in the lower section of the furnace, the envelope (with the crystal) is quickly removed from the growth furnace and placed into an annealing furnace at 300°C. The envelope is cooled to room temperature by decreasing the temperature at a rate not exceeding 8°C/h. This slow cooling is designed to minimize the production of thermal strains in the crystal. In our experience, there has been no sticking of the crystal to the crucible under these growth conditions.

(8) Impurity content. The impurity content was determined for several crystals grown from three different preparations of silver bromide precipitate, but using portions of the same batches of silver nitrate, hydrobromic acid, deionized distilled water, distilled bromine, and high purity nitrogen. The three melts were treated simultaneously with the Br₂-N₂ mixture. Specimens for analysis were cut from the center of mass of each crystal. Emission spectrographic analyses of both precipitates and crystals did not detect the presence of any of the 54 cations sought, with detection limits of ~1 ppm (ppm = 10^{-6} mole fraction). The iodide content was estimated from the ratio of the intensity of the low temperature 2.48 eV iodide luminescent band to that of the 2.14 eV emission band of other centers [6]. These results, along with the room-temperature conductivity measurements, are given in table 1. It should be pointed out that the extrinsic ionic conductivity is determined

Table 1
Iodide content of precipitates and crystals and ionic conductivity (25°C) of crystals

Crystal	Iodide (ppb)		Ionic conductivity (10^{-8} mho/cm at 25°C, 10 kHz)
	Starting precipitate	Crystal	
AgBr: 11	30 ± 1	5.5 ± 1	2.2
AgBr: 12	20 ± 1	2.7 ± 1	2.5
AgBr: 13	13 ± 1	3.4 ± 1	2.6

^{††} All quartz is Type 204, General Electric Co., Willoughby, Ohio, USA.

primarily by the free vacancies introduced by polyvalent cation impurities, and the values in the table should be compared with that expected for ideally pure AgBr, whose intrinsic conductivity is $\sim 10^{-8}$ mho/cm (obtained by extrapolating to 25°C the data of Moser, Burnham, and Tippin [3] for zone refined AgBr which was intrinsic almost to room temperature). The ionic conductivities of these three crystals indicate a concentration of cation vacancies of the order of 100 ppb.

(9) Discussion of impurity content. The low iodide content (10–30 ppb) of the precipitate compared with the apparent norm of 200 ppb previously encountered is attributed to the higher purity of the particular silver nitrate used, the high purity hydrobromic acid, and the deionized distilled water with a low conductance, as well as special precautions to decrease impurity introduction from the apparatus. The improvement from 10–30 ppb in the precipitates to 3–6 ppb in the crystals is attributed to the treatment of the melt with bromine vapor which itself had been purified by distillation. Clearly, an equilibrium should ultimately develop between the iodide in the melt and the low iodine concentration in the bromine vapor being bubbled through the melt. The residual variation of iodide in the three crystals may simply mean that equilibrium was not always completely established.

The ionic conductivities of these three specimens are comparable with those ($1.7\text{--}3.0 \times 10^{-8}$ mho/cm) routinely measured on many other silver bromide crystals prepared in this laboratory by the same method. They are also comparable with the reported conductivities of zone refined silver bromide [3].

Since ionic conductivity may be used to measure the polyvalent cation impurity content and since at present one has not yet reached the expected intrinsic level at room temperature, the interesting possibility arises of obtaining still higher purities by first zone refining the precipitated material. Of course, such an additional procedure would demand additional precautions against the reintroduction of iodide and the introduction of additional impurities. It is thus

suggested that unique silver bromide crystals might be grown by an extended bromine treatment of the central regions of zone refined ingots of the described precipitate, with low ionic conductivity being used in selecting the regions to be used for the melt. Such crystals should have iodide contents near or below the detection limit of ~ 1 ppb for the most sensitive test of I^- in silver bromide and may have, as well, low-temperature ionic conductivities, approaching those expected for ideally pure silver bromide.

