assigned to Ilford Ltd. is the following: unwanted water-soluble materials are separated by adding an anion soap (one of the commercially available detergents) to the silver halide suspension at a low pH or in the presence of an electrolyte. This is added in sufficient quantity to cause precipitation of the protein containing the halide. The precipitate is subsequently washed and the anion soap removed.

The swelling, when immersed in various processing solutions, of NIKFI type R plates has been compared with that of Ilford G.5 plates at temperatures of 0° C and 24° C by Uvarova and Anosova (UA 57). It was found that the swelling behavior of G.5 emulsion does not parallel that of gelatin, and that it has a greater stability. They deduced that the gelatin of the emulsion is partially replaced by artificial colloids, and furthermore that substances may be present that lower the surface tension in the emulsion layer. Limited swelling in acid media, such as that observed in G.5 emulsion, has considerable importance for the reduction of deformations in processing.

3.4 Mechanical Properties

Unmounted pellicles depend on their own rigidity and strength to maintain dimensions. It is important for accurate measurements to understand the elastic and plastic properties of emulsion and their dependence on the humidity. The emulsion density—which is of paramount importance for the accuracy of much emulsion data—varies with the water content of the emulsion, and with these two quantities, several mechanical properties vary rapidly. They can, therefore, be used to estimate density or water content. As the R.H. is lowered the emulsion becomes stiff, hard, and boardlike. The following table gives the measured Brinell hardness of Ilford G.5 emulsion near normal humidity as a function of the relative humidity.

R. H. (%)	Brinell hardness (kg/mm^2)
50	4.04
60	2.71
70	1.47

For measuring the hardness, a thickness of the emulsion should be used that is large compared to the depth of the depression made by the loaded sphere that is pressed into the surface. It is found that a polished steel ball 12.7 mm in diameter loaded with 5.8 kg is about right. The measured hardness does not vary with loading or ball diameter for a considerable range near these conditions. The depth of the depression is measured by its diameter. The ball is coated with a thin layer of stamp-pad ink, and the diameter, d, of the resulting spot is measured with a low-power microscope. For making the test, samples of emulsion 600 μ in thickness backed by a flat steel plate are used. One calculates the hardness from the formula:

$$\text{Brinell hardness} = 2P/\{\pi D[D - (D^2 - d^2)^{1/2}]\}$$

Here P is the load in kilograms, and D and d are respectively, the diameter of the sphere in millimeters and the diameter of the depression in millimeters.

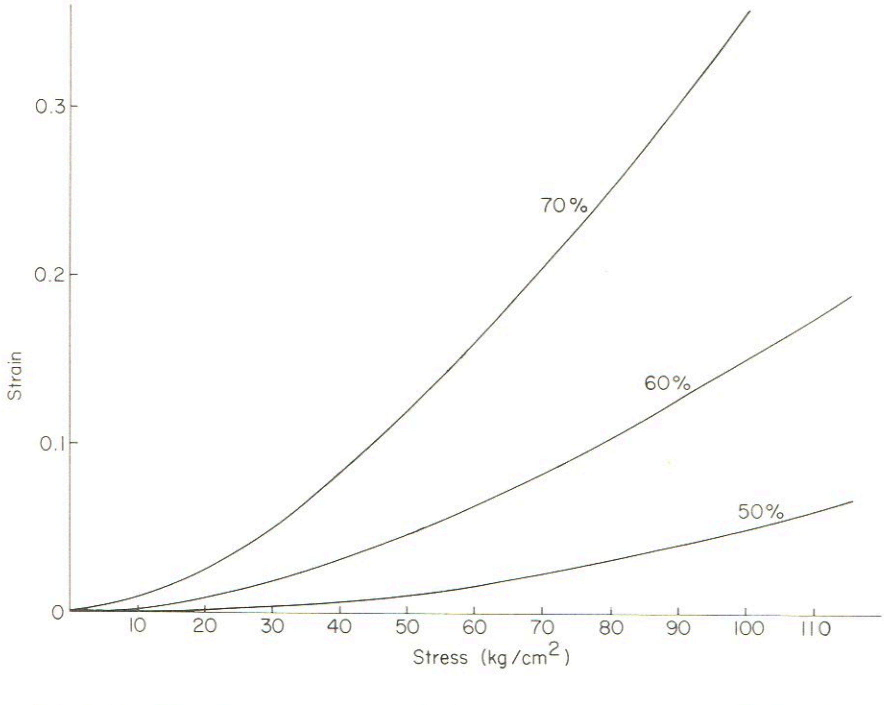

FIG. 3.4.1. The tension-strain curves for emulsion samples at three different relative humidities. The strains are those observed immediately after applying the tension.

