5.5 Development of Thick Emulsion Layers

Some of the special problems presented by concentrated nuclear research emulsions arise because great thicknesses must be processed with the general requirement that the development be uniform throughout the emulsion volume. A second requirement is that emulsion distortion in processing must be kept small.

Emulsions of 50, 100, and up to 200 μ in thickness can be developed at 20°C, as described in Section 5.3. This development is similar to that of photographic plates if the developing solution is diluted or the concentration of developing agent alone is reduced so as to keep the time of development long compared to the time of penetration of the solution into the emulsion. An obstacle exists to the extension of this method to still greater thicknesses. The developing agent becomes exhausted or is absorbed by the gelatin as the solution penetrates the emulsion. Attempts have been made to carry out two-solution development (BD 48) or development in two stages (B 56.1), but it is believed that these methods have now been largely superseded.

Adding potassium bromide to the developer as a restrainer and keeping the pH of the solution low are valuable means for slowing the development. Moreover, the rate of penetration of the developer into the emulsion can be increased by as much as a factor of two if the emulsion is presoaked in distilled water prior to development. However, the most important single device for obtaining uniform development of thick emulsions is the adjustment of the relative development and diffusion rates by control of the temperature. Either a two-temperature method (DOP 48) or isothermal development at a reduced temperature (S 53, Y 55, BF 60) is possible.

Except as necessary to eliminate temperature gradients, one does not agitate the developing baths. Agitation tends to increase the difference between the development received by the surface layer of emulsion and that of the deeper layers.

When emulsion is immersed in cold developer, sufficient time must be allowed for the developer to soak through to the glass. For amidol developer at 5°C, this is about 2.5 hr for 600 μ emulsion. At least 7 hr must be allotted for amidol developer to penetrate 1000 μ emulsion. Most other developing agents penetrate more slowly than amidol or amidol bisulfite.

Presoaking of the emulsions for 3 hr in distilled water was studied by Dainton *et al.* (DGL 51) as a means to shorten the penetration time of developers in all thicknesses of emulsion. The average reduction factors were for azol, 1.34 ± 0.05; D-19b, 1.70 ± 0.13; normal amidol,

1.86 ± 0.12; and amidol bisulfite, 1.65 ± 0.14. They also showed that amidol and amidol bisulfite penetrated equally fast. Their penetration time was less than half that required for azol or metol-hydroquinone solutions. The time of penetration for amidol in presoaked emulsion increased with the 1.4 power of the thickness and decreased with rising temperature. In Section 4.7 we found that diffusion times should theoretically depend on the square of the thickness, and other investigations (H 52, H 52.1) have not confirmed the 1.4 power-dependence.

Dainton *et al.* gave the pH values of their solutions as 10.0 for D-19b, 11.5 for azol, 7.2 for amidol, and 6.7 for amidol bisulfite. These figures could be reflected in their results. The solutions of high pH penetrated more slowly. An effect of the high pH is to cause the emulsion to swell greatly, thus complicating the diffusion problem. It may also be a partial explanation of the disagreement found in the law of the penetration time.

In this book we shall assume that the theoretical "thickness squared" law is a good approximation. For all large thicknesses, immersion-diffusion times may be calculated by this law from the times recommended for 600 μ emulsion.

The two-temperature method was developed by Dilworth, Occhialini, and Payne (DOP 48), by Wilson and Vaneslow (WV 49), by Dilworth, Occhialini, and Vermaesen (DOV 51), by Dainton, Gattiker, and Lock (DGL 51), and by Stiller, Shapiro, and O'Dell (SSO 52), and others, in an effort to achieve uniform and complete development.

As tabulated by Dilworth *et al.* (DOV 51), the ideal developing agent for the temperature method should: (1) be capable of penetrating the emulsion quickly, (2) possess a moderate or high temperature coefficient, (3) produce negligible background fog and stain in the emulsion, (4) be stable at temperatures up to 30°C, and (5) have a pH as close as possible to that of the emulsion.

Of the many agents tested in various experiments most were eliminated because they fell too far short of one or more of the above ideal characteristics. Amidol or amidol bisulfite are most nearly ideal.

The kinetics of development of G.5 emulsion in amidol bisulfite and in ID-19 developers has been studied by de Carvalho and da Silva (DD 60). They used the experimental data of Herz (H 52). It was found that a general relation (Sheppard's equation)

$$D = D_\infty \{1 - \exp [-K(t + t_0)]\}$$

fitted the data if t_0 is taken to be a time parameter allowing for the cold development and induction time. The quantity, D, in this case is the grain density and K is a constant. They also found that in the range

21° to 33°C, that the maximum development rate varied exponentially with $1/T$, T being the absolute temperature. This behavior follows from Arrhenius' equation:

$$\text{Rate} = Ae^{-E/kT}$$

where A is a constant, k is the Boltzmann constant, and E is an activation energy.

Dahl-Jensen (D 60.1) found a somewhat different time-dependence of the plateau blob density in hypersensitized K.5 emulsion. This is

$$B = 34[1 - (t/10)^{-3/2}]$$

where t is presumably in minutes and B is the number of blobs in 100 μ.

Birge *et al.* (B-W 54) also studied the kinetics of development. They found that a grain density of as much as 14 per 100 μ was developed in G.5 emulsion by the 5°C cold soak. The additional grain density, G_D, contributed by the warm stage could be fitted by a function of the form

$$G_D = G_R[1 - e^{-\lambda(T)t}]$$

where G_R is its asymptotic value, t is the time of the warm stage, and $\lambda(T)$ is a function of the temperature.

Development of thick emulsion layers is usually carried out with a variation of one of two conventional amidol formulas, which we shall designate the "Bristol formula," stemming from the work of Dainton, Gattiker, and Lock (DGL 51), and the "Brussels formula," from Dilworth, Occhialini, and Vermaesen (DOV 51). Each of these is capable of yielding quite a satisfactory product. The chief difference between them is that the Bristol formula contains sodium bisulfite or sodium metabisulfite whereas the Brussels formula uses a buffer of boric acid. The boric acid is good for this purpose but is difficult to put into solution. The substitution of citric acid, as recommended for processing the Russian emulsions, (Section 5.2) has often been suggested.

The Bristol formula is the following: to each liter of distilled water one adds 7.2 gm of anhydrous sodium sulfite, 1 gm of sodium bisulfite, 8.7 ml of a 10 % solution of potassium bromide, and 3.25 gm of amidol. The pH of the Bristol solution is about 6.6.

Sodium bisulfite solution in the United States is commonly prepared using sodium metabisulfite, $Na_2S_2O_5$. About 0.91 gm per liter of this compound is equivalent to 1 gm of sodium bisulfite.

The Brussels' formula is: to each liter of distilled water one adds 35 gm of boric acid, 17.5 gm of anhydrous sodium sulfite, 8 ml of a 10 % solution of potassium bromide, and 4.5 gm of amidol. This has a pH of about 6.4.

All amidol developers must be made up fresh for use because the amidol quickly oxidizes.

In preparing the developing baths, amidol must be added only to solutions from which oxygen has been eliminated by the presence of sulfite, or the amidol itself will react with dissolved oxygen and be destroyed. Usually the amidol and sodium sulfite are added just before development. One may keep a stock solution containing the other ingredients.

For the immersed hot stage (see below) Brussels' formula with 15 gm per liter of sodium sulfite is used because 17.5 gm starts to cause deposition of colloidal silver. "Dry" hot stage development requires more protection of the amidol from oxygen. Oxidation of amidol is revealed by staining of the gelatin—usually to a yellowish hue.

The most important variation of the two-temperature development procedure is in the means for carrying out the hot stage. The ideal temperature-cycle development contemplates (*a*) an emulsion layer uniformly saturated with developer at a temperature too low for silver reduction to proceed at an appreciable rate, (*b*) development to take place with the emulsion isolated so that new developer cannot penetrate it, while the temperature is raised to a uniform value throughout the emulsion volume, (*c*) development to be slowed throughout the emulsion volume by reduction of the emulsion temperature and immersion in a stop bath of low pH.

The procedure of Dilworth *et al.* (DOV 51) was to lower the emulsion temperature while it was immersed in the distilled water of the presoak. They state that the penetration of the developer starts to become rather difficult when the temperature is less than 5°C, and they did not carry it below that point. Only 15-20 μ of the ends of proton tracks were visible after the cold developer penetrated the 600 μ emulsion used by Dilworth *et al.* Ilford G.5 emulsion now appears to have altered in behavior, because development proceeds much farther in the cold solution; often minimum tracks are visible before the hot stage is commenced.

The hot stage of Dilworth *et al.* is to be carried out while avoiding a development gradient as the temperature is raised. They mention the three possibilities for accomplishing this: (*a*) diluting the developer solution; (*b*) covering the emulsion surface with oil, carbon tetrachloride, glass coated with beeswax, etc., to prevent the entering of further developer; (*c*) warming the plates dry.

The method (*c*) has been utilized extensively by Dilworth *et al.*, and apparatus for carrying out this procedure has been described by them (DOV 51, IO 58). Method (*b*) has not had much use.

In addition to the prime purpose of providing uniform development,

methods (*b*) and (*c*) prevent further swelling of the emulsion. When method (*a*) is used, specific steps must be taken to avoid further swelling if there is any reason to prevent it.

At several international conferences the writer has advocated an immersed hot stage in a solution of altered composition, the time and temperature of the immersion to be optimized (B 57.2, B 58.2, B.60). Some variation of the method (*a*) suggested by Dilworth and her collaborators seems ultimately to be the best approach for the following reasons:

1. One wishes to obtain uniform development from plate to plate as well as within a plate. To develop all the plates of a large stack by the dry, hot stage method with identical times and temperatures is not really possible. It also requires much space and a large amount of equipment.

2. There are many critical operations in method (*c*). Equipment unreliability, lack of operator care, or operator inexperience, can cause a processing catastrophe. Development of a large and valuable stack must be carried out in such a way that failure is an extremely remote possibility.

3. The amount of work involved in method (*c*) is large.

Although in the hands of an expert method (*c*) can give plates of good development uniformity, quite the opposite result often is obtained. Some of the reasons for such failures are the following: in order to prevent developer oxidation at the surface, an inert gas should be put in contact with the emulsion. This is often done inefficiently. Frequently the surface on which the plates are heated is not free of lateral temperature gradients. As Dilworth *et al.* point out, 1°C is a very important temperature difference in development. The cover of the processing tank above the emulsion must have the same temperature as the surface below, or there will be an exchange of heat by radiation and a temperature gradient will exist through the emulsion. Since heating is done by water jackets, etc., thermal lags caused by varying thermal conductivities and heat capacites are very likely to subject different plates to different temperature cycles. Radiofrequency heating has been used, but it has the same objection. In addition, the mopping up, as recommended, of excess developer remaining on the emulsion surfaces must be regarded with suspicion as a cause of distortions. (When this operation is carried out, photographic blotting paper is recommended for the job.)

It is the writer's opinion that the dry, warm stage produces conditions favoring corrosion and also may contribute to spurious scattering (Chapter 8).

A good compromise of the ideal with practical considerations may be found in an immersed hot stage. If present knowledge is utilized, the effect of fresh developer entry through the surface can be minimized.

In early trials by the writer, simple dilution of the normal solution by an equal volume of distilled water for the hot stage was so successful in providing a uniform grain density that no change in this procedure was felt necessary until a number of other variables were better controlled. Subsequently, precise measurements were able to establish that excess surface development remained in certain emulsion stacks.

The effect was found to be reversed, however, in stacks which had experienced a somewhat lower silver ion concentration in the fixing solution. Any experiments on the uniformity of development must also accurately control the fixation corrosion or the results are of little value.

Something less than half-normal concentration of hot stage developer is probably optimum. Curves have been given by Hauser (H 58.1), however, which indicate that for Agfa K.2 emulsion a concentration of 50 % is the best. Hooper *et al.* (HDN 60), under the same conditions, found that the grain density in G.5 emulsion near the glass was lower than that not so deep in the emulsion. They found a reduction at the surface, however. Such an effect is not normal with this type of development unless silver etching, latent-image fading, etc., has taken place.

With 50 % dilution the writer has found that the grain density that is typical, if not completely satisfactory, remains in a range of $\pm$ 5 % from the average. Perhaps 20 blobs per 100 μ in the middle layers of the emulsion, 21 at the surface, and 19 next to the glass are found at the minimum of ionization.

By simple refinements and variations of the technique, further improvements of the uniformity can be effected. Accidental variations of conditions occasionally, in fact, have yielded plates in which any nonuniformity of the grain density was very difficult to detect.

The immersed warm stage has recently been re-examined by Heughebaert, Igiuni, and Occhialini (HIO 58). They have tested a number of modifications of the method, and were favorably inclined toward a solution in which the amidol and sodium sulfite concentrations are lowered by about 40 % and the potassium bromide content increased from 0.8 gm to 2.2 gm per liter. In addition they proposed to add 12 % sodium sulfate.

Later, at Milan, a warm stage was tried in "dummy" developer containing no amidol (D 60.3). Only a 6 % development gradient was found. The surface 50 μ suffered a marked reduction of grain density, however. The developer containing sulfate was found not to improve the quality of the results.

Hooper *et al.* (HDN 60) found a compromise on lowering the amidol concentration from 4.5 gm to 1.9 gm per liter, and on raising the potassium bromide concentration from 0.8 gm to 1.6 gm per liter for the immersed warm stage.

Some tests of the dependence of grain density on developer components also have been made by the writer's research group. Data by D. DeLise and C. Cole indicate, for example, that both the track and background grain density fall slowly (for a fixed hot-stage development time of 30 min at 22°C) with increasing potassium bromide content of the developer. This amounts to a reduction to two-thirds in going from 2 to 12 ml of 10 % potassium bromide per liter.

Meulemans (M 54.1) studied the dependence of the grain density on the amidol concentration. At 25°C, p.5 gm per liter of amidol produced a grain density of about 16 per 100 μ; 1 gm per liter, 22 per 100 μ; and 4.5 gm per liter, 31 per 100 μ. The opacity of the emulsion also varied about proportionally.

Fatzer *et al.* (FWGH 57) reported that at temperatures below 14°C the development with amidol is improved by increasing the concentration of sodium sulfite. Sodium sulfite requires rapid stirring while it is added slowly. It must be dissolved *before* the amidol.

To determine the effects of the time and temperature variables, tests were made at the Lawrence Radiation Laboratory. The results are illustrated by Table 5.5.1. These data were obtained by D. DeLise. He measured the blob density on the plateau of ionization in K.5 emulsion. The emulsion was developed in Bristol developer. It was 200 μ in thickness and was presoaked in chilled water.

TABLE 5.5.1

BLOBS PER 100 μ VERSUS TIME AND TEMPERATURE OF DEVELOPMENT

Temperature (°C)	Development time (minutes)				
	30	50	70	100	150
0		No tracks		<7.5	15.1
5	<9	10.2	11.3	14.0	18.2
10	11.8	13.3	15.6	22.7	22.6
15	14.5	17.9	19.6	25.1	25.5
20	20.4	23.0	24.3	28.0	29.6
25	24.3	27.2	28.8	31.1	Opaque
30	26.2	29.0	30.3	Opaque	Opaque

Colloidal silver either uncolored or as dichroic fog deposits in the gelatin under certain conditions. The effect increases with sulfite concentration almost linearly for lower concentrations of sodium sulfite, and tends to saturate at higher concentrations. The effect increases rapidly with temperature and pH. The deposit of a surface layer of silver, often with a mirror finish, parallels this behavior. These effects are associated with physical development which coarsens the silver grains and which results from the presence of a silver halide solvent in the developer. The effect may also occur in the fixing solution if the silver ion content is high, and "minigrains," small random grains in the emulsion, can be formed when the silver concentration is high, according to Gailloud (D 60.3). This effect is the reverse of the etching that may occur near the surface in the fixer.