The author expresses his sincere appreciation to Dr. R.S. Eachus of the Research Laboratories of the Eastman Kodak Company for his interest in measuring the iodide contents of the precipitates and the crystals and for granting permission to include his data in this report. This work was supported in part by the Materials Research Center, University of North Carolina under Grant No. DMR-7203024 from the National Science Foundation and Grant No. NAS8-26601 from the George C. Marshall Space Flight Center of the National Aeronautics and Space Administration.

References

- [1] D.C. Burnham, Thesis, University of Illinois, Urbana, Illinois (1959).
- [2] N.R. Nail, F. Moser, P.E. Goddard and F. Urbach, *Rev. Sci. Instr.* 28 (1957) 275.
- [3] F. Moser, D.C. Burnham and H.H. Tippin, *J. Appl. Phys.* 32 (1961) 48.
- [4] P. Junod, B. Kilchör and H. Walliser, *J. Crystal Growth* 10 (1971) 144.
- [5] H. Kanzaki and S. Sakuragi, *J. Phys. Soc. Japan* 27 (1969) 109.
- [6] F. Moser and S. Lyu, *J. Luminescence* 3 (1971) 447.
- [7] H. Kazaki and S. Sakuragi, *Phot. Sci. Eng.* 17 (1973) 69.
- [8] von H. Hediger, J. Wulschleger and P. Junod, *Helv. Phys. Acta* 47 (1974) 442.
- [9] R.S. Eachus, private communication.
- [10] P.V. McD. Clark and J.W. Mitchell, *J. Phot. Sci.* 4 (1956) 1.

APPENDIX 2

THE UNIVERSITY OF NORTH CAROLINA

General Administration

P. O. BOX 2688

CHAPEL HILL 27514

WILLIAM FRIDAY
President

August 6, 1975

TELEPHONE: (919) 933-6981

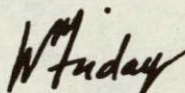
Mr. William Kinard
Long Duration Exposure
NASA
Langley Research Center
Hampton, Virginia 23665

Dear Mr. Kinard:

I am enclosing a proposal for which we are
requesting your support.

We hope that this proposal will merit your
kind consideration.

Cordially,



William Friday

THE UNIVERSITY OF NORTH CAROLINA
AT
CHAPEL HILL
27514

DEPARTMENT OF PHYSICS
AND ASTRONOMY

23 July 1975

Mr. William Kinard,
Long Duration Exposure Facility Project Manager
National Aeronautics and Space Administration
Langley Research Center
Hampton, Virginia 23665

Dear Mr. Kinard:

In response to the invitation contained in your letter of 20 June 1975, we are herein submitting a proposal for an experiment to be included on the Long Duration Exposure Facility. This proposed experiment is entitled "Dosimetry of High 'Z' Particles Using Silver Halide Single Crystals."

In formulating the Cost Plan in Section II-B of the proposal, we have been very conscious of the restricted funds available for the LDEF experiments so we have included the labor cost for the principal investigator at 1/4 full time. This is a five-year program, and there are several ways in which it could benefit from a full-time research associate; that would, of course, require additional funding, amounting to approximately \$85,000. As you know, of that amount, approximately \$35,000 would go to the university as indirect cost, leaving \$50,000 or \$10,000 per year for a research associate. Please let us know if it should develop that the LDEF Project considers that the proposal merits such additional funding.

We would be glad to supply you with any additional information about this proposal which you might desire.

Sincerely yours,

Charles B. Childs

Charles B. Childs

ORIGINAL PAGE IS
OF POOR QUALITY

"DOSIMETRY OF HIGH 'Z' PARTICLES USING SILVER HALIDE
SINGLE CRYSTALS"

An experiment proposed for inclusion
on the
Long Duration Exposure Facility.