The tensile strength of emulsion also changes rapidly with the R.H. in the range of normal humidities. At 50 % R.H. the writer measured a tensile strength of 230 kg/cm^2 for Ilford G.5 emulsion. At 60 % R.H.

the measured tensile strength was 190 kg/cm², while at 70 % R.H. it was 112 kg/cm².

The brittleness of emulsion becomes pronounced as it dries, and the fractional elongation that it will withstand without rupturing is a sensitive test of its dryness.

A true elastic constant apparently does not exist for this material. Under tension, no portion of the stress-strain relationship is linear. Furthermore it creeps so that after a load is applied the elongation continues to increase with time. Curves are shown in Fig. 3.4.1 of the instantaneous extension observed in Ilford G.5 emulsion.

The temporal behavior of Ilford G.5 emulsion at 50 % R.H. under a constant tension of 56 kg/cm² is shown in Fig. 3.4.2. This portion of the curve is labeled *a*.

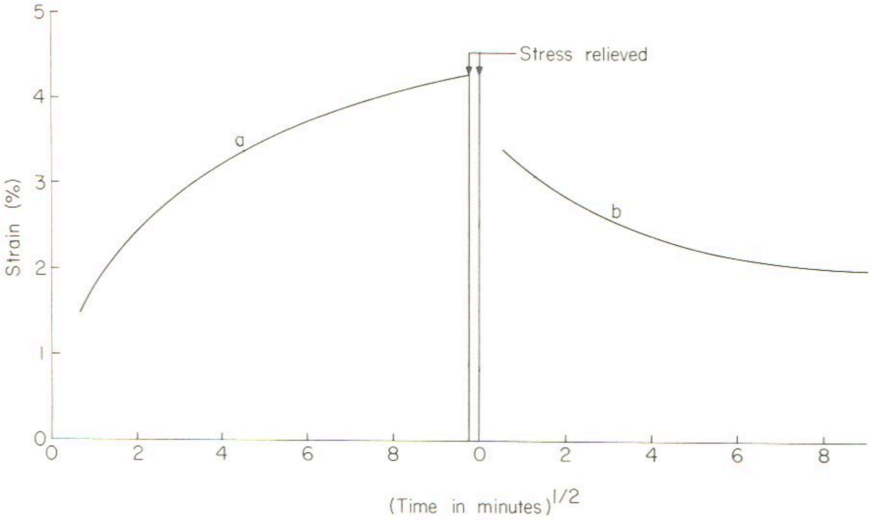

FIG. 3.4.2. The strain observed in a sample of Ilford G.5 emulsion in equilibrium with 50% R.H. as a function of time after subjecting it to a steady tension of 56 kg/cm² (part *a*), and after reducing the tension to zero (part *b*).

Part of the deformation is elastic and part plastic flow, as can be seen from the recovery portion of the curve, labeled *b*. An emulsion sample followed this portion of the curve after the tension was suddenly reduced to zero. It may be noticed that the emulsion requires many hours to readjust itself to the presence of a stress.

In order to obtain Young's modulus correctly for such a material, a dynamic method must be employed. This has been done by Uvarova and Myltseva (UM 58) who systematically studied the mechanical properties of layers of both NIKFI and Ilford emulsions. They found Young's modulus for Ilford G.5 emulsion at 58 % R.H. to be 9 kg/mm^2. Depending on the conditioning treatment given other layers of emulsion, the modulus varied from 6.5 to 340 kg/mm^2. When the highest modulus was measured only 1.2 % of water remained in the emulsion, and the relative humidity was 30 %.

A more thorough study of the mechanical properties of emulsion, particularly the effect of aging on the gelatin, the range of variation of different gelatins, and the behavior under extremes of humidity would be of interest.