The silver deposit in the developer can be controlled, according to Bonetti *et al.* (BDS 58), by putting a metal plate a few millimeters above and parallel to the surface of the emulsion. An excess of surface silver can also be bleached rather simply using a prescription given by Bonetti. To 1 liter of water 100 gm of copper sulfate, 100 gm of sodium chloride, and 25 ml of concentrated sulfuric acid are added. A short period of immersion in this bath, the time to be established by trial, is followed by washing and fixing of the plate.

The writer has tried this formula on plates having an opaque surface deposit of silver. With immersion times of 5-10 sec it proved to be a useful process. When the surface opacity is thus removed without rubbing, no gelatin layer is lost. If it is overdone, the tracks can be redeveloped by very dilute developer.

The development of NIKFI emulsions is rather conventional. For example, a development procedure for 400 μ NIKFI emulsion was described by Engelhardt *et al.* (EHK 60). The mounted emulsion is soaked in distilled water at 3°C for 2.5 hr. It is then placed for an additional 2.5 hr in a developer solution at 3°C. The developer composition is as follows: to 1 liter of distilled water, 12 gm of sodium sulfite and 2.5 gm of amidol are added. The pH is brought to 6.6 by the addition of about 2 gm of citric acid.

The hot stage of development is carried out at 24°C. The plates are taken out of the cold developer, gently wiped with filter paper, and immediately placed in a thermostat where they remain for 40 min.

Probably a large part of the processing damage which is evident as distortions of the finished emulsion occurs during fixing, dilution, and washing rather than during development. It is in these subsequent operations that the maximum swelling occurs, and the gelatin then is very soft and has little strength. Bonetti *et al.* (BDO 51) state that the

temperature changes to produce the hot stage of development probably have less deleterious effect on the emulsion than the later stages unless the developer is more alkaline than pH 10 or the temperature higher than 32°C. Sudden changes of temperature and/or pH in development, however, also were thought to be possible causes of distortions.

Bonetti (D 60.3) also reported recently that spurious scattering appeared to be lowest in emulsion that had had wet, hot-stage development.

To develop emulsion of any thickness, greater uniformity and probably less distortion can be obtained by using a low temperature and a prolonged time of development. Although unconventional in ordinary photography, there are no practical reasons why development cannot proceed for a day or so.

There are many other considerations, however, some of which are conflicting, so that one must compromise. A grain density near the maximum possible usually must be realized, distortion minimized, and the fog level relative to the track-grain density be held in check, all at the same time.

The Bristol formula was employed (BF 60) to study the possibility of prolonged development of G.5 emulsion at a very reduced temperature. A fair minimum blob density of 17.4 per 100 μ was achieved with a time of 10 hr at 0°C, but the fog level then was 22 grains/10^3 μ^3—too high to be satisfactory. This technique for obtaining good uniformity of development probably should be restudied with hypersensitized emulsions. Our result, nevertheless, confirms the conclusion of Herz (H 52.1): "For equal background a higher minimum grain density can be obtained with a warm stage than without one. This is the real justification for the temperature-cycle method—*uniformity* can be obtained with single temperature development whenever the time the developer needs to penetrate is short compared with the total developing time."

The ultimate in development procedure is not yet in sight. The approach, thus far has been almost completely empirical, and there are a very large number of variables to be simultaneously optimized. To complicate matters there appears to have been a change in the emulsion characteristics so that presumably well-established facts of several years ago are no longer true (O 57). Nevertheless, the average quality of the development in most laboratories probably is slowly improving, thanks to small refinements.

Many details remain to be studied. For example, when the temperature of emulsion is raised or lowered, an ambiguity always exists as to how and how rapidly to make the change.

The writer's procedure of immersing the cold-soaked emulsion in

warm developer could be altered by heating the tank of diluted developer. Some argue that this would eliminate "thermal shock," whatever that means. The uniform heating of a large volume of solution, which should at the same time be kept stagnant, offers difficulties. As far as the writer is aware, there is no distortion damage inflicted on the emulsion in this development procedure comparable to that sustained in mounting, that caused by excessive swelling, or that occurring during most drying procedures, so that these steps should be controlled before worrying greatly about what is probably a very minor effect.

5.6 Stopping and Fixing

The action of the developer can be stopped by lowering the temperature and the solution pH. A suitable stop bath is distilled water containing 0.2 % acetic acid (pH 3.2). The temperature of the stop bath should be about 5°C, and it is best if the emulsion temperature can be brought gradually to 5°C before immersion in the fixing bath. The time in the stop bath should be about 2.5 hr for 600 μ emulsion, and for another thickness, the time should be varied approximately with the square of the emulsion thickness. Prolonging the stop-bath immersion much beyond the time recommended is dangerous as reactions leading to dissolution of the silver grains take place.

Fixation is the process of dissolving the silver halide in an alkali thiosulfate solution. Sodium thiosulfate or "hypo" is most commonly employed in the fixing solution, but if the ammonium ion replaces the sodium, more rapid fixation is usually considered possible. According to Lonchamp and Braun (LB 54) this depends on the temperature and the concentrations when fixing Ilford C.2 emulsion. At 20°C, and at concentrations of 1.5 to 2.7 moles per liter, the times of fixation are minimum and equal. At 16°C the minimum times are the same but occur at 1.5 moles per liter for sodium and about 3 moles per liter for ammonium. At 3°C ammonium thiosulfate fixing time has a minimum of 1.2 hr for 100 μ emulsion at 2.2 moles per liter, whereas sodium thiosulfate has a minimum of 2.2 hr at 1.7 moles per liter. These writers could find nothing to make inadvisable the use of ammonium thiosulfate for fixing up to 600 μ emulsion at 5°C, although it was not contested that ammonium thiosulfate attacks the reduced silver more easily than does sodium thiosulfate.

The time for fixation to be complete at first falls rapidly as the concentration of the thiosulfate is increased. It passes through a minimum which at low temperatures is more marked than at high. At 19°C the

minimum is very broad, extending from 300 gm per liter to 700 gm per liter for sodium thiosulfate. It occurs at about 400 gm per liter at 16°C, and at about 500 gm per liter at 7°C according to Lonchamp and Braun, Birge *et al.* (B-W 54) found lower concentrations to be optimum when bisulfite is also present. The minimum time, t, for fixation of 100 μ G.5 emulsion in the interval 7° to 28°C was observed by Lonchamp and Braun to be about $t = 1.6\ e^{-0.03\theta}$ hr, where θ is the centigrade temperature.

In addition to sodium thiosulfate, fixer usually contains Na_2SO_3 and/or $NaHSO_3$. The acid sulfite arrests the action of the developer and lowers the pH so that the emulsion swelling is kept within bounds. The sulfite ion removes dissolved oxygen, the presence of which in the fixer causes corrosion of silver in thick emulsion layers.

Ammonium chloride added to the fixer usually has a clearing effect on the emulsion, but may also etch the track grains. Lonchamp and Braun found that, whereas the speed of fixing C.2 emulsion was increased 60 % by the addition of 50 gm per liter of NH_4Cl, when the concentration of hypo was 300 gm per liter, much smaller effects were general, and in G.5 emulsion the time for fixation was even increased for high hypo concentrations. The different behavior of C.2 and G.5 emulsion was attributed to the difference in their content of iodide.

Acid fixer generally increases the time of fixation 30 % or thereabouts, but tanning agents have less effect. Wetting agents have no effect. The addition of 5 % sodium sulfate increases the fixation time by 40 % according to Lonchamp and Braun.

Agitation of the fixer, particularly flow of the fixer over the emulsion surface, reduces the time of fixation. Although it varies with the emulsion thickness, this factor may be as much as 2.

Fixing is speeded if the plates are processed with the emulsion facing downward, the ratio being about 0.7.

Swelling of the emulsion in processing has been studied by a number of people. Among others, Bonetti *et al.* (BDO 51), Meulemans and Mignone (MM 51), Heughebaert and Heughebaert (HH 58), Major (D 60.3), and Oliver (O 58.1) have reported on the swelling behavior in fixer. Other conditions being held constant, the swelling increases with increasing temperature. Meulemans and Mignone report a 40 % increase in going from 5° to 20°C.

Swelling curves in fixing solutions as a function of time and pH have been given by Heughebaert and Heughebaert (HH 58). When the pH was 6.8 the emulsion swelled more than when it was 4.8. Nevertheless, because corrosion is somewhat higher in acid solution, and the time of fixing longer, a pH of 6.8 was recommended.

Rather than put bisulfite or metabisulfite in the fixing bath, which they fear may corrode the grains during the long time required for fixing thick emulsions, Bonetti, Dilworth, and Occhialini (BDO 51) suggest that the swelling be restrained during fixing by the addition of 7 to 10 % of sodium sulfate. Longer fixing times are then required (see above) and the resulting product is not so clear as the gelatin emerging from standard fixer. Therefore they suggest the further use of an acid clearing bath (Section 5.12) to produce a transparent product.

In a later publication Bonetti (BDS 58) advocates lowering the fixer pH to 5.0 by means of acetic acid.

In the fixer, the silver halide solvent may oxidize silver which is highly reactive when finely divided. Oxidation tends especially to occur when the fixer contains dissolved oxygen. Boiling of the water shortly prior to making up the solution is recommended (P 57). Pure chemicals are also demanded.

Thuro and Paic (TP 56) propose the following fixing solution:

Sodium hyposulfite ($Na_2S_2O_3 \cdot 5H_2O$)	360 gm
Anhydrous sodium sulfite (Na_2SO_3)	5 gm
Glacial acetic acid	4 ml
Boiled distilled water	to make 1 liter

While to reduce the emulsion swelling and to increase the transparency of the emulsion, an acid fixer is required, suppression of corrosion is more complete in a basic fixer.

Braun (B 59) has made a study of corrosion in which he reconsidered the influence of the fixer composition, pH of the fixer, the silver ion concentration, the oxidation-reduction potential, E_{red}, of the fixer, and the volume of the fixing solution present. He concluded that there are at least two causes of silver-grain corrosion: (*a*) an effect of relatively little importance is the attack of the silver by the sodium thiosulfate or its products of decomposition and atmospheric oxygen under certain conditions of pH. (*b*) A more important effect is due to the amidol or its oxidation products, accelerated by the oxygen of the atmosphere.

After experimentation he recommends the following fixing solution, which is said to cause no observable corrosion of 600 μ emulsion:

Water	1 liter
Disodium phosphate	70 gm
Ascorbic acid	2 gm
Sodium thiosulfate	350 gm
Monosodium phosphate to make a solution of pH	6.5

This solution clears a 600 μ G.5 plate in about 50 hr at 4°C. The volume of fixer is 1 liter per 2 gm of silver bromide in the emulsion, and the

emulsion is to be immersed 20 cm beneath the surface of the fixer. Sodium bisulfite, according to Braun, increases the corrosion while sodium sulfite increases the difficulty of fixing and the turbidity of the plates. For an antioxidant, he employed ascorbic acid. Adding of amidol to the fixing solution was found to cause a rapid solution of the silver grains, but hydroquinone or pyrogallol merely caused coloration of the gel.

A well-known means advocated for the reduction of corrosion is to avoid a low silver ion concentration in the fixer. Bonetti *et al.* (BDS 58) state that at least 6 gm of silver per liter should be present in all stages of fixing, and they add old emulsion or solutions of KBr and $AgNO_3$ to the fixing bath until this figure is exceeded. Low temperature and freedom of the emulsion from oxidized developer they also believe reduce the tendency for corrosion.

If care is taken to insure that the volume of fixer is not too large, reasonable silver ion concentrations are attained in a few hours after the start of fixation. This precaution seems to have been sufficient to inhibit corrosion in the writer's experience.

A high concentration of silver should also be avoided. More than 15 gm or so per liter slows the fixing, and it is believed that silver complexes form in the gelatin. They make the emulsion turbid, and are hard to remove. Dahl-Jensen (D 60.3) has cited the gelatin tanning effect of the triply charged silver ion. The silver-containing solution in a hypo bath tends to settle to the bottom of the vessel. Gentle flow of fixer over the emulsion surface is recommended. Stratification is then avoided and the efficiency of fixing improved.

The fixing solution that has been used by the writer's group of the Lawrence Radiation Laboratory is made with 300 gm of sodium thiosulfate per liter of water. In addition, 22.5 gm of sodium metabisulfite is added. This brings the pH down as low as 4.3. Few corrosion problems have been encountered. This may be partly owing to the use of the immersed warm stage of development which keeps the air away from the emulsion. Care is also taken that the emulsion does not remain for long periods in solutions of low silver ion concentration.

Engelhardt *et al.* (EHK 60) vary the stopping and fixing procedure somewhat for NIKFI emulsions. Development is stopped by immersing the plates in 1/2 % acetic acid at 3°C for 2.5 hr. The plates are washed by immersion for 2.5 hr in distilled water (this step is not usual in other laboratories and its purpose was not explained). The fixing is carried out with a 30 % filtered hypo solution at 5°C which flows across the plates at 6 cm/min. The time allowed for fixing is 50 % more than the clearing time—about 40 hr total. In the hypo solution there is enough

potassium metabisulfite to bring its pH to 6.9. The quantity of fixer used is adjusted so that, while it contains no silver at the beginning of the fixation, at the end it contains 8 gm per liter. They observe no corrosion.

Recently Van Heerden and McEwen (VM 60) rechecked several points of uncertainty in connection with fixing solutions. A fixing bath was made up of 1 liter of water in which were dissolved 400 gm of sodium thiosulfate and 30 gm of sodium metabisulfite. At 6°C they found the clearing time of 600 μ K.5 or G.5 emulsion to vary with the silver ion concentration about as follows: 0, 45 hr; 12 gm/liter, 80 hr; 20 gm/liter, 120 hr; and 30 gm/liter, 240 hr. The clearing time was defined to be the time required to dissolve the last trace of opaque silver halide. At that time about 90 % of the silver has been removed. The swelling was maximum at this point. There the thickness was 2.2 times the original thickness.

They also investigated the effect of acetic acid buffer and of potassium alum hardener. The hypo buffered with acetic acid to pH 4.8 and 6.8 cleared the emulsion slightly more rapidly, but it swelled more. Buffering to a pH of 6.8 is recommended (HH 58).

The presence of 10 gm of potassium alum hardener caused the rate of swelling and shrinkage to decrease. No further swelling then takes place in the subsequent washing.

It was verified that corrosion decreases slowly with silver ion concentration.

The long time required for thick layers of emulsion to be fixed has stimulated suggestions for methods to shorten this time. E. M. McMillan and G. Occhialini each have proposed that pellicles be mounted on glass containing fine pores through which the fixer could be drawn (LB 54). This would also provide a means to obtain uniform development of the emulsion. No completely suitable glass has been found for this purpose, but it seems possible that something may be developed that would make this process practical.

Agitation of the fixing solution with a supersonic generator has been tried by the writer, and a small increase in fixing rate obtained. A well-designed and powerful sound generator may well turn out to be useful for this purpose.

5.7 Dilution and Washing

After fixation is completed, the solution concentration should be gradually reduced until no hypo can be detected in the processed emulsion. The operation must be carried out with care because at this stage

the emulsion is soft and greatly swollen. To limit the swelling, the temperature is kept low and the rate of dilution adjusted for minimum swelling. This rate has been established by Oliver (O 58.1). One should not introduce dilution water at a rate of more than 3 % of the tank volume per hour. A temperature of 5° to 10°C is satisfactory. The dilution can start soon after the emulsion has cleared. Generally, demineralized tap water is satisfactory for the purpose. After diluting for about 3 days the rate may be increased and continued until the test for hypo (Section 5.13) gives a negative result.