July, 1975

Charles B. Childs
Charles B. Childs,
Principal Investigator
Materials Research Center
University of North Carolina
Chapel Hill, N. C. 27514
Telephone: (919) 933-5002

Lawrence M. Slifkin
Lawrence M. Slifkin,
Co-investigator
Department of Physics
University of North Carolina
Chapel Hill, N. C. 27514
Telephone: (919) 933-1185

E. N. Mitchell
E. N. Mitchell, Assistant Chairman
Department of Physics and Astronomy
University of North Carolina
Chapel Hill, N. C. 27514
Telephone: (919) 933-3013

George R. Holcomb
George R. Holcomb, Dean
of Research Administration
University of North Carolina
Chapel Hill, N. C. 27514
Telephone: (919) 933-1383

L. Felix Joyner
L. Felix Joyner, Vice President
of Finance
General Administration
University of North Carolina
910 Raleigh Road
Chapel Hill, N. C. 27514
Telephone: (919) 933-6981

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I. INVESTIGATION AND TECHNICAL PLAN
A. Investigation and Technical Plan

1. Summary

This investigation will be conducted by preparing specimens of specially doped silver halide single crystals which will be exposed to cosmic radiation during the flight of LDEF. The specimens will then be decorated by the Haynes-Shockley technique and evaluated as heavy ion dosimeters for future long-duration space flights. (Details of the processes and techniques are given in Section 1-c.)

2. Objectives and Significant Aspects of the Experiment

The objectives of this experiment are to develop, test, and evaluate silver halide crystals as heavy ion dosimeters for future long-duration space flights. These objectives are a continuation of the research by the present authors who first developed the use of silver halide crystals as dosimeters for high "Z" particles. These silver halide dosimeters are already known to have several advantages over dosimeters now existing. Among those advantages are:

1. sensitivity is not affected by vacuum, humidity, gravitational fields, or magnetic fields;
2. complete passivity, with no power requirements during space flights;
3. insensitivity to singly charged, relativistic particles, thereby enabling the study of the heavy chemical component of cosmic radiation in the presence of high integrated flux of single charged particles such as will occur during long-duration space flights;
4. non-chemical development which provides the potential of a simple, on-board method of acquiring data on the heavy ion component during long-duration space flights;

5. high stopping power, up to 6.5 gms/cm;
6. spatial resolution of less than one micron because development produces no distortion in the latent images of the tracks;
7. latent image stability of at least six months at 25°C.

While the crystals have performed well as heavy ion dosimeters for machine-accelerated heavy ions and for short-term space flights, the crystals have not yet been exposed to the actual environment of long-duration space flights; therefore, there is a need to have crystals on long-duration space flights for the dual purpose of testing their use on such flights and for obtaining data on the high Z component of cosmic radiation.

LDEF will make possible the complete test and evaluation of silver halide crystals as dosimeters for future long-duration space flights by exposing the crystals to the actual physical conditions of extended space flights and to the effects of both the low Z and high Z components of the cosmic radiation, the combination of which cannot be simulated on earth.

Up to the present time, silver halide crystals have been used as heavy ion dosimeters, but those crystals were not subjected to either the mechanical or thermal conditions offered by LDEF. Such conditions are necessary to make a valid evaluation of the crystals as heavy ion dosimeters during future extended space flights.

3. Investigative Approach

a. LDEF Experiment

The experiment will be performed in three stages. The first stage will include the production of high purity silver halides and their use in growing specially doped crystals of the composition which have been previously used for heavy ion dosimetry. The crystals will be prepared into samples which will

then be assembled into prototype flight units. These units will be designed, fabricated, and given the environmental tests required for L. D. E. F., followed by exposure to heavy ion beams at the Lawrence Laboratory at Berkeley. The units will then be given additional environmental tests and the crystals photoelectronically developed to compare their features with those of controls.

The second stage of the program will be the production of the actual flight and back-up units.

The third stage will include development of the crystals after flight and acquisition of data, culminating in an analysis and report of all findings.