3.5 Chemical Composition

Data are obtainable from the manufacturers on the composition of some types of emulsion. A report by Swinnerton and Waller (SW 57) provides detailed information on the density and chemical composition of Ilford emulsions. On the basis of the obtainable information, Table 3.5.1 has been prepared. Further explanations and information are, however, necessary. A serious effort has been made to reproduce the same composition of Ilford emulsion in every batch. Nevertheless the density varies somewhat from batch to batch, even at a fixed relative humidity. For a group of 40 batches at 58 % R.H. the density measurements fell in the interval 3.78 to 3.88 gm/ml. The mean of the distribution was 3.8278 with a standard deviation for a single batch of 0.0177 gm/ml. The mode was between 3.80 and 3.82 gm/ml.

Whereas the mean weight of silver halide per gram of emulsion at 58% relative humidity was 0.82746 ± 0.00268, the actual weight varied from 0.8215 to 0.8355, the mode falling between 0.8235 and 0.8275.

Swinnerton and Waller found for the 40 batches of G.5 emulsion a standard deviation of 0.46 % from the mean batch density, while the writer obtained standard deviations within the batches of 0.21 and 0.23 % when the densities of whole pellicles of two batches of K.5 emulsion were determined. Within a third batch, however, a variation of 0.44 % was found. In an earlier study (B-T 57) when small samples (1 gm) of G.5 emulsion were measured, a standard deviation of only 0.25 % in a single batch was observed. The measurement error in all cases was too small to affect the variance. The composition of Ilford C.2 and E.1 emulsions differs slightly from that given in Table 3.5.1. At 58 % R.H. the bromine content of these emulsions is 1.324 gm/m. and the iodine content is 0.052 gm/ml.

TABLE 3.5.1

CHEMICAL COMPOSITION OF NUCLEAR RESEARCH EMULSIONS[a]

Element	Ilford G. K, and L Series (58% R.H.)	Ilford, diluted 2 × (58% R.H.)	Ilford, diluted 4 × (58% R.H.)	Ilford, diluted 8 × (58% R.H.)	Eastman NTA and NTB (50% R.H.)	Eastman NTC (50% R.H.)	Eastman NTB-4 (50% R.H.)	Fuji ET-7A (65% R.H.)	RIAN P-8 (dry)	RIAN P-9 (dry)	Agfa Wolfen K-2 (60% R.H.)	NIKFI Type T-2	NIKFI Type T-1, T-3	NIKFI Type R	"Standard"
Ag	1.817 ±0.029	1.250	0.769	0.435	1.35	0.74	1.755	1.74	↑	↑	1.76	↑	↑	↑	1.8088
Br	1.338 ±0.020	0.920	0.566	0.320	0.97	0.53	1.335	1.26	2.3[b]	3.3[b]	1.29	3.92[b]	2.77[b]	3.77[b]	1.3319
I	0.012 ±0.000	0.008	0.005	0.003	0.04	0.02	—	0.049	↓	↓	0.05	↓	↓	↓	0.0119
C	0.277 ±0.006	0.362	0.435	0.485	0.29	0.40	0.289	0.278	↑	↑	0.29	↑	↑	↑	0.2757
H	0.0534 ±0.0012	0.0677	0.0799	0.0886	0.05	0.07	0.0534	0.0344	|	|	0.06	|	|	|	0.0538
O	0.249 ±0.005	0.321	0.382	0.425	0.39	0.46	0.313	0.161	0.8[b]	0.6[b]	0.30	0.51[b]	0.74[b]	0.54[b]	0.2522
N	0.074 ±0.002	0.104	0.130	0.148	0.09	0.11	0.0764	0.010	|	|	0.07	|	|	|	0.0737
S	0.007 ±0.000	0.005	0.003	0.002	—	—	—	0.015	↓	↓	0.02	↓	↓	↓	0.0072
	3.8278 ±0.0345	3.038	2.370	1.907	3.19	2.34	3.8218	3.75	3.10	3.9	3.84	4.43	3.51	4.31 ±0.08	3.815

[a] Concentrations in grams per milliliter of each element at the stated relative humidity.

[b] The number entered is the sum of the constituents of its group, as indicated by the arrows. Information on the individual elements was not available.