Sometimes the emulsion is poorly attached to the glass at certain points. Then in the fixer a small pocket of liquid may exist between the glass and pellice. When a plate having such a defect is subjected to dilution, the concentration gradient produces a pressure differential (osmotic pressure) tending to drive water into the pocket. The pocket swells and the bump will grow on the pellicle. In the affected area the emulsion becomes irreversibly stretched, and when dried it will not return to its undistorted condition. It is good practice to inspect the plates at the beginning of dilution for such blisters. With a hypodermic needle one may puncture each and withdraw the fluid so that the emulsion lies flat on the glass. It is convenient to attach the needle to an aspirator. Of course the needle always does some damage.

If a blister is not drained it will collapse on drying, but a ridge will be left at its periphery, and considerable track distortion in this area probably will be observed.

If the emulsion is not dried with alcohol, organisms may grow in the gelatin. Bonetti *et al.* (BDS 58) recommend the use of 0.1 % of santobrite in the wash water and a lowering of the temperature to 5°C to discourage such growths. They suggest that the washing temperature be under 15°C in any case. When the plates are not dried in alcohol containing glycerin, they also require a plasticizing bath of about 5 % glycerin in water prior to drying. To restrain the swelling and to clear the emulsion about 0.5 % of acetic acid is sometimes added to the wash water in the final stages. Swelling is also sometimes restrained by adding sodium sulfate to the wash water.

5.8 Drying

Early methods of emulsion drying involved the use of closed drying spaces in which the relative humidity of the air was controlled by a saturated solution of a suitable salt, or by a glycerin solution (Section 4.6). More rapid drying at the edge of the pellicle was inhibited by surrounding each pellicle with a guard ring of wet emulsion or gelatin.

To control the drying rate, dryers were developed such as one described by Rechenmann (R 56). This was made for drying freshly poured emulsion, and also keeps out dust and light.

Air drying has now been almost completely superseded by alcohol or polyethylene glycol drying. Alcohol drying is less troublesome and the emulsion distortion is usually less than when it is air dried. The growth of microorganisms in the emulsion is also inhibited. The drying is usually in several stages. The first stage may consist of immersion in an alcohol-water solution at the same temperature as the wash water. Such an immersion should be for about the penetration time (2.5 hr for 600 μ emulsion). Subsequently the emulsion may be immersed in other alcohol solutions of progressively higher temperatures and concentrations. The last can be in a solution of 90 % alcohol, 5 % glycerin, and 5 % water. The time in this stage should be longer, perhaps 8 hr. The alcohol usually has reached room temperature before the plates are removed. The writer uses three baths, each containing 5 % glycerin. The first contains 50 % alcohol, and the second 75 %. Each solution is admitted in turn to the main processing tank in which are racks containing the plates.

After they are removed from the alcohol, the plates are permitted to remain at 55 % relative humidity until the thickness of the emulsion layer has stabilized. Then the thickness of each is measured with the microscope. Before processing, the thicknesses of some or all of the pellicles are determined. The ratios of thicknesses before and after processing are then calculated. The ratio should be the same for all pellicles. This common shrinkage factor can then be used to calculate the original thickness of each pellicle. The information should be recorded. Whatever the subsequent humidity conditions, the original thickness will then be available for evaluating the shrinkage factor applicable at that time.

The glycerin content of the last drying solution affects the shrinkage factor; the more glycerin, the smaller the shrinkage factor. Glycerin is not suitable for producing a unit shrinkage factor, however, because the emulsion becomes too soft and hygroscopic.

Some laboratories use more stages of alcohol drying, starting with 10 % or 20 % concentrations. Information is not at hand to indicate whether or not the extra effort is justified.*

Other laboratories use a solution of polyethylene glycol (Carbowax) for drying. At Laussanne (D 60.2) the Carbowax solutions used are 30 %, 60 %, and 100 % in three baths.

* H. De Carvalho has expressed the opinion that a starting concentration of 50 % is too high.

At CERN, industrial alcohol containing 2 % acetone is used in four stages of 20 %, 40 %, 60 %, and 80 %, each also containing 50 ml per liter of glycerin.

After drying or in the process of drying it is possible to impregnate the emulsion with a filler that gives it a shrinkage factor near unity. Sometimes this may be a useful procedure. For the purpose an alcohol solution of pine resin (colophony) has often been used. (As mentioned above glycerin is a possibility, but if it is used the emulsion surface should be coated with a material impervious to water. The index of refraction of glycerin is not very suitable either.)

F. C. Gilbert (G 58) studied impregnation of emulsion with alcohol solutions of resin. He was always able to approximate a shrinkage factor of unity by varying the concentration of the solution and the time of soaking. About 35 gm of resin per 100 ml of alcohol produced a shrinkage factor of unity. Kodak acid fixer was used because neutral hypo seemed to inhibit the penetration of the resin solution.

The lightest, clearest type of Spanish colophony is recommended by Catala and Casanova (CC 58). This has an index of refraction of 1.5192 $\pm$ 0.0096—very close to that of processed emulsion gel and the immersion oil normally used. They found that the emulsion must be washed in distilled water or demineralized water for good results. Then with a 33-34 % solution of resin in ethyl alcohol they obtained a shrinkage factor of unity. The time of immersion is not critical except that it must not be too short or the ultimate quantity of resin will not be absorbed. After impregnation the plate thickness varies virtually not at all with varying ambient humidity. Another advantage is that the background is reduced in such plates because, the depth of focus being finite, the number of grains in focus is reduced in inverse proportion to the swelling.

It has not been established that by such methods of impregnation the resin is uniformly distributed through the layer of emulsion, so that distortions of vertical dimensions may remain. The results also do not always seem to be reproducible from one emulsion to another.

Catala and Casanova gave references to other impregnation work that has been done. Certain synthetic epoxy resins have recently been considered for this purpose (D 60.3). The use of tert-butyl alcohol both for drying and plasticizing was also suggested.

After drying if a deposit of silver or an excess of silver grains remains on the pellicle surface, ethyl alcohol on soft paper or chamois skin can be used to rub it off. This should be done gently. It must not be done at all if ranges of grazing-incident particle tracks are to be measured. It is easily possible to rub off 10 or 100 μ of track.

5.9 Processing Unmounted Pellicles

A number of laboratories carry out the developing and fixing of emulsion pellicles before mounting them. Demers (DD 58.1) describes how he handles pellicles of 700 μ thickness; Rosenfeld *et al.* (R 55) give details of processing pellicles up to 900 μ in thickness; and Yagoda (Y 57) has described procedures for processing layers of emulsion up to 2 mm in thickness—too thick to be studied with a high-power refracting microscope objective.

The advantages cited for free processing of pellicles are:

(*a*) More uniform development is obtained and time is saved, because the solutions can penetrate the pellicles from both sides.

(*b*) The bending of tracks by the unequal swelling of the emulsion resulting from the attachment of only one surface to the glass is avoided.

(*c*) Tracks can be viewed from either surface of the pellicle.

(*d*) Thicker pellicles can be successfully processed off glass.

Some of the disadvantages are:

(*a*) The free pellicles seldom return to their original dimensions after drying, and ranges, angles, and other measured quantities cannot be measured as well as in premounted pellicles. The use of a contact-printed grid on the free pellicle, however, helps greatly when it is to be used for quantitative measurements or when tracks are to be traced from one pellicle into another.

(*b*) Processing of free pellicles is complicated by the possibility that they may stick together or to surfaces with which they come in contact.

(*c*) A stack of several hundred large pellicles offers a very considerable task in processing, as the handling of free pellicles requires much more personal attention than those that are more ruggedly mounted.

(*d*) A pellicle seldom lies in a plane, and in order to scan it conveniently it is often found best to cement it to glass with rubber cement or otherwise, thus losing some of the advantages cited above.

NIKFI emulsions to be processed off the glass are subjected to a tanning agent which improves the strength of the pellicles.

Marguin (M 57) has developed means for processing unmounted pellicles of 1-mm thickness while holding them in frames of Plexiglas. On both sides of each pellicle is very fine stainless steel mesh (40 μ wires, 100 wires per centimeter) which permits solutions to reach the pellicle. During processing the pellicle is permitted to expand to triple its normal volume.

The method employed by Demers (DD 58.1) to process his pellicles off the glass is completely automatic, the cycle being controlled by clockwork. The pellicles, which are suspended in the baths, generally increase in lateral dimensions by about 7 % in processing.

Figure 5.9.1 is the photograph of a typical 5 × 7 inch × 1000 μ unmounted pellicle from a stack of the Enrico Fermi Institute.

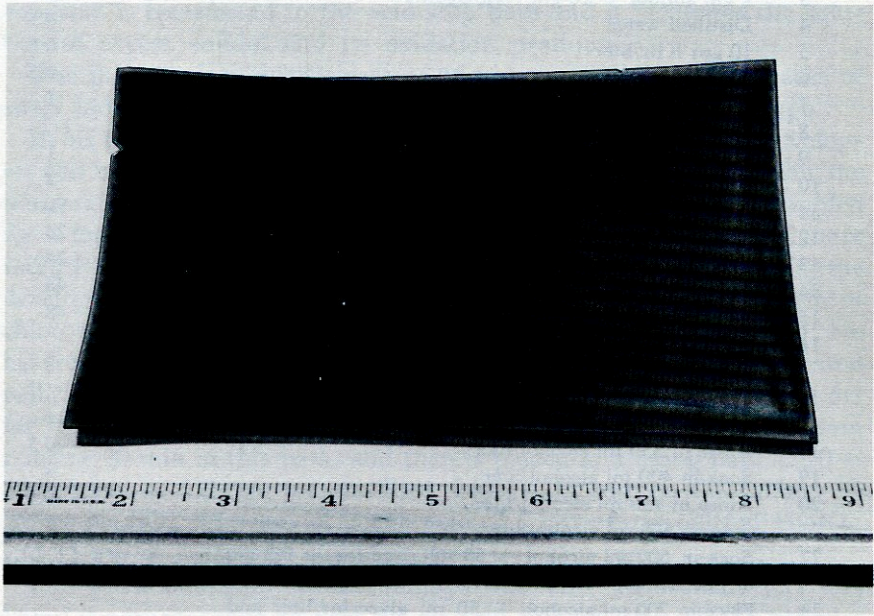

FIG. 5.9.1. Photograph of a 1000 μ pellicle processed while unsupported (IDLRL).

H. Yagoda (Y 57.1), probably more than any other individual, has developed the technique of processing thick layers of emulsion without support. He has been able to achieve uniform development of thicknesses of 1500 μ or more by an elaborate isothermal process, which he has outlined in the 27 steps of Table 5.9.1.

A temperature of 5°C is employed throughout stages 1 to 22. The changes in pH from stage to stage are brought about in many steps to control the swelling and avoid emulsion distortion. By the end of stage 12 the emulsion has experienced its maximum distortion—about 20 % in lateral dimensions and four or five-fold in thickness. By the termination of stage 23 the lateral dimensions are found to be only 3 ± 1 % over their original values. The trays in which the pellicles are processed

TABLE 5.9.1
DEVELOPMENT OF 1500 μ THICK PELLICLES[a]

Stage	Operation	pH	Duration (hr)
1	Presoak in freshly distilled water	7	2
2	3 gm amidol + 6 gm Na_2SO_3 + 2 gm $NaHSO_3$/liter	6.5	3
3	2 gm amidol + 16 gm Na_2SO_3/liter	7.5	2
4	Distilled water	7	0.5
5	10 gm KBr/liter	7	0.5
6	30 gm ammonium acetate + 10 gm citric acid/liter	5	0.5
7	5 ml glacial acetic acid/liter	4	1
8	Water wash and surface cleaning		1–2
9	100 gm $Na_2S_2O_3$/liter	~7	1
10	200 gm $Na_2S_2O_3$/liter	~7	2
11	300 gm $Na_2S_2O_3$ + 3 gm Na_2SO_3/liter	~9	96
12	200 gm $Na_2S_2O_3$ + 50 gm $(NH_4)_2SO_4$/liter	~8	24
13	100 gm $Na_2S_2O_3$ + 100 gm $(NH_4)_2SO_4$/liter	~8	24
14	100 gm $(NH_4)_2SO_4$/liter	7	48
15	50 gm $(NH_4)_2SO_4$/liter	7	48
16	10 gm thiourea + 10 gm citric acid + 30 gm ammonium acetate/liter	5	8
17	5 gm thiourea + 5 gm citric acid + 15 gm ammonium acetate + 300 ml of absolute ethyl alcohol/liter	5	16
18	Rinse in 350 ml alcohol/liter		~0.5
19	Soak in 400 ml alcohol/liter		3
20	Rinse in 450 ml alcohol/liter		0.5
21	Soak in 500 ml alcohol/liter		7
22	Soak in 600 ml alcohol + 50 ml glycerine + 0.2 gm of phenol/liter		16
23	Place in 700 ml alcohol + 50 ml glycerine/liter and warm to room temperature (25°C)		~3
24	Rinse in 750 ml alcohol/liter, drain surfaces on cellulose acetate sheeting, and mount on subbed glass		
25	Dry at 50 to 60% R.H.		72
26	Clean dry surface with cotton moistened with equal volumes of xylene and absolute ethyl alcohol		2
27	Dip plate in solution composed of 1 part of Duco cement and 2 volumes of acetone. Drain off excess, support glass plate horizontally on 3 thumbtacks, and cover with evaporating dish moistened with acetone		4

[a] Yagoda.

are lined with 2 or 3 mm of filter paper and the pellicles are also covered with sheets of filter paper at the end of stage 8 so that the ease of access of the solutions to the pellicle is the same from both sides. Great care

is exercised to eliminate bubbles which might be trapped in contact with a pellicle. Their presence while the pellicle is in the fixation bath causes a complete loss of track image in the areas covered by air bubbles. This is attributed by Yagoda to a solvent action of sodium thiosulfate in combination with oxygen on the developed silver.

The presoak time is restricted to 2 hr and it is stated that more than 4 hr appears to weaken the latent image.

The special buffer, stage 6, provides a transition in pH before the emulsion is transferred to the acid stop bath and it facilitates the solution of excess amidol and its oxidation products.

The thiourea introduced in stage 16 aids in the solution of traces of silver iodide which may not have been dissolved in alkaline hypo.

In his earlier experiments, Demers (D 58) used standard D 19 developer, and also D-8 diluted 2:1 or 1:1 at 20°C for 3 to 6 min. When the temperature was lowered he found that the development time doubled for a temperature reduction of 5.26°C. For very low fog he has more recently employed a formula in which chlorohydroquinone is the developing agent. For 1 liter of solution 30 gm of Na_2SO_3, 45 gm of chlorohydroquinone, 32.5 gm of KOH, and 8 gm of KBr are used. He has recommended to store the solution under nitrogen. Unmounted pellicles of 300 μ thickness are developed in this solution diluted 6:1 for 7.5 hr at 0°C. The stop bath, also at 0°C, is 1 % acetic. The emulsion remains 20 min in this bath, and then is placed in a fixing bath at 20°C constituted as follows: for one liter, 240 gm of sodium thiosulfate, 15 gm of Na_2SO_3, 13.5 gm of acetic acid, 7.5 gm of crystallized H_3BO_3, and 15 gm of potassium alum are used.

For thick emulsion layers Samoylovich and Tarasenkov (ST 62) found it advantageous to pass an electric current through the pellicles while they were developing. This apparently accelerates the diffusion of the developing agent.

They varied the developer composition, the electrode material, and the electrode spacing, as well as the temperature and time of development, to improve the uniformity of development. They found that complete development of layers up to 1200 μ in thickness could be achieved in 15 to 20 min.

5.10 Test of Development

One of the most useful tests of the quality of the development of an electron-sensitive emulsion is the minimum ionizing track test. It consists merely of comparing the ease and certainty with which minimum

ionizing particles can be traced through samples of emulsion which have been developed in different ways. Although a high blob density at minimum is an important factor in the ease of following a track, it is not the only consideration. The single grain and electron densities are very important, and in thick emulsions the track contrast deep in the emulsion may be the decisive factor.