The following section gives a short summary of the processes which enable the use of silver halide crystals as heavy ion dosimeters.

b. Radiation Effects in Solids

When a material is penetrated by a heavy charged particle, the particle loses most of its energy by two processes. First, it may lose energy by coulombic interaction (elastic collision) of its nucleus with that of nuclei in the material. In such elastic collisions, the electrons play no role other than partial screening of the nuclear charge. Second, the particle may lose energy by direct interaction with electrons, which results in the host electrons being elevated to excited states (inelastic collision). Of these mechanisms, the dominant one is energy loss by inelastic collision, with the rate of loss being a function of the particle charge, velocity, mass, and the density of the penetrated material.

The electronic excitation, resulting from direct interaction between the host atomic electrons with either the incident particle or from primary recoil atoms, may produce what Seitz and Koehler have termed "thermal spikes."¹ These spikes are regions in which the localized energy loss is mainly converted into heat.

In these thermal-spike regions, the material is heated to several hundred degrees and is then thermally quenched. This heating process takes place in less than 10^{-10} second and forms "large" concentrations of point defects which may form stable clusters during the subsequent rapid cooling. In addition, these defects and the increase in temperature can produce a disordering which causes a volume change ΔV . The volume change depends upon the number of defects, their volume, and atomic rearrangement resulting from thermal quenching. This ΔV plus the intense temperature gradients produce a stress field which results in plastic flow near the spike and thereby forms permanent linear imperfections (dislocations) at distances much greater than the radius of the transiently molten core of the spike.

Another type of spike concept has been formulated by Brinkman.² He proposed that since the time of the molten state is greater than the relaxation time, there is sufficient strain energy, released after density fluctuations have relaxed, to raise the temperature even higher and thereby extend the period of existence of the liquid state. This resulting temperature extension produces turbulent motion so that most of the atoms will occupy new lattice sites. Such a region which has undergone melting and resolidification is a "displacement spike."

Regardless of the validity of these models of the radiation damage process, one expects the track of a heavy charged particle to be surrounded by a cylindrical region of intense thermal strain. In a soft material such as silver chloride, such a region would be highly dislocated, and hence contain a high density of electron traps. It is this circumstance which provides the basis for track decoration by the method to be employed in the proposed experiments.

c. Decoration of Imperfections

The essential act in using crystals for radiation detection is making microscopically visible the radiation-induced imperfections. The method used to achieve this is called "decoration."

Decoration is a term which refers to making crystal imperfections microscopically visible by deposition of material at the imperfections. The decoration can then be observed when the deposited material is sufficiently opaque to illumination transmitted by the crystal. For practical studies of radiation-induced imperfections, the decoration should be observable with a conventional microscope using visible-light illumination.

There have been three techniques reported for decoration of silver halides (AgBr and AgCl). One of these techniques was developed by Bartlett and Mitchell³ who exposed doped crystals to ultraviolet light and then annealed them to obtain decoration. Since here, as with the alkali halide decoration, there is an anneal which would affect radiation-induced imperfections, we will consider this technique as improper for radiation detection. The two remaining methods produce imperfection decoration at room temperature, thereby being of primary interest for radiation studies by decoration.

A second procedure for decoration of a silver halide was formulated by Hedges and Mitchell.⁴ Their initial decoration was obtained by merely exposing AgBr to light in its long-wave edge absorption band at room temperature. They also could decorate dislocations lying within 30 microns of the surface by means of exposure to tungsten filament lamps and mercury lamps. One interesting observation was that the silver specks (decoration) did not grow indefinitely but reached a maximum size.

In pursuing this technique, Jones and Mitchell⁵ found the decoration to be dependent upon individual properties of the crystals, while the depth of decoration varied with exposure duration. Parasnis and Mitchell⁶ studied copper-doped silver chloride but their results did not settle the problem of impurity effects on decoration, as illustrated by silver formation in silver bromide.

While the Hedges-Mitchell method does produce decoration to a depth of $\sim 30\mu$, it is far from providing decoration for depths approaching the thickness of the widely used 600μ G-5 emulsions. This limitation of decoration-depth at room temperature was overcome by modifications developed by the present authors.