At a fixed relative humidity the emulsion density suffices to describe it. For the hydrogen content of Ilford G, K, and L emulsions, which is critical for neutron detection experiments, Swinnerton and Waller have given the following formula:

$$\rho_{\mathrm{H}} = 0.1283 - 0.0196\rho \pm 0.0006 \text{ gm/ml}$$

Here ρ is the emulsion density and ρ_{H} the hydrogen density. They have given similar formulas for other components viz.:

$$\begin{aligned}
\rho_{\mathrm{Ag}} &= 0.7169\rho - 0.9271 \pm 0.0047 \text{ gm/ml}\\
\rho_{\mathrm{Br}} &= 0.5281\rho - 0.6830 \pm 0{,}0053 \text{ gm/ml}\\
\rho_{\mathrm{I}} &= 0.0047\rho - 0.0061 \pm 0.0000 \text{ gm/ml}\\
\rho_{\mathrm{C}} &= 0.6871 - 0.1072\rho \pm 0.0036 \text{ gm/ml}\\
\rho_{\mathrm{O}} &= 0.5613 - 0.0815\rho \pm 0.0029 \text{ mg/ml}\\
\rho_{\mathrm{N}} &= 0.2182 - 0.0378\rho \pm 0.0011 \text{ gm/ml}\\
\rho_{\mathrm{S}} &= 0.0214 - 0.0037\rho \pm 0.0001 \text{ gm/ml}
\end{aligned}$$

The electron density can also be calculated similarly. Designating the number of electrons per milliliter by the symbol n_{e}, we obtain

$$n_{\mathrm{e}} = (1.005 + 2.475\rho) \times 10^{23} \text{ per ml}$$

The above formulas apply when the emulsion density changes as a result of fluctuations in the silver halide to gelatin ratio, but they are not strictly applicable when the water concentration is changed. This case will be treated in Section 3.8. The formulas are particularly useful for diluted emulsions at 58 % R.H.

Only nominal values are given in Table 3.5.1, based on published mean halide concentration, but a comprehensive study of the elementary composition of NIKFI types R, K, Я-2, and T-3 emulsions has been made by Rodicheva (R 60). Emulsions made in pure gelatin, some containing surface-active substances, variously hardened emulsions, and diluted emulsions also were analyzed. Bogomolov *et al.* (BSDU 57) have indicated that the silver halide content of NIKFI type R emulsion fluctuates with a standard deviation of only 0.6 %.

The composition of Fuji plates ET-7A was obtained from the Fuji Photo Film Co. Ltd. The percentages of the stated contents add up to 97.23 %.

Ilford C.2 and E.1 emulsions loaded with lithium or boron are available. Standard loadings give 16 mg of lithium and 23 mg of boron per milliliter, respectively. Separated Li^6, Li^7, B^{10}, or B^{11}, supplied by the customer in the form of lithium sulfate or sodium borate, may be incorporated in C.2 or E.1 emulsion at the customer's risk. Emulsion may also be ordered "with extra plasticizer" for use in vacuum or under very dry conditions. Special loading or preparation of the emulsion can often be arranged by correspondence with the manufacturer. The elements Ca, P, Cr, Si, and Na are also found in Ilford emulsions in trace quantities.

The composition given in Table 3.5.1 for Eastman Kodak emulsions, except for Eastman Kodak NTB-4, are approximations deduced from the information supplied by the manufacturer on the relative proportions of the chemical elements present, and from measured densities. Eastman Kodak NTA and NTB emulsions are supplied with boron or lithium loading if desired.

The composition of Agfa-Wolfen emulsion is from a report of Lanius (L 53), while the information on the PR-8 and PR-9 emulsions is derived from the halide concentrations mentioned by Perfilov *et al.* (PNP 57)

Swinnerton and Waller state that in the manufacture of Ilford emulsions glycerol is added in direct proportion to the silver halide content of the emulsion. Sulfur also varies in proportion to the silver halide content in Ilford emulsion, while the Eastman Kodak Company analyses of their emulsions do not mention its presence.

The last column of Table 3.5.1 is an emulsion composition that the writer has taken as standard. As stated above the mean density of the commonly employed Ilford G.5 emulsion at 58 % R.H. comes out to be 3.8278 ± 0.0354 gm/cm³. Several years ago, before these density figures were available, it was necessary to select a standard emulsion density in order to calculate a range-energy relation. "Standard" emulsion at that time was defined to have a density of 3.815 gm/ml. To change this would cause undue confusion and work in connection with the range-energy relation. It is much simpler merely to assume a relative humidity of 60 %; then from Ilford Ltd. data, one obtains a composition of G.5 emulsion. This is the composition defined as "standard emulsion." It turns out, in fact, to be probably the most frequently occurring composition.