The writer's research group recently conducted some tests on the hot stage development of 600 μ Ilford K.5 emulsion samples from a single batch. The formulas for Brussels and Bristol developer given above were used. In all cases the presoak was in distilled water at 5°C for 2 1/2 hr. The cold soak in the developer was also for 2 1/2 hr at 5°C. In each developer the hot stage was carried out with the emulsion immersed in a solution consisting of the developer diluted with an equal volume of distilled water. Temperatures were as follows: 22°C and 25°C. Times of hot-stage immersion were: 30 min, 50 min, and 70 min. All combinations of developer, temperature, and time were tried. In a certain sense it can be said that development is not critical, because all combinations tried developed tracks of minimum ionizing particles so that they were visible, but they were not equally easy to see.

The combination of Bristol developer, for 50 min at 25°C was actually chosen for use.

It may be desired to test another aspect of the development—namely the discrimination obtainable. Then a correct procedure is to compare the values of σ_g/g (see Chapter 9) obtainable from track segments of equal length.

Emulsion sensitivity and degree of development are conventionally given as blobs per 100 μ at the minimum of ionization. Development uniformity, however, is best measured by observing g, rather than the blob density, because as reported by Dilworth *et al.* (DOV 51) especially near saturation, tracks are sensitive to the amount of physical development which affects the grain size, but perhaps not the number of grains. Tests of development uniformity for nonminimum tracks are easily made on slightly inclined tracks of relativistic alpha particles which traverse the whole pellicle thickness with no appreciable change in velocity. Alternatively different tracks, known to have the same velocity, can be measured at various depths and in various plates.

A precaution in interpreting the measurements is necessary. When tracks are deep in the emulsion, contrast is lost and small gaps are not seen well. To be quite sure that a depth effect exists, it is wise to use a long-focus objective and measure the ionization of the same track both in the normal way and through the glass with the plate upside down on the microscope stage.

5.11 Track Intensification

Methods for intensifying photographic negatives were developed many years ago (M 54). Such techniques as replacing the silver by another metal or by adding more silver were applied to particle tracks by Powell *et al.* (POLC 46) with some success. Discrimination tends to be lost, however, in thus increasing the track visibility.

Lonchamp and Morgenthaler-Metz (LM 55) have enumerated conditions under which track reinforcement may be useful, and have studied extensively various methods of chemical intensification. They state that track intensification is capable of saving accidentally underdeveloped plates, and the surface layer of thick plates that were etched in processing.

Moreover, they believe that for particle discrimination this technique may be useful. One of the means used for discrimination is underdevelopment. They suggest combining underdevelopment with track intensification to make visible the partially developed grains. They also suggest development at a low temperature to attain uniformity in thick plates, with subsequent intensification of the tracks.

Lonchamp and Morgenthaler-Metz, after testing a number of other processes, recommend the following procedure: immerse the plate for 30 min in a bleaching solution of 3 % $HgCl_2$ and 3 % KBr. Then a blackening solution is prepared by adding 18 gm of Na_2SO_3, 34 gm of H_3BO_3, 8 ml of a 1 % solution of KBr, and 4.5 gm of amidol to a liter of distilled water. This solution is diluted by a factor of 10 for use. A dry underdeveloped plate is impregnated first with a 0.5 % solution of sodium carbonate for 24 to 48 hr, depending on the dryness. Then the plate to be reinforced is placed for 30 min in the intensifying solution. The intensification may be repeated. Figure 2.3.4 shows the effect of successive reinforcements of alpha-particle tracks. While the growth of the fog grains is conspicuous, a net gain in visibility may have been achieved.

5.12 Clearing Solutions

Often the processed plate is lacking in clarity. The cause may be tiny scattering centers of colloidal silver which produce a loss of contrast in the image, especially deep in the emulsion. A treatment that the writer has found satisfactory for this condition is the following: the emulsion is soaked in a solution of 1 part concentrated hydrochloric acid solution to 1000 parts of water for 1 hr at 5°C. The emulsion is then refixed in hypo of 10 % normal concentration, rediluted, and washed.

The process is repeated if necessary. This is a slight variation of a method used by Dilworth *et al.* (DOS 48) for progressive, controlled clearing of plates.

Often merely refixing alone for a short time, a process that has few hazards, is very effective in clearing the emulsion. A little NH_4Cl in the fixer has a marked clearing effect, but etching of the developed grains also takes place unless it is done carefully.

When the silver ion concentration is high, silver-hypo complexes are believed to form (BDVP 57). These are removed from the emulsion only with difficulty, but washing and refixing are usually effective. A clearing procedure recommended by Stiller, Shapiro, and O'Dell (SSO 54) is to soak the emulsion for 8 hr in a solution made as follows:

Dissolve 10 gm of thiourea, 10 gm of citric acid, and 30 gm of C.P. ammonium acetate in 1/2 liter of water at 40°-50°C. Filter and dilute to 1 liter. After soaking for 8 hr at 5°C, the emulsion is washed and dried as usual.

A more drastic treatment has been published by Herz (H 52.1). After fixing and washing, the emulsion is soaked in a 0.06 % solution of potassium ferricyanide. This is followed by a short, second fixation and washing.

In Section 5.5, a procedure was given that bleaches excess surface silver.

5.13 Test for Hypo

For determining whether or not washing is complete, the following instructions are given: lift plate from wash water and let it drip for a moment. Let the last few drops drain into about 10 ml of the following solution: potassium permanganate, 0.5 gm; NaOH, 1.0 gm; and distilled water, 1 liter. Dilute 20 to 1. The presence of hypo will cause the violet color to turn orange in about 30 sec. Higher concentrations cause it to turn yellow. The color change is most easily seen by looking down into a test tube containing the liquid. It is best to place the test tube in a rack with a white surface beneath the tube and to place adjacent to it a comparison solution containing no hypo.

5.14 Measurement of the Silver Ion Concentration

One of the simplest ways to determine the silver ion concentration is to observe the change in specific gravity of the fixing solution brought about by the solution of silver halide in it. Using a good hydrometer,

the specific gravity of the solution prior to use is determined at the temperature at which it is to be employed. Then as fixing proceeds, the temperature remaining constant, measurements of the density by means of the hydrometer can be made. A change of density of 0.0013 gm/ml corresponds to a change in silver concentration of 1 gm/liter at 10°C. The density changes 0.00036 gm/ml per degree centigrade in the specific gravity range, 1.15 to 1.17.

Of course the silver concentration also can be determined by standard methods of analytical chemistry, but this is slow. Dahl-Jensen (D 60.2) described electrical means for measuring and controlling the silver concentration in a hypo bath. The potential difference between a silver and calomel electrode (see Chapter 2) depends on the silver concentration and on the hypo concentration. This relationship is given graphically. When the silver concentration is about 5 gm/liter, for example, the change in potential is about 10 mv for a change of 1 gm of silver per liter. This value is almost independent of the hypo concentration. The potential difference is not affected by sodium bisulfite, sodium sulfite, sodium acetate, acetic acid, or gelatin in the hypo bath.

5.15 Recovery of Silver from Fixer

Recovery of the silver can be effected by electrolysis of the used fixing solution. If a difference of potential of 1.5 volts is maintained between a positive graphite electrode and a negative stainless-steel electrode in used fixing solution, silver will plate out on the stainless steel. It is important to keep the liquid in rapid motion with respect to the electrodes, and no pockets of stagnant liquid should be permitted. There is danger otherwise that the sodium thiosulfate will decompose.

If the voltage is kept fixed, the current will depend on the silver ion concentration. It is unwise to raise the voltage above 1.5. If the stainless steel is in the form of thin sheets, flexing the sheet is usually effective in causing the silver to flake off, thus simplifying its recovery.

It is also possible to recover the solution itself for reuse, but frequently the emulsion to be processed is so valuable that one does not care to compromise its quality by using any fixer but one of the highest purity.

After prolonged electroplating, the pH of the fixing solution changes and requires adjustment if the solution is to be reused.

In Fig. 5.15.1 is shown the appearance of a rotating stainless-steel electrode heavily encrusted with the silver that was recovered from a small emulsion stack.

The simplest method for recovering the silver is to precipitate it as

silver sulfide. One merely adds sodium or potassium sulfide to the bath. Hydrogen sulfide fumes are evolved, however, and this operation as well as electrolysis should be carried out in a hood. Silver can also be displaced from solution by such metals as zinc, iron, or copper.

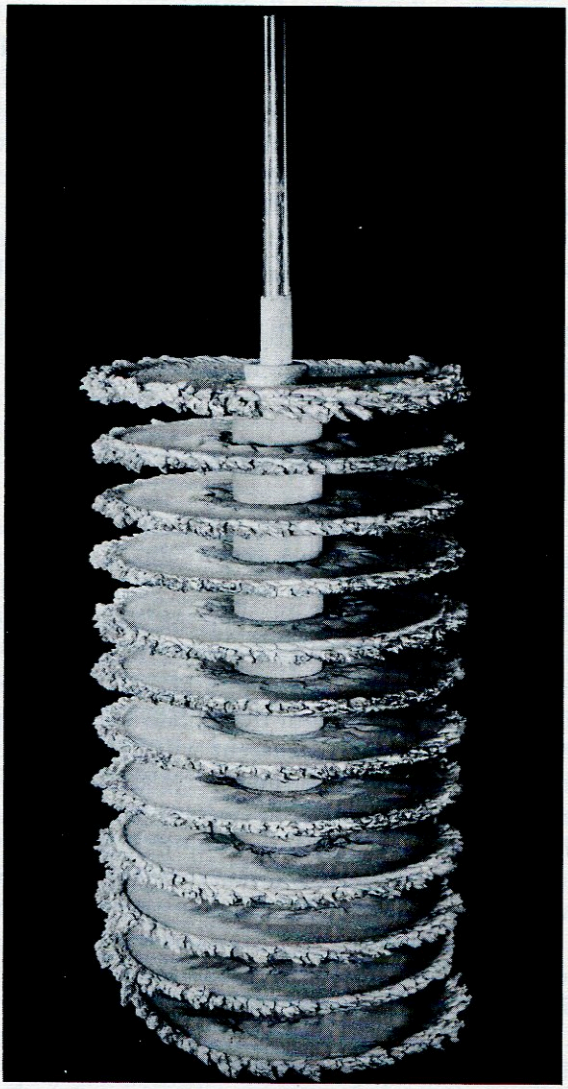

FIG. 5.15.1. Silver recovered from bath used to fix a small emulsion stack (IDLRL).

5.16 Discrimination between Heavy Ion Tracks by Controlled Development

In Chapter 2 the general principles that can be employed for discriminatory development have been discussed.

A particular problem of this sort is presented when one wishes to record the tracks of fission fragments while discriminating against alpha particles or even more highly ionizing particles. In Ilford K minus 2 or K minus 3 emulsion developed with a modification of a formula published by Stevens (S 51), the tracks of fission fragments and argon ions are visible while the tracks of alpha particles remain undeveloped, and even tracks of carbon and nitrogen ions are very weak. The developer is made up from 3 stock solutions: solution A′ consists of 15 gm of *p*-aminophenol hydrochloride, 2.5 gm of sodium bisulfite, and water to make 1 liter. Solution B is made up of 50 gm of tribasic crystalline sodium phosphate, 40 gm of sodium sulfite, 4 gm of potassium bromide, and water to make 1 liter. Solution C is made by adding 50 gm of dibasic crystalline sodium hydrogen phosphate, 40 gm of anhydrous sodium sulfite, and 4 gm of potassium bromide to water so as to make 1 liter of solution.

Varying the ratio of solutions B and C varies the pH and varying the quantity of solution A′ varies the concentration of developing agent. A preferred composition consists of 2 parts of solution A′, 50 parts of B, and 50 parts of C. Development is for 40 min at 70°F.

Studies have been made by Gegauff and Lonchamp (GL 58) of discriminatory development for such heavy ions as C, N, O, Ne, and A. The mean track widths were shown to increase with development time, but to depend little on whether the developer was ID-19 or an amidol solution.

CHAPTER 6

The Processed Emulsion

6.1 Fog and Foreign Matter in Emulsion

The development process, as mentioned in Chapter 2, is always a differential one; the grains containing latent images caused by an ionizing particle are chemically reduced more rapidly than the unaffected grains. All will be affected, however, under the prolonged action of a sufficiently active reducing agent. Random developed grains constituting "fog" are found in emulsion because there are always a few grains of the immense number present that develop as quickly as the track grains. Although they are often large, many of the fog grains do not appear to differ much in size or in any other visible way from the track grains. Apparently there is some way in which a grain can exist in a condition of developability without having been traversed by an ionizing particle.

The character of the defect that makes an unexposed grain developable is evidently not a latent image of the same type as that produced by an ionizing particle, because the number of fog grains that develop increases linearly with the time of development (D 58) for perhaps twice the period required for a track to attain its maximum grain count. If the time of development is extended still more, the density of fog starts to rise more rapidly than linearly with the time. The discrimination between fog and track grains also varies with the type of developer.

When the emulsion sensitivity is too low for tracks of weakly ionizing particles to be recognized, the occasional grains produced by passage of such radiations through the emulsion may have the appearance of true fog. There is always some tritium present, and in a sensitive emulsion the low energy electrons from it will produce some single-grain tracks indistinguishable from fog.

Fog produced by visible light is limited (except in very fine-grain translucent emulsions) to the surface, only a thin layer of which is penetrated by the light. Accidental exposure of a plate to light may not destroy its usefulness as the surface layer can be scrubbed off with alcohol. Often large blocks of stacked emulsion are left unprotected from the light, because only superficial layers at the edges of the pellicles are affected when the clamping members are opaque.

Yellowish staining of the gelatin is usually caused by oxidation products of the developing agent, and is especially likely to occur if the developer is old, if the development is prolonged, or if a stop bath is

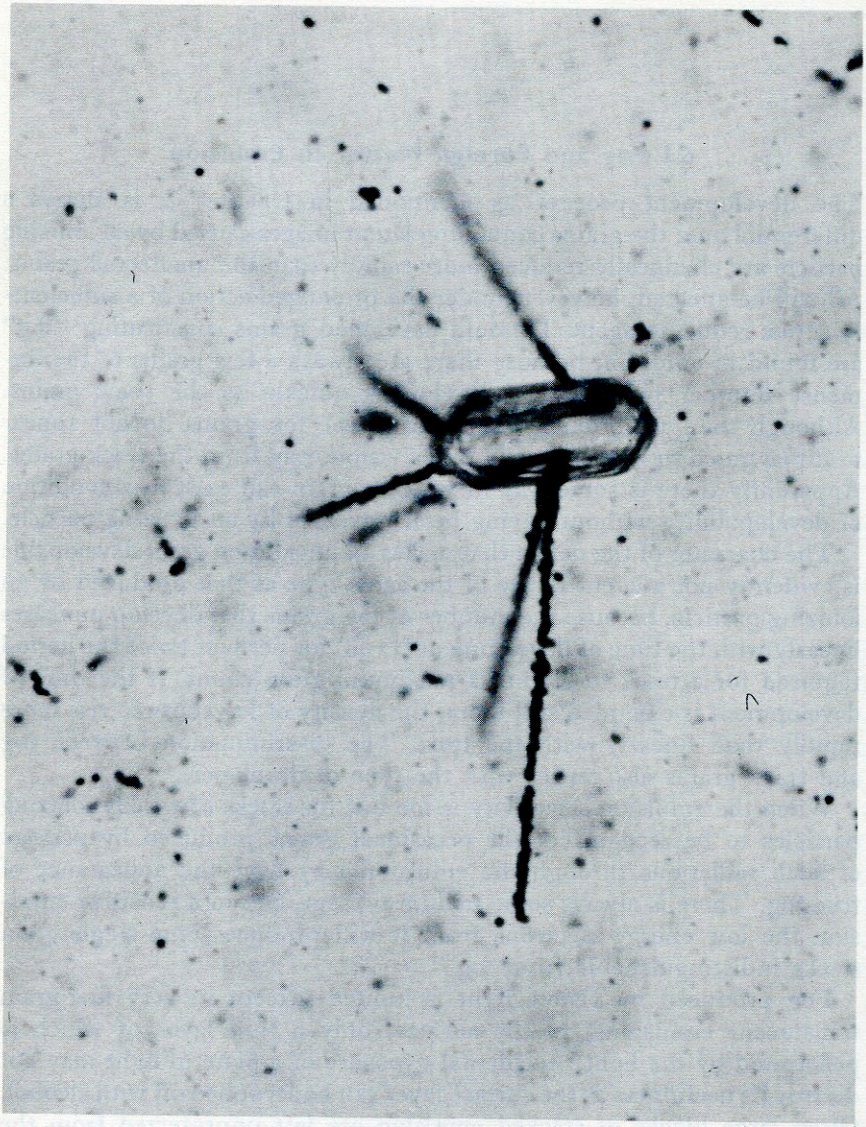

FIG. 6.1.1. An occlusion found in Ilford G.5 emulsion. It is believed to be a zircon crystal. The alpha-particle range is the same as that of thorium C′. (Courtesy H. Yagoda.)

not used after development. Prolonged immersion in a stop bath can produce a reaction in which a combination of developer, acetic acid, and oxygen stains the gelatin pink and dissolves the developed silver. Some emulsions are more prone to staining than others. Ilford's G.5 emulsion stains less readily than their C.2 emulsion.