In 1960, using a technique first developed by Haynes and Shockley to study electronic properties of silver halides, it was demonstrated at U.N.C. that room temperature decoration in silver chloride crystals could be obtained when photoelectrons were swept into the crystals.⁷ The interesting factors here were the ability to achieve decoration at room temperature and the large thickness (5000μ) of the decorated region. Thus were solved the problems of high temperature anneals for decoration and the shallow decoration depths of the Hedges-Mitchell technique for silver halides.

The earliest studies were plagued with considerable difficulties in producing consistent data from adjacent crystals from the same boule. Moreover, no consideration had been previously given to the effects of minute amounts of impurities present at the level of several parts per million. It was the difficulty in producing consistent data which previously had caused the general abandonment of this technique before any consideration was given to the second factor of impurities. Whereas earlier investigators had obtained inconsistent

results for crystals from the same ingot, Childs⁸ reported that those failures were caused by a severely deformed surface region containing an extremely high density of electron traps. He showed that careful removal of this deformed region resulted in good penetration of photoelectrons in all crystals. Later, he showed that the ability to decorate also depended upon the impurity content of the crystals, and that crystals with greater than about 10 ppm of Al, Fe, and Ni would not decorate.⁹ These elements, notably nickel, were assumed to be very good electron traps, an assumption in agreement with earlier photoelectronic studies by Michel and Sliker.

The paths of charged particles have thus been made microscopically visible as a result of sweeping photoelectrons into silver chloride at room temperature. We now present a brief description of that method and some of its results.

d. Decoration of Radiation Induced Imperfections of Charged Particles in Silver Chloride

Experimental Methods

In 1948 Haynes and Shockley performed unique experiments in which they measured various electronic properties of silver chloride. In these experiments, the crystals were placed between two quartz plates each with an outer surface of an electrically conducting, ultraviolet-transmitting thin film. A potential difference of several kilovolts charged the resulting capacitor, the crystal being the major dielectric. At the end of the charging cycle, the internal field was, nevertheless, zero, having been cancelled by ionic charge drift within the crystal. The external field was then abruptly removed and an ultraviolet lamp was flashed. The internal field immediately became several kilovolts/cm by virtue of the ionic surface charges; this field has a time constant for decay of a few hundred microseconds. The light flash duration of one microsecond and the electron lifetime of several microseconds were both much less than this time constant; therefore, with the light being absorbed

within one micron of the negatively charged surface, the photoelectrons were swept toward the opposite positively charged surface.

The decoration mechanism is as follows. An absorbed quantum forms a chlorine atom and an electron near the surface, from which the chlorine escapes. The electron, driven into the crystal by the polarization field, becomes trapped at an imperfection. An interstitial ion, with a jump frequency of about 10^{11} per second at 300°K , combines with the trapped electron, thereby forming a silver atom. This atom may subsequently trap another electron, with the process repeating many times, to form microscopic specks of silver atoms, each probably nucleated at charged dislocation jogs.

Decoration of Imperfections Induced by Primary Cosmic Rays, Proton Collisions, and Alpha Particles

The apparatus used in the experiments at U.N.C. provided potential differences of two kilovolts, pulsed at a rate of 1000 per second. The light flash provided 10^9 photons per cm^2 at the crystal surface. With this apparatus, the first decoration of radiation tracks in silver chloride was obtained in 1961.¹⁰ Since that first report and continuing up to the present date, there have been numerous articles on the use of this technique to study: particles produced by collision of singly charged relativistic particles with Ag and Cl nuclei; tracks of non-relativistic and relativistic particles of machine accelerated carbon and oxygen ions; primary cosmic radiation; and identification of heavy ions in AgCl single crystals by multiple coulomb scattering.

Because of the well-known photosensitivity of silver chloride, we should mention that the decorated tracks are nevertheless stable against visible illumination. There is no qualitative change when decorated tracks are exposed to Wratten K2 filtered microscope illumination for 40 hours and to the noon-day sun for 30 minutes.