If the density of the halide is ρ_h, the density of the gel is ρ_g, and if the fraction by weight of halide in the emulsion is γ, then:

$$\frac{1}{\rho} = \frac{1}{\rho_g} + \left(\frac{1}{\rho_h} - \frac{1}{\rho_g}\right)\gamma\,.$$

where ρ is the density of the emulsion. Putting $\rho_h = 6.473$ gm/ml, then from the composition of standard emulsion, we find for the density of standard gel 1.2914 gm/ml, and the equation becomes:

$$1/\rho = 0.7744 - 0.6199\,\gamma$$

This equation describes the variation of density of the emulsion at standard R.H. (60 %), as the concentration of silver halide varies. It is valid for any degree of dilution of the halide by standard gel.

If C is the fraction of the emulsion volume occupied by halide, the concentration of halide in grams per milliliter is $C\rho_h$. Then

$$C = 0.19306\rho - 0.24968 \text{ gm/ml}$$

When no other information is available, this formula can be used to estimate an emulsion composition.

3.6 Atomic Averages for Standard Emulsion

Emulsion being a composite material is not equivalent to any single element for all processes, but for each type of interaction it has an equivalent atomic number and atomic weight. These may be determined by proper averaging. Also, various functions of atomic number and atomic weight are parameters governing physical processes in matter. In emulsion, effective values of these functions may be defined. In order for these functions to be reducible to numbers, however, an emulsion composition must be assumed. Some numerical averages have been calculated corresponding to the composition of standard emulsion.

From Table 3.5.1 for the chemical compositions we first prepare Table 3.6.1 giving the atomic weight, A_i, number of atoms, N_i per milliliter, and moles per milliliter for the element of atomic number Z:

TABLE 3.6.1[a]

Element	Z_i	N_i ($\times 10^{20}$)	A_i	Moles/ml ($\times 10^{-3}$)
Ag	47	101.01	107.88	16.764
Br	35	100.41	79.916	16.673
I	53	0.565	126.93	0.094
C	6	138.30	12.0000	22.698
N	7	31.68	14.008	5.147
S	16	1.353	32.06	0.216
H	1	321.56	1.0080	53.571
O	8	94.97	16.0000	16.050

[a] Totals: $N = 7.898 \times 10^{22}$ atoms/ml; $n_e = \Sigma N_i Z_i = 1.0446 \times 10^{24}$ electrons/ml $= 1.7338$ moles/ml.

Then:

$$
\begin{aligned}
\langle Z\rangle &= \sum N_iZ_i/N &&= 13.225\\
\bar{Z} &= \sum N_iZ_i^2/n_e &&= 34.56\\
\langle A\rangle &= \sum N_iA_i/N &&= 24.564\\
\bar{A} &= \sum N_iZ_i^2\Big/\sum \frac{N_iZ_i^2}{A_i} &&= 74.4\\
\langle Z^2\rangle^{1/2} &= \Big(\sum N_iZ_i^2/N\Big)^{1/2} &&= 21.38\\
\langle Z^{1/3}\rangle &= \sum N_iZ_i^{1/3}/N &&= 1.927\\
\langle Z^{2/3}\rangle &= \sum N_iZ_i^{2/3}/N &&= 4.658\\
\langle Z^2/A\rangle &= \sum \frac{N_iZ_i^2}{A_i}\Big/N &&= 6.1468\\
\langle A^{1/3}\rangle &= \sum N_iA_i^{1/3}/N &&= 2.377\\
\langle A^{2/3}\rangle &= \sum N_iA_i^{2/3}/N &&= 7.615\\
\langle \ln Z\rangle &= \sum N_i(\ln Z_i)/N &&= 1.593\\
\langle Z^2\ln Z\rangle &= \sum N_iZ_i^2(\ln Z_i)/N &&= 1681\\
\langle Z(Z+1)\rangle &= \sum N_iZ_i(Z_i+1)/N &&= 470\\
\langle Z^{1/3}(Z+1)\rangle &= \sum N_iZ_i^{1/3}(Z_i+1)/N &&= 43.148\\
\left\langle \frac{Z(Z+1)}{A}\right\rangle &= \sum \frac{N_iZ_i(Z_i+1)}{A_i}\Big/N &&= 6.8299\\
\overline{(Z+1)\,Z/A} &= \sum N_iZ_i(Z_i+1)\Big/\sum A_iN_i &&= 16.155\\
\left\langle \frac{Z^{1/3}(Z+1)}{A}\right\rangle &= \sum \frac{N_iZ_i^{1/3}(Z_i+1)}{A_i}\Big/N &&= 1.568\\
\langle Z(Z+1)\ln Z\rangle &= \sum N_iZ_i(Z_i+1)\ln Z_i/N &&= 1726
\end{aligned}
$$