There may also be seen in a processed plate black lumps of reduced silver that can be attributed to imperfect conditions of manufacture. They are generally a micron or more in diameter and may consist of several crystals close together. The individual crystals do not develop independently, and each therefore probably is not separately contained in an envelope of gelatin. These are called "agglomerates."

Another type of fog is generally attributed to colloidal silver. It consists of a very large number of grains so tiny as to be barely visible with the highest resolution. This type of fog has been observed to increase as the sulfite content of the developer is increased. If serious, it can easily be removed by a clearing solution as described in Section 5.12.

In addition to the normal components of emulsion, foreign matter may be introduced inadvertently in manufacture. This often consists of dust in which particles of almost any sort can be found. Occasional microorganisms and vegetable fibers are also present. While this sort of "trash" is not a serious problem with emulsions now available, instances where foreign matter destroyed the possibility of analyzing an important event are known. Occasionally, also, transparent, insensitive masses of an unknown nature have been found in the emulsion (see Fig. 6.1.1).

If the pH of the fixer is too low sulfur may be precipitated, and if the emulsion is not washed well, sodium thiosulfate crystals may be found in the processed emulsion. If incompletely fixed, silver halide may be found in the gelatin. Radioactive atoms may cause alpha-particle stars, and radioactive dust particles may produce characteristic structures in the emulsion (Y 49, PFP 59, D 58) (see Fig. 4.1.2).

Occasionally a horizontal plane, that appears to be the interface between two bodies of emulsion, is found. In it an increased density of grains is usual. It has been assumed that sometimes thick emulsion is poured in stages, and the interface may be the boundary between layers poured at different times.

6.2 Track Visibility

The presence in the gelatin of anything (including too many tracks) that scatters and absorbs light is deleterious to the emulsion quality. When scattering material is present, tracks deep in the emulsion lose

contrast, are difficult to see, and require large corrections in photometry. When there is much light scattering, red illumination is recommended. If a large amount of light is absorbed in the emulsion a strong source is required. Then the emulsion may be damaged by the heating that occurs where the intense light beam is focused on it by the condensing lens. It may be necessary to use heat-absorbing glass or a water filter in the light beam.

Fog is an especially serious problem when one is making a study for which it is essential to see tracks near the minimum of ionization. In modern studies it is becoming more and more common to trace minimum tracks through many pellicles. The presence of a near-minimum decay track distinguishes a positive muon from a zero-prong negative pion coming to rest in emulsion. It also distinguishes positive K mesons or charged hyperons, in some decay modes, from protons. Failure to see the track of the weakly ionizing particle in these cases is serious, and as the fog density increases the probability of seeing a near-minimum track falls rapidly.

It sometimes happens that near the surface the fog increases. This is particularly observable if too little restrainer was used in developing a thick emulsion layer. A fog gradient can be a sensitive indicator of a development gradient. At the very surface a layer of fog grains often is found. The surface layer is usually most strongly developed. It may be somewhat light-struck, also, and this layer of the emulsion may have been affected by external chemical agents. Besides, at the surface the emulsion is subject to abrasion. Another contributing effect is the luminescence that is produced when pellicles that adhere to each other are pulled apart.

Surface graininess tends to cause an uncertainty in the location of the point where a track enters or leaves a pellicle, particularly if it enters at grazing incidence. This effect may be important in range measurements (Chapter 10). Surface graininess is useful in a certain respect; one would have no way to focus on a surface devoid of grains or other optical features.

For its recognition as the path of a particle, the grain density in a particle track must be larger when the density of background grains or fog goes up. The grain density required for a track to be visible appears to rise as the square root of the in-focus areal grain density of the fog. This has been called the "law of Selwyn-Coates" by Demers (D 58).

Well-processed Ilford G.5 emulsion has been observed to contain about 5×10^9 fog grains per cubic centimeter, and tracks with a grain density of less than about 10 grains per 100 μ are generally not recogniz-

able in this amount of fog. In a plane containing a density, ρ, of random points, the mean distance from a point to its nearest neighbor is $1/(2\rho^{1/2})$. If an objective lens is used with a depth field such that ρ grains per unit area are in focus, then a flat track with a mean grain density of about $2\rho^{1/2}$ may be barely discernible. (A complete three-dimensional study of the problem of track visibility is yet to be made, and it must be admitted that most of the present information on this subject is qualitative.)

If the grain size is reduced, and the depth of field correspondingly lowered by using a larger numerical aperture, a higher ratio of fog density to track density can be tolerated because a smaller proportion of the fog grains will be in focus, whereas the grains in a straight, flat track can all remain in focus simultaneously. Unfortunately the field of view then is restricted. The AgBr concentration also affects the track visibility, and as the concentration is varied a point of maximum track visibility is found. This point is reached when the silver halide content is about 3.5 gm/ml, according to Demers (D 58).

The track visibility is reduced if the track is inclined, and especially if it scatters greatly. On the other hand, if the track is flat and straight, its visibility would be enhanced by swelling the emulsion, because then the track grains all will remain in focus, but the areal density of fog grains in focus will be reduced by the swelling. Berriman (B 51.1) mentions the sudden rise of visibility of a track when the ionization increases to a point where double grains begin to appear along its trajectory, doublets having direction and being rare in fog.

Wide-field oculars are advantageous for improving the visibility of tracks. For the same depth of field, a longer visible track segment gives more information about the track.

The single-grain fog background has one deleterious effect not mentioned heretofore. It reduces the accuracy of grain density measurements because the background, the effect of which is not easy to evaluate, must be subtracted from the observed grain density.

The presence of other tracks in excess of a certain density starts to interfere seriously with the viewing of any particular track, especially if it is deep in the emulsion. In some types of research one may wish to maintain the visibility of proton or alpha-particle tracks in the presence of large numbers of tracks made by electrons from gamma-ray irradiation of the emulsion. It is possible to utilize discriminatory development, as described in Chapter 2, to retain the visibility of the heavy particle tracks in spite of the ionization produced by the electrons.

Good track visibility is extremely dependent on the microscope equipment at hand and on its correct adjustment.

6.3 The Optical Characteristics of Processed Emulsion

After fixing, the matrix material of the emulsion, except for its soluble components, remains. It may also contain some added glycerin, and its water content changes with the ambient relative humidity. For dry gelatin the index of refraction is about 1.53. The index of processed emulsion containing the usual amount of water and glycerin has been found to be in the neighborhood of 1.51-1.52. The immersion oil generally used has an index of 1.515, so the optical match here is very good, and often depth measurements are made in emulsion by measuring the amount of displacement of an oil immersion objective while making no correction for differences of refractive indices.

Sometimes newly processed plates that appear dry may have a milky translucent appearance. This has been noticed especially in plates that are dried with alcohol. The effect is attributed to the incomplete collapse of the voids formerly occupied by silver halide crystals. These holes are, in order of magnitude, equal in size to a wavelength of light, and if filled with air each is an effective scattering center for light. If the plates are stored for a few hours or days they become transparent, presumably because the voids have collapsed.

As discussed below (Section 6.5), gelatin under stress becomes doubly refracting, and the strain pattern in an emulsion sheet can be examined by the use of a polarimeter of the sort used in the study of stressed models of engineering structures.

If dried emulsion is cloudy and difficult to see through, often refixing has a remarkable clarifying effect on it. Refixing is apparently more effective than increasing the time of the original fixation. The clearing procedures described in Chapter 5, Section 5.12, also may be found useful.

6.4 Shrinkage and Stretching of Emulsion

After processing emulsion it will occupy less volume than before unless some material has been added to replace the silver halide dissolved by the fixer. For glass-mounted plates the most conspicuous evidence of this effect is a simple reduction of the thickness of the emulsion layer.

Both gelatin and glycerin are hygroscopic so that the actual equilibrium thickness and index of refraction of the processed as well as the unprocessed emulsion depends on the ambient humidity. The thickness of the layer of emulsion at the time of exposure divided by its thickness at the time of scanning is called the *shrinkage factor*. It is important to know the precise original thickness of the emulsion for any quantitative

measurements of track densities, ranges, and angles in the emulsion. Because the index of refraction of the emulsion may not be the same as the oil used for an immersion objective (and certainly will not be the same as air when an air objective is used), depth measurements in emulsion, even using a microscope with a linear, accurately calibrated focusing motion, may not be correct. The most satisfactory procedure is to observe the apparent thickness of the processed emulsion in whatever units the z-motion is calibrated. This is then divided into the known original thickness of the emulsion at the point in question to obtain an effective shrinkage factor. This must be done at the time a measurement requiring knowledge of the shrinkage factor is made, because the emulsion changes its thickness as the humidity changes. The shrinkage factor also may be different at different depths in the emulsion if equilibrium with respect to the water content of the atmosphere has not been attained, or if the glycerin is not uniformly distributed throughout the emulsion. It has been reported that the shrinkage factor may vary with depth in the emulsion for other reasons in addition (GM 57). It has also been stated that the shrinkage factor may vary seriously from place to place on a given plate, but the writer has not observed this effect.

Normal processed emulsion changes its thickness with the ambient relative humidity in a way that is given roughly by:

$$\frac{\Delta t}{t} = \frac{r^2}{30{,}000}$$

Here t is the nominal thickness, Δt is the increase from the dry thickness, and r is the relative humidity in per cent. This relationship is based on measurements near a relative humidity of 60 %. It is desirable that the room where the plates are stored and scanned be maintained at a relative humidity between 50 and 60 %. The shrinkage will then vary little from time to time, and transient humidity changes will not be the cause of variations with depth of the shrinkage factor.

Vigneron *et al.* (VGC 55) have found in plates dried with alcohol or acetone, that when apparently dry, the plates may not have contracted the expected amount. It is supposed that the emulsion then has a somewhat open structure. This effect is not believed to be permanent, however, as the gelatin tends to assume normality in a few days.

To know whether or not the emulsion is in equilibrium with the atmospheric water, repeated weighings will reveal loss or gain of water. Another method is to place a drop of immersion oil on a smooth surface of the emulsion. After an hour or more the oil may be carefully wiped off and the surface of the emulsion examined under grazing-incidence light. If a bump remains on the emulsion, it is not evidence that the oil

has penetrated the emulsion, but rather that the emulsion around the oil drop lost water during the time the drop was present. Conversely, if a depression appears, the emulsion has gained water from the atmosphere. Since time is required for water to diffuse through the emulsion to or away from the glass, the shrinkage factor and index of refraction will vary with depth. A way to minimize this effect is to coat the entire emulsion surface with immersion oil. The chief objections to this procedure is that it is "messy" and the oil catches dust. Sometimes lacquers or plastic coatings are put on emulsion to protect it and prevent water exchange with the air. Unfortunately, 600 μ emulsion, which is rather standard, is the limiting thickness that can be scanned with high-power immersion objective lenses because of their small working distance. Only a very thin coating is at all acceptable in this case, so that it is seldom waterproof. In addition, it may have to be removed if it is later decided to subject the plates to further processing. The coating employed, of course, must be unaffected by oil, water, and the moderate heat of the intense light passing through it. It must be flexible and tough so as not to chip off, and it should dissolve easily in a solvent that does not adversely affect the emulsion. The coating must, of course, be smooth and transparent. While no really satisfactory coating is known to the writer, thinned Duco cement has been used for this purpose, in spite of the fact that it is somewhat brittle and not waterproof. Krylon plastic spray and Eronel Spraypeel No. 725 $\times$ 1 are additional products that have been used for this purpose. Plates have also been embedded in a block of polystyrene, but then only long-working distance objectives are useful.

As emulsion contracts on drying, it may produce striking evidence of the magnitude of the stresses developed. For example, a surface layer of glass may be torn loose from the glass plate with the emulsion. (This is not likely to happen if the emulsion pellicle does not extend to the edge of the glass.) The glass plate on which a pellicle is mounted may bend and even break when the humidity is lowered. These effects are greatly reduced by introducing glycerin into the processed emulsion. It tends to keep the pellicle moist and pliable.

When a pellicle is not mounted on glass, its length and width as well as its thickness may alter in processing. By putting unmounted pellicles through a sequence of baths, Yagoda (Y 57) and Demers (D 58) have been able nearly to restore them to their original areas.

If the resultant alteration of the emulsion is merely to convert an elementary cube into a rectangular parallelepiped, simple scaling of the dimensions along the three axes will suffice to correct for the geometrical alterations. Angles will be changed, but straight lines remain straight lines.

6.5 Shears and Nonlinear Distortions

In order to make precise and meaningful measurements in emulsion, its behavior should be thoroughly understood. Then possible sources of error can be anticipated and circumvented, or, at least, intelligent estimates of errors can be made. Failure to take account of known behavior characteristics of emulsion has been the source of many serious errors in emulsion experiments. Among the most spectacular have been some measurements affected by emulsion distortion.

In addition to simple alterations of the three linear dimensions of an emulsion sheet by constant factors, usually more complex changes of geometry occur between exposure and the time when processing is complete. If the emulsion is supported on glass throughout the operations, these distortions generally are less severe than the distortion of unsupported emulsion. Those that are commonly seen have several causes: (*a*) Release of tensions existing in the emulsion at the time of exposure. [In processing such tensions will relax, so that even if no new distortions are introduced, the release of the old ones will have the same effect as the introduction of new distortions. It has been found (SSO 54) that stresses are released during the eradication process (Section 4.2) with a noticeable reduction in multiple-scattering error. Possibly some kind of annealing process prior to exposure should be adopted for any emulsion in which distortions must be kept small.] (*b*) Soft emulsion remaining for a period with its surface not level. (*c*) Maltreatment, such as touching the soft emulsion with the fingertip during fixing or washing. (*d*) Uneven drying.

An unmounted pellicle, especially one of large area, may suffer distortions from mechanical stresses. It may be bent, or it may be stretched if, for example, it is pulled when it adheres to another pellicle. These experiences introduce irreversible strains into the pellicle. As shown in Chapter 3, emulsion is not a simple elastic material, and does not obey Hooke's law. Some of the most serious distortions a pellicle suffers may be received in the mounting operation. In order to cause the pellicle to adhere well to the glass, it is sometimes rolled through a wringer or otherwise caused to experience large local forces and shearing stresses. Such outrageous but sometimes necessary treatment of the pellicle introduces distortions so complex as to be largely unanalyzable.