References

1. F. Seitz and J. S. Koehler, Solid State Physics 2, 377 (1956).
2. J. A. Brinkman, J. Appl. Phys. 25, 961 (1954); Am. J. Phys. 24, 246 (1956).
3. J. T. Bartlett and J. W. Mitchell, Phil. Mag. 3, 334 (1958).
4. J. M. Hedges and J. W. Mitchell, Phil. Mag. 44, 223 (1953); Phil. Mag. 44, 357 (1953).
5. D. A. Jones and J. W. Mitchell, Phil. Mag. 2, 1047 (1957).
6. A. S. Paronis and J. W. Mitchell, Phil. Mag. 4, 171 (1959).
7. C. B. Childs and L. M. Slifkin, Phys. Rev. Letters 5, 502 (1960).
8. C. Childs, M. S. Thesis, University of North Carolina (1959).
9. C. Childs, "Impurity Effects on Decoration and Speculation on Self-Decorating Silver Chloride Crystals," Reunion de Travail sur l'enregistrement des traces de particules chargees dans les cristaux, CNRS Strasbourg (1963).
10. C. Childs and L. Slifkin, Bull. Am. Phys. Soc. 6, 52 (1961).

4. Crew Support

There will be no functions or demands on either the shuttle crew or the ground support crew.

5. Supporting Studies

The supporting studies will include determination of the effects of temperature on tracks, in both Bridgman-grown and Czochralski-grown crystals. Also, further studies of the effects of impurities, dopants, and replacement of AgCl by AgBr would be desirable.

There are no anticipated obstacles or uncertainties.

B. Experimental Hardware

1. Modules: Four Flight and Four Back-up

Each flight module will be 12" by 12" by 3" with a mass of approximately 8 kilogram. Total flight mass is 32 kilograms.

2. Power

There will be no required data or power systems for or in the modules.

3. Electromagnetic and Vibration Susceptibility and Production

There is no susceptibility to or production of electromagnetic interference, contamination, acoustics, vibration, or any other interferences during ground handling, launch, and recovery.

4. Experiment Initiation Requirements

There is no requirement to signify experiment initiation.

5. Ground Support Equipment and Operations

There will be no ground support equipment or ground operations.

6. Special Requirements

There are no requirements for special electrical, mechanical, or shield controls.

7. Temperature Limits

The preferred temperature limits during handling, launch, space exposure, and recovery are -20°C to $+40^{\circ}\text{C}$.

8. Pre-launch and Post-launch Support

There is neither a pre-launch nor post-launch support requirement.

9. Present Stage of Development

The present state of development includes knowledge of the effects of various dopants (trace impurities) on the formation of stable

latent images of tracks in silver chloride crystals. Such crystals are now grown up to 2 cm by 2 cm by 20 cm by the Bridgman method. Samples from properly doped crystals have been employed as dosimeters for various heavy ions.

10. Design Life of Experiment

The design life of the experiment is a minimum of six months, the expected duration of the LDEF flight.

11. Desired Location on LDEF

The desired location of the modules is an experimental tray along the outer circumference of LDEF, and the modules may be placed under certain other experiments such as those studying micrometeorites.

C. Data Reduction and Analysis

The data reduction will include that which is obtained during calibration at the heavy ion accelerator at the Lawrence Radiation Laboratory at Berkeley, with the analysis showing track grain density and track width as a function of particle charge, velocity, and calculated energy loss of the particles in the crystals. These measured parameters will be compared with the crystals' dopants, electrical conductivity, and optical properties. Reports of these findings will be made quarterly.

D. Results Expected

The basic objective of the LDEF silver halide experiment is to test and evaluate the use of silver halide crystals as heavy ion dosimeters for long-duration space flights. This objective will be accomplished by two different sets of data obtained after the LDEF flight. The first set of data will provide actual information on the heavy ion component of the cosmic radiation, including charge distribution, energy, and flux. The second set of data will show what, if any,

effect the space flight had on the ability of the crystals to perform as reproducible heavy ion dosimeters when exposed to the environmental conditions of extended space flights.