3.7 Characteristic Lengths for Processes in Emulsion

As we have seen, emulsion is a medium comprised of many kinds of atoms. When a particular fast particle penetrates the emulsion, a differential cross section exists for every kind of interaction with each nuclear type, but often these are not separately measurable. For many processes, however, a single number, the "mean free path" can be measured. This may be particularly convenient for describing the frequency of such events in emulsion.

Suppose the mean free path for all nuclear processes is L, and if the mean free paths for the separate possible processes are L_1, L_2, L_3, etc., then:

$$\frac{1}{L} = \frac{1}{L_1} + \frac{1}{L_2} + \frac{1}{L_3} + \cdots$$

An important concept is the "geometrical" free path. This is the mean free path a particle would have in emulsion if each nucleus had its geometrical cross section for interaction with the particle. It has a simpler significance for emulsion than has the nuclear radius, for example. The nuclear size is to a certain extent indefinite, partly because the density of nuclear matter tapers off radially so that the nuclear "surface" is undefined. For some purposes a radius $R = r_0 A^{1/3}$, A being the mass number, with $r_0 = \hbar / M_\pi c \approx 1.4 \times 10^{-13}$ cm, is most useful.

From the standard composition given in Table 3.5.1 the geometrical mean free path, when r_0 is taken to be 1.4×10^{-13} cm for all the emulsion elements, is 27.0 cm.

We know that strongly interacting particles such as pions, nucleons, antinucleons, hyperons, and negative K mesons will penetrate emulsion average distances comparable to the geometrical free path before interacting with nuclei. If the mean free path is somewhat less than the geometrical path, as is the case for antiprotons and negative K mesons, it implies a large elementary cross section between the particle and a nucleon so that even in the low density "atmosphere" of the nucleus an appreciable probability for interaction exists. By the same reasoning, a mean free path for a nuclear interaction large compared to the geometrical, such as that measurable for a muon, an electron, a photon, or a neutrino, indicates a small elementary cross section or processes that are inhibited by the nuclear binding. Mean free paths can be quoted either corresponding to total cross sections or to differential cross sections. For example, a mean free path could be quoted for 100 Mev K^+ mesons inelastically scattering into the 20-30 Mev energy interval, and into the angular interval 30°-40°. Mean free paths for interactions with particular nuclei, such as with hydrogen, can also be quoted. Setting the proton "radius" equal to the Compton wavelength of the pion the geometrical free path for hydrogen interactions is 495 cm. This corresponds to a hydrogen cross section of 63×10^{-27} cm^2.

Processes other than direct interactions with nuclei also are often best described in terms of characteristic lengths. Electromagnetic interactions such as the radiation length, or the pair conversion length are of this character. These will be treated in Volume II.

The geometrical free paths in centimeters for interactions with each elemental component of standard emulsion are:

Ag, 70.9; Br, 87.1; I, 1.16×10^4; C, 224; H, 495; O, 269; N, 876; S, 1.16×10^4.

It is also possible to state the free paths in grams per square centimeter. The geometrical free path in emulsion corresponding to $r_0 = 1.4 \times 10^{-13}$ cm is 103 gm per square centimeter.

3.8 Emulsion of Altered Composition

One may think of altering the composition of standard emulsion by adding to or subtracting from the concentrations of the constituents already present, or by adding a new material as loading.