An effect of drying the emulsion, especially when done without alcohol, is often to produce surface stresses in it. The distortion from this cause is reduced by slow drying and by the use of guard rings (DOV 51). These may be emulsion or some other moist material placed around the plate to eliminate the humidity gradient at the edge of the plate. Drying

produces a type of distortion that is present at the edge in most plates (see Section 6.7).

Emulsion that is drying contracts and pulls on adjacent areas. If these are not equally as dry, or if such an area is adjacent to an edge, the unbalanced tension will cause a displacement. Near the center of a plate such tensions are generally in equilibrium with each other and there is no preferred direction of strain. Near the edges, however, the strain vector is perpendicular to the edge, and the gelatin will not be isotropic. Polarized light passing through a sheet of emulsion can be used to exhibit the areas of strain. In Fig. 6.5.1 is shown a sheet of emulsion that was dried without plasticizer. The areas of strain are clearly seen.

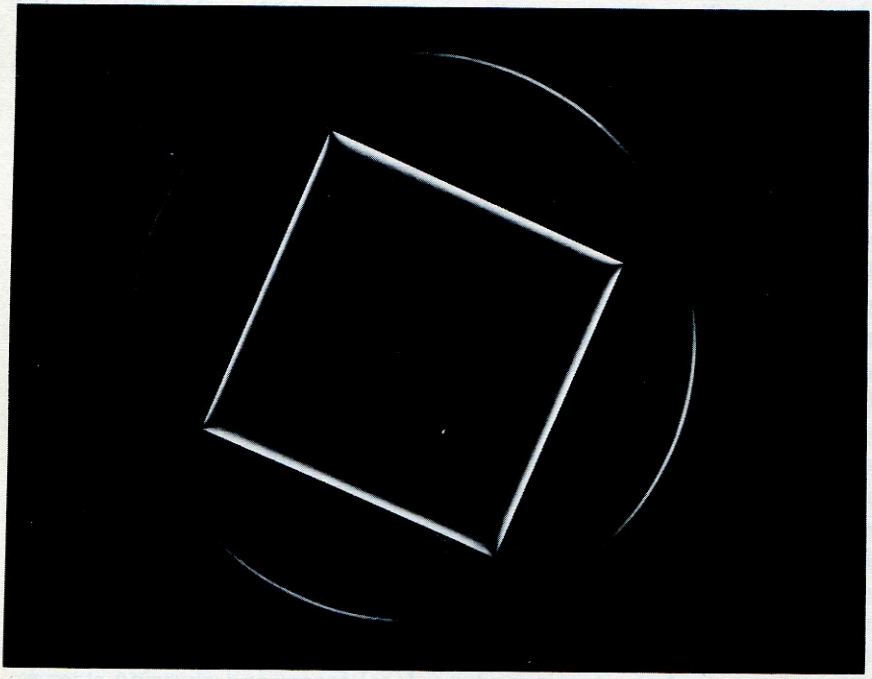

FIG. 6.5.1. Strain near the edges of an emulsion pellicle revealed by the double refractivity of the strained gelatin. (Photograph by A. Oliver.)

Distortions may be introduced if there is any large-scale inhomogeneity of the emulsion. Density gradients can be caused by imperfect mixing or by silver halide sedimentation, and the shrinkage factor increases with increasing emulsion density. In justice to the manufacturers, it

must be said that emulsion heterogeneity seriously in excess of inevitable statistical fluctuations is seldom encountered.

Violent microscopic distortions always occur when the silver halide crystals are dissolved in fixing. Voids are left which are filled by the collapse of the gelatin as the emulsion dries.

Experiments of J. P. Dentan (D 58.2) indicate that the pH of the developer when it is below 7.5 does not influence the distortion. When the pH goes above 7.5 the distortion rapidly increases. He also found no effect of the temperature on the distortion below 15°C, but, when the temperature went above this value, the distortion rose rapidly. Presumably these temperature and pH effects are indirect, the direct cause of the distortions being the emulsion swelling.

The swelling of the pellicle while in the fixing and washing stages is a primary cause of distortion, and is one reason for keeping the swelling within limits in these phases of the processing. When the pellicle swells, it tends to do so in all directions, but one surface of a mounted pellicle is constrained to retain its dimensions. The other tends to swell and the shearing stresses introduced may well lead to irreversible distortions of the pellicles. At this stage the soft emulsion is also exceedingly vulnerable to damage from any other kind of mechanical stress to which it may be subjected. Sudden changes of temperature or pH are thought to be harmful by Bonetti, Dilworth, and Occhialini (BDO 51). Too high temperatures and/or alkalinity in the warm stage of development (as observed by Dentan), sudden changes in salt concentrations, too rapid or vorticose motion of liquid in contact with the emulsion surface, or too high temperatures in fixing or washing are all possible causes of distortions according to Bonetti *et al.*, who have studied the problem in some detail. If emulsion is dried in air rather than alcohol, they warn against drying too quickly, at too high a temperature, and under uneven conditions of temperature or humidity. If the emulsion is not level when it is being dried, it will be subject to a shearing stress produced by gravity that increases in proportion to the distance from the emulsion surface. This leads to a standard "C" type of distortion.

A special and often serious type of distortion is that produced when blisters form between a pellicle and the glass. These tend to swell by osmotic pressure in the dilution and washing stages of processing. Some advocate doing nothing to a blister. It collapses on drying, but the gelatin will generally have stretched irreversibly, and a ridge will remain at the edge of the blister site. On the other hand, if the fluid is withdrawn from the blister with a hypodermic needle before it grows large, no great distortion occurs except at the puncture where the structure of the emulsion is destroyed.

Another cause of distortion and even destruction of the emulsion is the growth of microorganisms. Before fixing there is little danger of this happening, as the silver bromide protects the gelatin, but the processed gelatin may become a culture medium if it is kept moist and warm and is not protected by a disinfectant.

Although many possible causes of distortions have been put forward, their relative importance is not well known. It is here that a type of irrationality may have been introduced into emulsion processing. It has not always been proven that some of the more tedious operations said to be required to reduce distortion are justified. Sometimes any possible reduction is overwhelmed by other uncontrolled sources of distortion.

6.6 The Measurement of Distortion

If a beam of particles, sufficiently high in energy so that the scattering is negligible, is caused to pass through an emulsion layer at a known angle, the distortion of the tracks can be measured provided that the original thickness of the emulsion is also known. In order to determine accurately the dip angle of the particles, Apostolakis and Major (AM 57) put emulsion on both surfaces of a thin piece of glass. The entrance and exit points on the glass surfaces cannot move during processing if there is a good bond between emulsion and glass, so, knowing the thickness of the glass, the original angle of each track can be determined, and its path through the unprocessed emulsion layers established.

The original thickness of the emulsion was determined by measuring projected track lengths of particles traversing the emulsion layer. If the angle of dip measured in the glass is δ_0, and the projected track length is R_p, the original emulsion thickness, T, was $R_p \tan \delta_0$. After processing, using an oil immersion objective, the thickness, $t = R_p \tan \delta$, can be determined by focusing successively on the top and bottom surfaces of the emulsion, and measuring the distance the objective is moved. If the immersion oil has the same refractive index as the gelatin the distance moved is the thickness, t, of the gelatin layer. The ratio $T/t = S$ is the measured shrinkage factor. These results are only approximate because R_p itself may be seriously affected by the distortion, especially when steep tracks are measured. The dip angle should be kept small for this measurement or the emulsion thickness before and after processing should be determined by other means.

Knowing the emulsion thickness and the original dip angle, the original x and y coordinates of the particle trajectory will be known for each value of the vertical coordinate, z. The actual x and y coordinates

can be measured to observe the displacement of the track. Since the z coordinate is affected by the shrinkage, and the x and y coordinates are affected by shear distortions, with each point of the original emulsion one can associate a three-component displacement vector. When the shrinkage in z is corrected in the usual way, the remaining distortion is attributed largely to the lateral displacements in the x and y directions. Apostolakis and Major found that such a displacement, ϵ, of a point in the emulsion could be expressed by:

$$\epsilon = \mathbf{K}_1 \left(\frac{z_0}{T}\right) + \mathbf{K}_2 \left(\frac{z_0}{T}\right)^2$$

Here T is the thickness of the unprocessed emulsion, and z_0 is the distance from the displaced point to the glass in the unprocessed plate. $\mathbf{K}_1$ and $\mathbf{K}_2$ are in general vectors having different directions, but they found them to satisfy the relation $\mathbf{K}_1 = -2\,\mathbf{K}_2$ with remarkably little dispersion.

Barkas, Smith, and Birnbaum (BSB 55) measured the lateral position of steep tracks in nuclear-track plates at points equally spaced in depth in the emulsion. It was found that the second differences of the x and y coordinates came out roughly constant with depth and the same constant was derived for adjacent tracks in a limited area. The deviations from constancy were random and approximately of the magnitude expected from multiple scattering of the particle. This indicated that the distortion was chiefly of the quadratic type, and at least for emulsion that has never experienced surface stresses, the assumption of quadratic distortion seems justified.

Many other measurements of the macroscopic distortions have led to roughly the same results. Microscopic distortion of a different character is encountered when measuring the multiple scattering of very high-energy particles (see Chapter 8).

6.7 The Distortion Vector

Let the components of a continuous vector function of position, η, describing the macroscopic displacements be α, β, and γ, each in general being a function of position. Normally the distortion vector will not be derivable from a scalar potential, so that the description of the distortion by a single function is not possible.

The geometry of the distorted emulsion is Riemannian in the coordinates of the undistorted emulsion, and, for a complete and general discussion of the transformations of points and of track segments, use

should be made of the appropriate mathematical apparatus (E 26). Our present more modest objectives draw on only the concepts of Euclidean differential geometry.

A vector element of track $d\mathbf{R}_0$ having components dx_0, dy_0, and dz_0 in the unprocessed emulsion is transformed into an element $d\mathbf{R}$ as a result of the processing distortion. The important matrix transforming a differential element of particle path in the unprocessed emulsion to an element of track in the processed emulsion is the following:

$$\mathbf{M} = \begin{pmatrix} 1+\alpha_x & \alpha_y & \alpha_z \\ \beta_x & 1+\beta_y & \beta_z \\ \gamma_x & \gamma_y & 1+\gamma_z \end{pmatrix} \tag{6.7.1}$$

where a subscript denotes partial differentiation with respect to that undistorted coordinate. The vector components of the track element in the processed emulsion, therefore, are:

$$\begin{aligned} dx &= (1+\alpha_x)\,dx_0 + \alpha_y\,dy_0 + \alpha_z\,dz_0 \\ dy &= \beta_x\,dx_0 + (1+\beta_y)\,dy_0 + \beta_z\,dz_0 \\ dz &= \gamma_x\,dx_0 + \gamma_y\,dy_0 + (1+\gamma_z)\,dz_0 \end{aligned} \tag{6.7.2}$$

The determinant, M, of the matrix $\mathbf{M}$ is equal to $1/S_0$ where S_0 is the macroscopically constant shrinkage factor. When distorted components dx, dy, dz of the vector element $d\mathbf{R}_0$ have been measured, the undistorted components can be found by inverting Eq. (6.7.2):

$$\begin{aligned} dx_0 = S_0[&dx(1+\beta_y+\gamma_z+\beta_y\gamma_z-\beta_z\gamma_y) \\ &+ dy(-\alpha_y+\alpha_z\gamma_y-\alpha_y\gamma_z) \\ &+ dz(\alpha_y\beta_z-\alpha_z\beta_y-\alpha_z)] \end{aligned} \tag{6.7.3}$$

$$\begin{aligned} dy_0 = S_0[&dx(\beta_z\gamma_x-\beta_x\gamma_z-\beta_x) \\ &+ dy(1+\gamma_z+\alpha_x+\gamma_z\alpha_x-\gamma_x\alpha_z) \\ &+ dz(-\beta_z+\beta_x\alpha_z-\beta_z\alpha_x)] \end{aligned}$$

$$\begin{aligned} dz_0 = S_0[&dx(-\gamma_x+\gamma_y\beta_x-\gamma_x\beta_y) \\ &+ dy(\gamma_x\sigma_y-\gamma_y\alpha_x-\gamma_y) \\ &+ dz(1+\alpha_x+\beta_y+\alpha_x\beta_y-\alpha_y\beta_x)] \end{aligned}$$

Rather than carrying out the transformations (6.7.3), correction for the shrinkage is often carried out merely by writing

$$\begin{aligned} dx_c &= dx \\ dy_c &= dy \\ dz_c &= S\,dz \end{aligned} \tag{6.7.4}$$

where S is the estimated shrinkage factor. Then the partially corrected vector element of path, $d\mathbf{R}_c \equiv d\mathbf{R} + \mathbf{k}(S - 1)\,dz$, differs from $d\mathbf{R}_0$ because allowance has not been made for the lateral distortion, and the estimated shrinkage factor, S, may not be equal to the true shrinkage factor, S_0.

Solutions of the partial differential equation $S_0 M = 1$, that satisfy the boundary conditions, correspond to all possible continuous distortion-vector functions consistent with a constant shrinkage factor. Some of the most important cases are relatively simple.

A typical kind of distortion, which we shall call gravitational distortion, takes place if the glass on which soft emulsion is mounted is not kept level. There is then a component of shearing stress arising from the component of gravitational field parallel to the emulsion surface. (Considered infinite in extent.) At the glass-emulsion interface $\alpha = \beta = \gamma = 0$, and at the free surface we take $\alpha_z = \beta_z = 0$. Also $\alpha_x = \beta_x = \gamma_x = \alpha_y = \beta_y = \gamma_y = 0$ throughout the emulsion volume. Then $S_0(1 + \gamma_z) = 1$, so we have $\gamma = z_0[(1/S_0) - 1]$. The derivatives α_z and β_z are subject to the condition that they be zero at the free surface, while α and β vanish at the glass surface. The simplest, nontrivial solution that meets these conditions is:

$$\begin{aligned} \alpha &= \frac{K}{T^2}(2z_0 T - z_0^2)\cos\theta \\ \beta &= \frac{K}{T^2}(2z_0 T - z_0^2)\sin\theta \\ \gamma &= z_0\left(\frac{1}{S_0} - 1\right) \end{aligned} \tag{6.7.5}$$

where T is the original emulsion thickness, θ is a constant determined by the direction of the gravitational field, and K is the magnitude of the tangential distortion vector at the free surface. If K is measured in microns and T in millimeters the quantity $D = K/T^2$ measures the distortion in units of the *covan* (*Co*syns-*Van*derhaeghe). This is an example of the "C" type of distortion, but gravity can produce more complex effects, for example, an "S" type of distortion.

If a wet plate is set on edge to dry, the drying will proceed inward and the rigidity of the emulsion will vary with z_0. Then when the drying has been partially completed, if the plate is rotated so that it sets on another edge, the direction of the stress will be altered, and the direction of the distortion vector will vary with z_0.

Distortion describable by Eq. (6.7.5) is frequently found in emulsion and may usually be caused by stresses other than those produced by gravity. We shall, however, refer to this type of distortion as "gravitational."