E. Shuttle Crew

There will not be any tasks for the shuttle crew.

F. Recovery Requirements

There are no special recovery requirements except that the flight modules should be maintained in the same temperature range as they experienced during flight and they must subsequently be returned to the University of North Carolina for processing.

II. MANAGEMENT PLAN AND COST PLAN

A. Management Plan

1. Principal Investigators

Charles B. Childs, of the Materials Research Center of the University of North Carolina at Chapel Hill, is designated the Principal Investigator. Dr. L. M. Slifkin, of the Department of Physics of the University of North Carolina at Chapel Hill, is designated the Co-investigator.

2. Materials Research Center of the University of North Carolina at Chapel Hill

The Materials Research Center for several years has been engaged in developing methods for preparing high purity and deliberately doped silver halides, some of which have been used in fabricating crystals used as heavy ion dosimeters. The manufacture of those crystals is now a routine process.

3. Work Plan for Test Modules

The test modules will involve work at the University of North Carolina, NASA Langley Research Center, and the Lawrence Radiation Laboratory at Berkeley.

a. University of North Carolina

The Principal Investigator will procure the starting materials for

making the high purity silver halides, synthesise the silver halides, and have them analyzed for trace impurities. He will then use the tested material in growing specially doped crystals by both the Bridgman and Czochralski methods. Those crystals will then be used for preparing small samples of silver halides for the test module. The test module will be designed at the University of North Carolina, with the design being approved by Langley Research Center before actual construction at the University of North Carolina. After design approval has been obtained from Langley Research Center, the module will be constructed and crystals will be assembled into the module for testing at Langley Research Center.

b. Langley Research Center

The assembled test module will be submitted for the LDEF environmental tests at Langley Research Center, including both temperature and mechanical tests. Any failure resulting during the tests will be analyzed for its source and modifications made, and the resulting new design will be given the complete LDEF series of environmental tests.

c. Lawrence Radiation Laboratory at Berkeley

The assembled test module, after passing the environmental tests at Langley Research Center, will be exposed to relativistic heavy ions ($Z \geq 4$) at the Bevalac, at the Lawrence Radiation Laboratory at Berkeley. The module will then be taken to the University of North Carolina at Chapel Hill for the final evaluation.

d. University of North Carolina at Chapel Hill

The module exposed to heavy ion at Lawrence Radiation Laboratory will be disassembled. The silver halide crystals will then be divided into

two groups. One group will be decorated immediately; whereas the other group will be exposed to the expected LDEF temperature cycle and then decorated. Comparisons will be made of the decorated tracks in the two groups.

4. Work Plan for Modules

- a. A critical design review will be made by Langley Research Center before beginning construction of flight modules and this construction will be correlated with the LDEF schedule.
- b. The crystals and eight modules will be constructed at the University of North Carolina after all testing and design approval has been obtained from Langley Research Center.

5. Progress Reports

Progress reports will be submitted quarterly and at the conclusion of each stage of the testing and construction.

B. Facilities and Equipment

1. Major Facilities in Existence

- a. The LDEF environmental tests will be performed at Langley Research Center.
- b. The exposures to heavy ions will be done at the Lawrence Radiation Laboratory at Berkeley.
- c. The Materials Research Center of the University of North Carolina will grow the silver halide crystals, fabricate test crystals, construct flight modules, and decorate the crystals after exposure to heavy ions at the Lawrence Radiation Laboratory and after the LDEF flight. The Center will provide a Leitz Ortholux microscope with all necessary accessories.

2. Laboratory Equipment to be Acquired from Sources Outside
the University of North Carolina

a. Furnance Equipment

It will be necessary to purchase the temperature regulators, power equipment, and related equipment for the five Bridgman furnaces to be constructed at the University of North Carolina. This equipment represents "on-the-shelf" items, with a lead-time of approximately one month after being ordered.

b. Automatic Polishing and Etching Equipment

Equipment will be purchased for the automatic polishing and etching of samples for the modules. These pieces of equipment are "on-the-shelf" items and could be obtained within one month after being ordered.

c. Crystal Pulling Furnace

The pulling furnace lead-time is estimated to be three months.

d. Microscope Equipment

The special high power, high numerical aperture microscope objective and condenser, for working with specimens with an index of refraction of silver halides, will have a lead-time of six to nine months .