It is of interest also to study another type of distortion that occurs at the edge of a plate. Suppose the edge is parallel to the y axis. A coordinate frame with its x axis lying in the emulsion-glass interface is selected, so the emulsion is entirely in the first quadrant. Then both α and γ vanish at $z_0 = 0$. We can then treat the distortion in the $x - z$ plane as a two-dimensional problem. We set $\beta = \alpha_y = \beta_y = \gamma_y = 0$, and the differential equation becomes

$$\alpha_x + \gamma_z + (\alpha_x\gamma_z - \gamma_x\alpha_z) = \frac{1}{S_0} - 1 \tag{6.7.6}$$

A solution describing distortion like that usually observed at the edge of a plate can then be found for Eq. (6.7.6):

$$\begin{aligned}
\alpha &= \frac{T}{2\lambda}\left[\ln\left(\frac{u_1 - aS_0u^2 + aS_0u_1u}{u_1}\right)^3 - \frac{u_1}{R}\ln\left(\frac{(u_1 - R)(2u - u_1 - R)}{(u_1 + R)(2u - u_1 + R)}\right)\right] \\
\gamma &= z_0\left[\frac{1}{S_0} - 1 + a\left(u - \frac{u^2}{u_1}\right)\right] \\
u &= z_0 \exp\left(-\frac{\lambda x_0}{T}\right) \\
R &= \left(u_1^2 + \frac{4u_1}{aS_0}\right)^{1/2} \\
u_1 &= T \exp\left(-\frac{\lambda x_1}{T}\right)
\end{aligned} \tag{6.7.7}$$

The quantity a defines the magnitude of the distortion. The constants, λ and u_1, are determined by the mechanical properties of the processed emulsion. The quantity, x_1, is the x_0 coordinate at which the vertical shrinkage passes through the value S_0. The shape of the emulsion edge distorted according to Eq. (6.7.7) is shown in Fig. 6.7.1. This, of course, is a special case and many variations satisfying Eq. (6.7.6) are possible.

The simple distortions mentioned thus far may occur in combinations. In actual emulsions one may find that the distortion vectors at different values of z in the plate are parallel, but it is more common for them to rotate as the depth of observation is varied. Thus, for example, in a typical plate the vectors representing the x-y displacement were found to vary as shown in Fig. 6.7.2. The measurements were made at

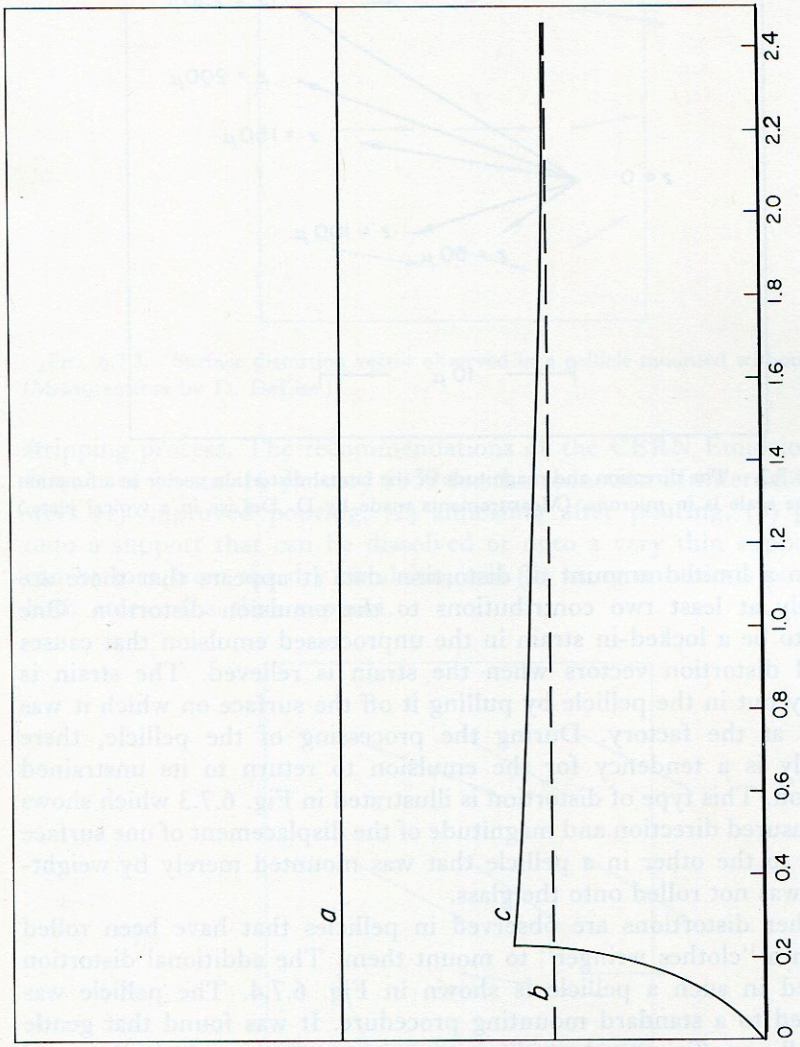

FIG. 6.7.1. In this diagram are shown: (*a*) the original emulsion surface at an edge; (*b*) processed emulsion, surface shrunken, but otherwise undistorted; and (*c*) shape of the distorted surface. The quantity X_1 here is

distances of 50 μ, 100 μ, 150 μ, 200 μ, and 231 μ (surface) from the glass, on 6.2 Bev proton tracks that had entered the emulsion normal to its surface.

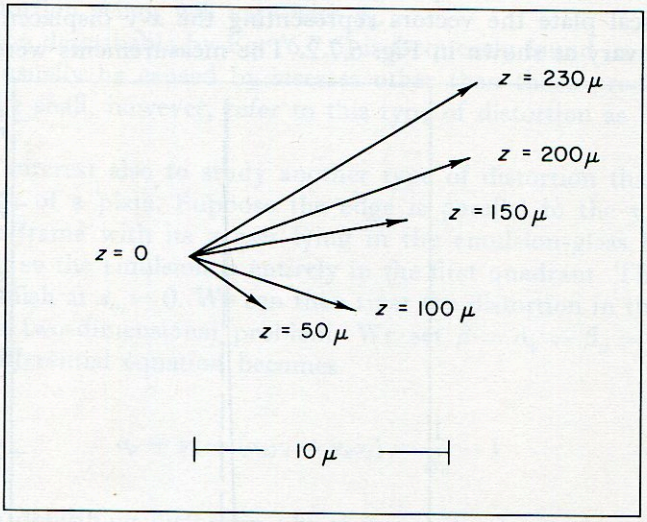

FIG. 6.7.2. The direction and magnitude of the lateral distortion vector as a function of z. The scale is in microns. (Measurements made by D. DeLise in a typical plate.)

From a limited amount of distortion data it appears that there are generally at least two contributions to the emulsion distortion. One seems to be a locked-in strain in the unprocessed emulsion that causes parallel distortion vectors when the strain is relieved. The strain is possibly put in the pellicle by pulling it off the surface on which it was poured at the factory. During the processing of the pellicle, there probably is a tendency for the emulsion to return to its unstrained condition. This type of distortion is illustrated in Fig. 6.7.3 which shows the measured direction and magnitude of the displacement of one surface relative to the other in a pellicle that was mounted merely by weighting; it was not rolled onto the glass.

Further distortions are observed in pellicles that have been rolled through a "clothes wringer" to mount them. The additional distortion observed in such a pellicle is shown in Fig. 6.7.4. The pellicle was subjected to a standard mounting procedure. It was found that gentle hand rolling sufficient only to eliminate excess mounting solution between pellicle and glass did not seem to contribute appreciably to the measured distortion.

Measurements on emulsions that were mounted without any shearing forces being applied still reveal shear-type distortions. This seems to imply that this distortion is introduced by the manufacturer in the

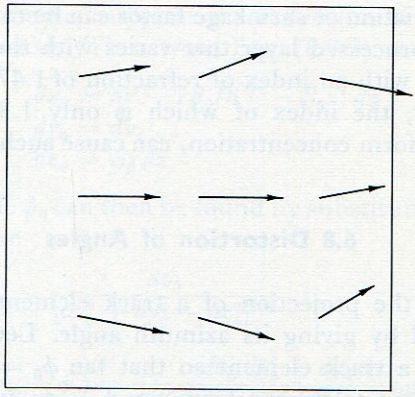

FIG. 6.7.3. Surface distortion vector observed in a pellicle mounted without rolling. (Measurements by D. DeLise.)

stripping process. The recommendations of the CERN Emulsion Conference of 15 and 16 January 1959 for the reduction of internal stresses were (1) improved pouring, (2) annealing after pouring, (3) pouring onto a support that can be dissolved or onto a very thin support that can be torn from the dry emulsion, and (4) using emulsions poured on both sides of a thin support.

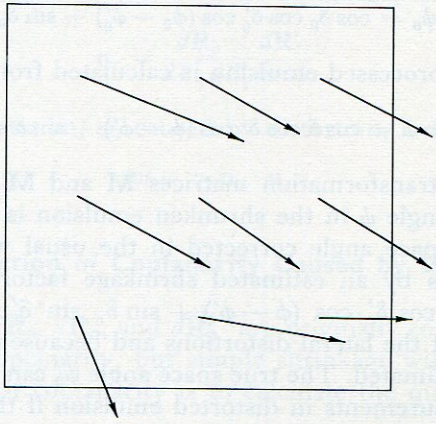

FIG. 6.7.4. Surface distortion vector observed in a pellicle mounted by a usual rolling procedure. (Measurements by D. DeLise.)

Cooksey and Cooksey (CC 30) found long ago that they could reduce the distortions of emulsions used for X-ray spectroscopy by soaking them in water and drying them with alcohol prior to exposure to the X-rays.

An apparent variation of shrinkage factor can be caused by an index of refraction of the processed layer that varies with the distance from the surface. Glycerol, with an index of refraction of 1.4729, matches gelatin fairly well. Water, the index of which is only 1.33, however, if not present with a uniform concentration, can cause such optical distortions.

6.8 Distortion of Angles

The direction of the projection of a track element on the horizontal plane is described by giving its azimuth angle. Let ϕ_0 be the original azimuth angle of a track element so that $\tan \phi_0 = dy_0/dx_0$. Then the distorted angle, ϕ, is calculated from $\tan \phi = dy/dx$; dy and dx being obtained from Eq. (6.7.2). Similarly the original dip angle δ_0 is obtainable by calculating $\sin \delta_0 = dz_0/dR_0$. The distorted dip angle is then found from $\sin \delta = dz/dR$. (The vector $d\mathbf{R}$ has components dx, dy, and dz.) Making a correction for the shrinkage by estimating a shrinkage factor S, the true shrinkage factor being S_0, we obtain a partially corrected dip angle δ_c given by $\sin \delta_c = S(dz/dR_c)$. The vector $d\mathbf{R}_c$ has components dx, dy, and Sdz according to Eq. (6.7.4). The undistorted angle, ψ_0, between two track elements, represented by the vectors $d\mathbf{R}_0$ and $d\mathbf{R}'_0$, is calculated from

$$\cos \psi_0 = \cos \delta_0 \cos \delta'_0 \cos (\phi_0 - \phi'_0) + \sin \delta_0 \sin \delta'_0 \qquad (6.8.1)$$

The angle ψ in processed emulsion is calculated from

$$\cos \psi = \cos \delta \cos \delta' \cos (\phi - \phi') + \sin \delta \sin \delta'$$

by introducing transformation matrices $\mathbf{M}$ and $\mathbf{M}'$ for the two track elements. The angle ψ in the shrunken emulsion is seldom of interest, however. The space angle corrected in the usual way by multiplying all z dimensions by an estimated shrinkage factor S is found from: $\cos \psi_c = \cos \delta_c \cos \delta'_c \cos (\phi - \phi') + \sin \delta_c \sin \delta'_c$. This differs from $\cos \psi_0$ because of the lateral distortions and because the shrinkage factor was inexactly estimated. The true space angle ψ_0 can be found, however, from track measurements in distorted emulsion if the distortion vector is known. Into the Eq. (6.8.1) one merely must put the components of Eq. (6.7.3).

If the two track elements are contained in a sufficiently small volume of emulsion the primed matrix $\mathbf{M}'$ can be taken identical to the unprimed matrix $\mathbf{M}$, but in general two different distortion matrices enter the calculation of angles between track segments.

For a simple example, suppose the distortion is that produced by gravity along the x axis. Then from Eqs. (6.7.3) and (6.7.5)

$$\begin{aligned} dx_0 &= dx - 2S_0D(T - S_0z)\,dz \\ dy_0 &= dy \\ dz_0 &= S_0\,dz \end{aligned}$$

The true space angle ψ_0 can then be found by substituting these quantities in Eq. (6.8.1) where

$$\sin\delta_0 = \frac{dz_0}{[dx_0^2 + dy_0^2 + dz_0^2]^{1/2}}$$

$$\cos(\phi_0 - \phi_0') = \frac{dx_0\,dx_0' + dy_0\,dy_0'}{[(dx_0^2 + dy_0^2)\,(dx_0'^2 + dy_0'^2)]^{1/2}}$$

etc.

6.9 Rotation of a Plane Caused by Distortion

The unit normal to a plane defined by the track elements $d\mathbf{R}_0$ and $d\mathbf{R}_0'$ is:

$$\mathbf{n}_0 = \frac{d\mathbf{R}_0 \times d\mathbf{R}_0'}{dR_0\,dR_0'\sin\psi_0}$$

The distorted unit normal, corrected for shrinkage, is:

$$\mathbf{n}_c = \frac{d\mathbf{R}_c \times d\mathbf{R}_c'}{dR_c\,dR_c'\sin\psi_c}$$

and the angle of rotation, ω, caused by distortion, is found from:

$$\cos\omega = \mathbf{n}_0 \cdot \mathbf{n}_c \tag{6.9.1}$$

6.10 Destruction of Coplanarity Caused by Distortions

If track elements $d\mathbf{R}_0$, $d\mathbf{R}_0'$, and $d\mathbf{R}_0''$ are originally coplanar, distortion may destroy the coplanarity, but simple shrinkage will not. A method often applied to test coplanarity is to calculate the quantity,

$$\frac{(d\mathbf{R} \times d\mathbf{R}') \cdot d\mathbf{R}''}{dR\,dR'\,dR''\sin\psi} \tag{6.10.1}$$

If it fails to vanish, either the tracks originally were not coplanar or distortion and measurement errors, including the multiple scattering of the tracks, have destroyed the coplanarity. When the distortion vector is known, a better quantity to use for a test of coplanarity is:

$$\frac{(d\mathbf{R}_0 \times d\mathbf{R}_0') \cdot d\mathbf{R}_0''}{dR_0\, dR_0'\, dR_0'' \sin \psi_0} \tag{6.10.2}$$

6.11 Spurious Scattering of Tracks Caused by Emulsion Distortion

The measurement of the scattering of a track is generally carried out by aligning the track approximately with the x axis, and at equal intervals along the x axis reading the y coordinate of the track. Let the ith such measurement be y_i. The undistorted value of this coordinate was $y_{0i} = y_i - \beta_i$. The scattering is calculated from the absolute mean value of the second difference, D_i:

$$D_i = | 2y_i - y_{i-1} - y_{i+1} |$$

For an originally straight track, $2y_{0i} - y_{0i-1} - y_{0i+1} = 0$, so the spurious second difference D_{si} caused by distortion is in first approximation (we have neglected distortions which alter the cell length),

$$D_{si} = | 2\beta_i - \beta_{i-1} - \beta_{i+1} |$$

Suppose one measures n cells in a track with an original inclination δ_0 in emulsion of original thickness T. Then $T/(n \sin \delta_0)$ is the cell length, and $T/n = z_{i+1} - z_i$. Then $z_i = iT/n$. If the distortion is given by

$$\beta_i = \frac{K}{T^2}(2Tz_i - z_i^2) \sin \theta$$

then $D_s = (2K/n^2) \sin \theta$. It is notable that when the distortion has this form, the third difference vanishes. If the distortion is of the common type, the third difference may be more useful than the second difference as a measure of multiple scattering.

The effect of the shear distortions is very marked when a track is steep, and too frequently one must abandon attempts to measure the scattering of such tracks because of unanalyzable emulsion distortion. Distortions arising from microscopic displacements of the grains also set a lower limit on the magnitude of D that can be measured even in tracks with no dip (see Chapt. 8).

6.12 Effects of Emulsion Distortion on Particle Range and Range Straggling

If a track happens to lie in the emulsion in such a way that the distortion vector stretches or contracts it, significant errors in range are possible. Randomly oriented tracks in emulsion that is distorted also suffer a deleterious increase in the straggling of their ranges (BSB 55).

If, as is usual, the surface of the emulsion suffers the maximum displacement, tracks that enter through the surface and stop near the glass will suffer the largest changes of range when the distortion vector is in the plane defined by the track and the normal to the emulsion surface. When the magnitude of the surface displacement is K and the emulsion thickness is T, the maximum percentage effect on the range occurs when $\tan 2\delta \approx 2K/T$, δ being the angle of dip. The fractional error in this case is approximately K^2/T^2. A value for K of 35 μ with T equal to 200 μ is not unusual, so that a 3 % error in range because of distortion is commonplace. For unmounted emulsion the effects may be much larger.

The error in the range caused by distortion can be approximated by writing $\Delta\eta$ for the magnitude of the difference of the distortion vectors (corrected for shrinkage) in the emulsion at the beginning and end of a track. Then $\Delta\eta \cos \omega$ is an estimate of the range error where ω is the angle between the vector $\Delta\eta$ and the vector connecting the terminus with the beginning of the track, again corrected for shrinkage. A more analytical approach follows:

The vector element of range, $d\mathbf{R}_c$, corrected only for shrinkage, is related to the uncorrected range element, $d\mathbf{R}$, by the relationship:

$$d\mathbf{R}_c = d\mathbf{R} + \mathbf{k}(S - 1)\, dz$$

where $\mathbf{k}$ is the unit vector along the z axis. Therefore,

$$dR_c^2 = dR^2 + (S^2 - 1)\, dz^2$$

The true range element $d\mathbf{R}_0$ is given by

$$d\mathbf{R}_0 = d\mathbf{R} - d\boldsymbol{\eta}$$

where

$$\begin{aligned} d\eta = \nabla\eta \cdot d\mathbf{R}_0 = {} & \mathbf{i}(\alpha_x\, dx_0 + \alpha_y\, dy_0 + \alpha_z\, dz_0) \\ & + \mathbf{j}(\beta_x\, dx_0 + \beta_y\, dy_0 + \beta_z\, dz_0) \\ & + \mathbf{k}(\gamma_x\, dx_0 + \gamma_y\, dy_0 + \gamma_z\, dz_0) \end{aligned}$$

The connection between $d\mathbf{R}_c$ and $d\mathbf{R}_0$ can now be written:

$$\begin{aligned} d\mathbf{R}_c = d\mathbf{R}_0 + [\mathbf{i}\{\alpha_x\, dx_0 + \alpha_y\, dy_0 + \alpha_z\, dz_0\} \\ + \mathbf{j}\{\beta_x\, dx_0 + \beta_y\, dy_0 + \beta_z\, dz_0\} \\ + \mathbf{k}\{S\gamma_x\, dx_0 + S\gamma_y\, dy_0 + (S - 1 + S\gamma_z)\, dz_0\}] \end{aligned} \tag{6.12.1}$$

$\mathbf{i}$, $\mathbf{j}$, and $\mathbf{k}$ being unit vectors along the coordinate axes.

To include all possible components of distortion in the analysis makes it rather cumbersome. For an illustration of the effect of distortion on ranges let us treat the case of gravitational distortion. Then

$$\alpha = D(2z_0T - z_0^2)\cos\theta$$

$$\beta = D(2z_0T - z_0^2)\sin\theta$$

$$\gamma = z_0\left(\frac{1}{S_0} - 1\right)$$

$$\alpha_x = \alpha_y = \beta_x = \beta_y = \gamma_x = \gamma_y = 0$$

$$\alpha_z = 2D(T - z_0)\cos\theta$$

$$\beta_z = 2D(T - z_0)\sin\theta$$

$$\gamma_z = \frac{1}{S_0} - 1$$

Then the range R_c is related to the true range R_0 by integrating:

$$\begin{aligned} R_c = \int_0^{R_0} \Big[1 + \frac{S^2 - S_0^2}{S_0^2}\sin^2\delta_0 + 4D^2(T - z_0)^2\sin^2\delta_0 \\ + 2D(T - z_0)\sin 2\delta_0\cos(\phi_0 - \theta)\Big]^{1/2} dR_0 \end{aligned} \tag{6.12.2}$$

For the general case of a scattered particle in which δ_0 and ϕ_0 vary along the track, the integration may be carried out by introducing average values of these quantities. It is nearly equivalent to assume that the track was originally straight. This case can be integrated exactly, but leads to a formula too long to reproduce here. In most instances $2D(T - z_0)\sin\delta_0$ is small compared to unity, and an expansion through second order in $\sin\delta_0$ can be made.

Then Eq. (6.12.2) integrates to give:

$$\begin{aligned} R_c = R_0 \Big[1 &+ \frac{S^2 - S_0^2}{2S_0^2} \sin^2 \delta_0 + D(T - \overline{z_0}) \sin 2\delta_0 \cos(\phi_0 - \theta) \\ &+ 2D^2T^2 \sin^2 \delta_0 - 4D^2T\overline{z_0} \sin^2 \delta_0 + 2D^2\overline{z_0^2} \sin^2 \delta_0 \\ &- \frac{D^2T^2}{2} \sin^2 2\delta_0 \cos^2(\phi_0 - \theta) \\ &+ D^2T\overline{z_0} \sin^2 2\delta_0 \cos^2(\phi_0 - \theta) \\ &- \frac{D^2\overline{z_0^2}}{2} \sin^2 2\delta_0 \cos^2(\phi_0 - \theta) + \cdots \Big] \end{aligned} \tag{6.12.3}$$

Here $\overline{z_0}$ and $\overline{z_0^2}$ are mean values for the track.

The main effects on the range of the distortion can be deduced from the first-order correction term:

$$DR_0(T - \bar{z}_0) \sin 2\delta_0 \cos(\phi_0 - \theta).$$

It is proportional to the magnitude of the distortion as measured by D, and to the range, R_0. It is maximum near the glass where $\bar{z}_0$ is small. The track length is reduced or stretched as the dip angle increases, and it is at a maximum when the projection of the track lies along the direction of distortion.

Straggling of ranges is introduced by emulsion distortion because tracks of true length R_0 will have variable observed lengths R_c, depending on how they are oriented in the emulsion. For example, in lowest order of approximation, the standard deviation, σ, of the range caused by randomness of the angle ϕ_0 is:

$$\sigma = \frac{\sqrt{2}}{2} R_0 D(T - \bar{z}_0) \sin 2\delta_0 \tag{6.12.4}$$

It is assumed here that all the tracks have the same dip angle, δ_0, and that they are all confined to a portion of the emulsion having a mean z_0 coordinate equal to $\bar{z}_0$. For more complex distributions, further obvious averaging operations are required.

6.13 Range Error Caused by Tilt of Shrunken Emulsion

If the z axis of the microscope is not strictly parallel to the direction of shrinkage of the emulsion, then a first-order error in the range measure-

ment will be introduced. Suppose an element of track has a length dR_0 in the unprocessed emulsion and a dip angle δ_0 with $\phi_0 = 0$. Then after processing its actual length will be

$$dR = (dx_0^2 + dz^2)^{1/2}$$

Let the inclination angle of the track in the processed emulsion be δ, found from $\tan \delta = \tan \delta_0 / S_0$, so that $dx_0 = dR \cos \delta$, $dz = dR \sin \delta$.

Now suppose the plate itself is inclined so that the angle between the axis of the microscope and the processed track is $(\pi/2) - \delta - \alpha$. Then $[\sin(\delta + \alpha)] = \sin \alpha_0 \sin \theta \cos \delta + \cos \alpha_0 \sin \delta$ where α_0 is the angle of tilt of the plate and θ is an angle defining the axis of rotation of the plate. Now the true length of the track segment, dR_0 before processing is found from:

$$dR_0^2 = dR^2 \cos^2 \delta + S_0^2 \, dR^2 \sin^2 \delta$$

while the tilted plate provides the following estimate of the length of the track segment:

$$dR'^2 = dR^2 \cos^2 (\delta + \alpha) + S_0^2 \, dR^2 \sin^2 (\delta + \alpha)$$

so that:

$$dR' = dR_0 \left(\frac{\cos^2 (\delta + \alpha) + S_0^2 \sin^2 (\delta + \alpha)}{\cos^2 \delta + S_0^2 \sin^2 \delta} \right)^{1/2}$$

Or to first-order in α,

$$dR' = dR_0 \left[1 + \frac{\alpha}{2} \left(S_0 - \frac{1}{S_0} \right) \sin 2\delta_0 \right]$$

Since to this order, $\alpha = \alpha_0 \sin \theta$, dR' can be written:

$$dR' = dR_0 \left[1 + \frac{\alpha_0}{2} \left(S_0 - \frac{1}{S_0} \right) \sin 2\delta_0 \sin \theta \right]$$

In unfavorable situations this may amount to 1.3 % per degree of tilt if $S_0 = 2$, and even more when the shrinkage factor is higher. Of course the first-order error disappears in virtue of the factor $\sin \theta$ if the average effect on randomly oriented tracks is measured, but the range variance produced will remain.

6.14 Mechanical Behavior of Tracks

A track is not merely a series of points but a physical object. The track of a highly ionizing particle in a sensitive emulsion may approach

the condition of a silver rod having well-defined mechanical properties of its own. This is not a new idea. Very early Rotblat and Tai (RT 49) in an emulsion to which they ascribed an unusual shrinkage factor of 2.7 seemed to find differences of alpha-particle ranges depending on how steep the tracks were in the emulsion. The effect was an apparent longitudinal incompressibility of the track.

Their observation now, however, has been largely discounted and ascribed to an incorrect estimate of the shrinkage factor. That the tracks do indeed shrink longitudinally with the emulsion has been demonstrated many times by experiments such as one described by Vigneron (V 49). Tracks of monoenergetic alpha particles oriented at random in emulsion were employed in this experiment. For straight tracks it is assumed that

$$R^2 = X^2 + S^2Z^2$$

where X and Z are respectively the horizontal and vertical projections of the track length and S is the shrinkage factor. The experiment consists of plotting Z^2 for each track against X^2 on rectangular coordinate paper. Each track is indicated by a point on this diagram, and all the points are found to lie on a straight line. The intercept on the X^2 axis is taken to be the square of the true range and the intercept on the Z^2 axis divided into the square of the range gives the square of the shrinkage factor (GH 53).

It is found that there is agreement between this measurement of the shrinkage of the track and the average shrinkage of the emulsion. This fact, and the consideration that the locus of points is straight, has been generally accepted as satisfactory evidence that the tracks observed in these experiments are freely compressible.

The leading sentence of this section is nevertheless true, as shown by some recent experiments. It was observed in early experiments at Bristol and more recently by Muga at Berkeley that, in some uranium-loaded emulsions, tracks of highly ionizing particles are sometimes crooked. This effect was investigated by Dr. H. Heckman and the writer, and it was found that the uranium played no essential part in the process. As development of the tracks of argon ions in unloaded G.5 emulsion with D-19 developer was prolonged, the tracks began to buckle. Tracks of neon, oxygen, nitrogen, and carbon ions likewise could be made to buckle by prolonged development, but as the atomic number was reduced, the effect became harder to produce. Very little, if any, buckling occurred when the tracks were developed in amidol-sulfite developer. It is therefore supposed that physical development, causing the grain size to grow with increasing development time, plays an important role

in the process. An obvious explanation is that as the grains grow they become crowded longitudinally and the track, being restrained by the gelatin from longitudinal extension, finds that it requires less energy to undergo lateral displacements resulting in a buckled track. The grains evidently cohere, because they retain their relative positions and are not merely displaced individually. The configurations assumed by the tracks are illustrated in Fig. 2.3.1. A heavy cosmic ray particle would also be expected to exhibit the same behavior. The track shown in Fig. 9.10.1 does indeed show the effect, but because this track was steep and the possibility of emulsion distortion existed, it was not possible originally to make a definite interpretation of the track behavior.

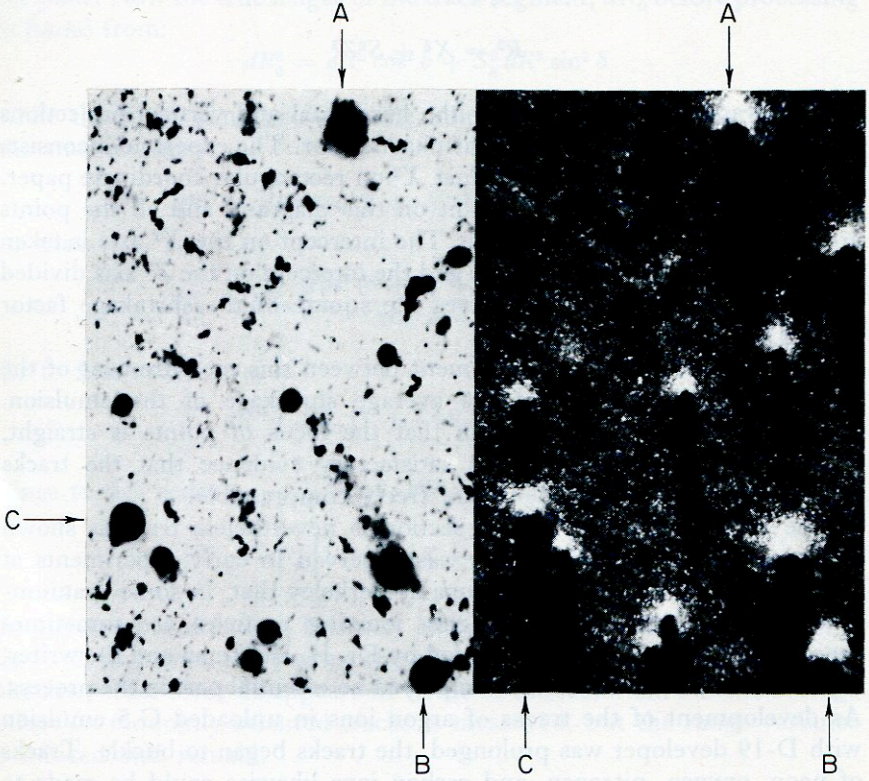

FIG. 6.14.1. Photomicrographs of carbon and argon ion tracks that enter G.5 emulsion normal to its surface. The same field is photographed twice. The light portion shows the tracks viewed in the usual way by transmitted light. Three argon tracks are indicated by arrows. The dark portion is a photograph of the same field of view with the microscope focused on the emulsion surface and the surface illuminated by grazing-incident light. Shadows cast by the protruding tracks can be seen (IDLRL).

Further experiments (H-B 60) have provided remarkable evidence for the longitudinal incompressibility of highly ionizing tracks when they are strongly developed. Carbon and argon ions were caused to traverse a 50 μ G.5 plate perpendicular to its surface. They were developed in D-19 developer diluted 6 to 1 for 1 hr at 22°C. After the plate was developed, fixed, washed, and dried the tracks were found to extend above the emulsion surface for several microns; the argon about 5 μ and the carbon 2 or 3. Each track raised a pointed boss on the emulsion surface, the track acting like a tent pole. The effect is shown in Fig. 6.14.1. Clearly, in this case the tracks did not freely compress as the emulsion shrank. A further study of the mechanical behavior of tracks was made by exposing a plate to ions entering the emulsion with a small angle of dip. After processing, the track grains create a gelatin-covered ridge on the surface of the emulsion.

It is well known that the width of saturated tracks increases with the ionization even when the particle is moving too slowly to produce delta rays of appreciable range. It is probable that part of this track swelling is a mechanical one caused by the growing of crowded grains as a result of physical development.