3. Laboratory Equipment to be Manufactured at the
University of North Carolina

The University of North Carolina will construct the five Bridgman furnaces for growing silver halide crystals. These five furnaces will be duplicates of the furnaces which are now being used for silver halide crystal growth. The lead time for these furnaces is from two to three months.

C. Cost Plan and Funding Plan

1. Direct Labor, Overhead, General and Administrative

The table below gives the total Cost Plan and Funding Plan for the proposal.

COST PLAN AND FUNDING PLAN

Item	Year				
	1	2	3	4	5
Equipment	\$18,100.00	1,000.00	-----	-----	-----
Modules	9,000.00	12,000.00	12,000.00	-----	-----
Langley Trips (2)	240.00	240.00	120.00	120.00	240.00
Supplies, Data Reduction, Computer Time	1,215.13	972.61	1,341.94	1,456.53	1,863.53
Berkeley Trip	600.00	600.00	-----	600.00	600.00
Labor (Principal Investigator, 25% of time)	4,125.00	4,335.00	4,550.00	4,725.00	5,015.00
Indirect Overhead (48.13% of labor)	1,985.36	2,086.44	2,189.92	2,274.14	2,413.72
Fringe Benefits					
Social Security, 5.85%	241.31	253.60	266.18	276.41	293.38
Retirement, 9.12%	376.20	395.35	414.96	430.92	457.37
Hospitalization, 1/4 of \$19.50/month	58.50	58.50	58.50	58.50	58.50
Disability, 1/4 of \$234	58.50	58.50	58.50	58.50	58.50
Total	\$36,000.00	22,000.00	21,000.00	10,000.00	11,000.00

2. Materials

The major item among the materials is the special high purity silver nitrate manufactured by the Eastman Kodak Company. This material is sold only in bottles of 50 gms each, with a current price of \$147.80 per 500 gm. bottle. This price is expected to increase during the next two years because of the projected increase in the price of silver. The total price of materials per flight module is approximately \$2,800.00 with each module having 100 crystals, each 2 cm by 2 cm by 5 mm thick.

It is estimated that each module will require approximately \$200.00 of additional materials for such things as manufacture, track decoration, and recording of data.

The price per module will be approximately \$3000.00 each. There will be four flight modules and four back-up modules with a total cost of \$24,000.

3. Special Equipment

Special glass manifold, vacuum system, and related parts	\$1,600.00
High purity water, filters, water storage containers, and drying trays for making high purity silver halides	\$1,000.00
Crystal-Pulling Furnace with related equipment	\$3,500.00
Five Bridgman furnaces with related equipment	\$6,000.00
Miscellaneous equipment (timers, pulse generators, power supplies, special lamps, polishers, non-strain crystal cutters)	\$6,000.00

4. Travel for One Person

Langley Research Center: eight trips to coorelate design, testing, and installation of modules, approximately two days per trip:	\$ 960.00
Lawrence Radiation Laboratory at Berkeley: testing, calibration of crystals by exposure to heavy ions, approximately five days per trip due to uncertainty with which the Bevalac operates, four trips:	\$2,400.00
TOTAL:	\$3,360.00

5. Allocation of Cost

Design, including first year labor, indirect cost, one module, two trips to Langley Research Center and one trip to Berkeley	\$ 18,000.00
Hardware, including all equipment and supplies, modules, labor	\$ 59,000.00
Testing, including the calibration trips to Lawrence Radiation Laboratory	\$ 6,000.00
Data Reduction, analysis, publication cost, and computer time	\$ 17,000.00
TOTAL:	\$100,000.00