Instead of considering separately each chemical element, it is often possible to discuss altered compositions of the Ilford G, K, or L emulsion in terms of three or four components. These could be halide, gel, water (and loading), but we prefer to consider a linear combination of standard emulsion, halide, water (and loading). In principle, the concentration of a component then could be negative. The loading must be inert for this analysis to apply; its volume must add directly to the emulsion volume.

At low water concentrations, the combined volume is less than the sum of the standard emulsion plus water volumes. We treat this effect by defining a factor r, so that when a small mass w of water is absorbed by emulsion, the volume of the emulsion is increased by the quantity rw.

If emulsion is placed in vacuum, the water gradually diffuses out of it, and its mean density increases. In Table 3.8.1 data on G.5 emulsion behavior during a period of vacuum desiccation is presented.

It is well known (M 54) that the water content of emulsion displays hysteresis when the ambient relative humidity varies. The effect seems to remain even when months of time are permitted to elapse between observations, so that equilibrium should be approached. In a prolonged series of measurements Mr. C. Cole and the writer studied the behavior of the factor r in Ilford G.5 emulsion that had been brought to equilibrium at various humidities. Quite separate curves of r as a function of relative humidity were found for increasing and decreasing humidity. These loci are shown in Fig. 3.8.1.

TABLE 3.8.1

VACUUM DESSICATION OF EMULSION[a]

Time in vacuum (hours)	Mean density (gm/ml)
0	3.812
4.5	3.853
21.0	3.871
27.5	3.927
45.5	3.962
69.5	3.986
172	4.010
334	4.021
502	4.022

[a] Mean density of a sample of unmounted 600 μ G. 5 emulsion as a function of time in vacuum. (For mounted emulsion multiply all time intervals by four.)

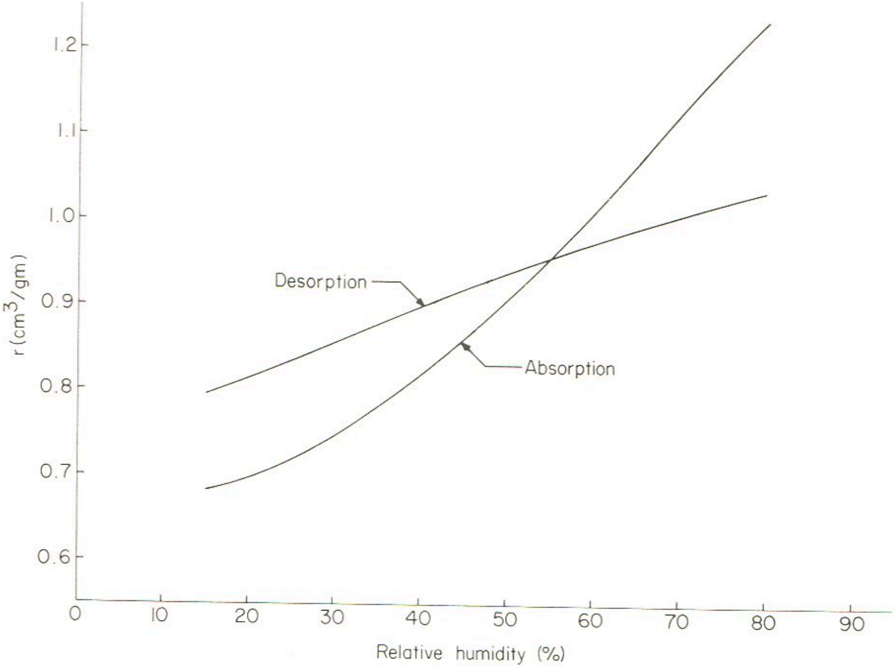

FIG. 3.8.1. The volume change observed per unit mass of water absorbed or desorbed by emulsion as a function of relative humidity.

Such data are difficult to obtain reliably. The approach to equilibrium is very slow and the changes in weight and volume are small. It is suspected that emulsion behavior changes with its age and with other conditions. Surprisingly, both curves seem to rise above unity at high humidities. Of course r must tend toward unity eventually. On the basis of their observations, Swinnerton and Waller (SW 57) took r to be a constant, 0.84 ml/gm, for normal emulsion. Fortunately its effect is small, and usually one can assume that the emulsion is affected little by its previous history. G.5 emulsion density versus relative humidity is graphed in Fig. 3.8.2 for five samples of G.5 emulsion. Three of these were measured by Swinnerton and Waller.

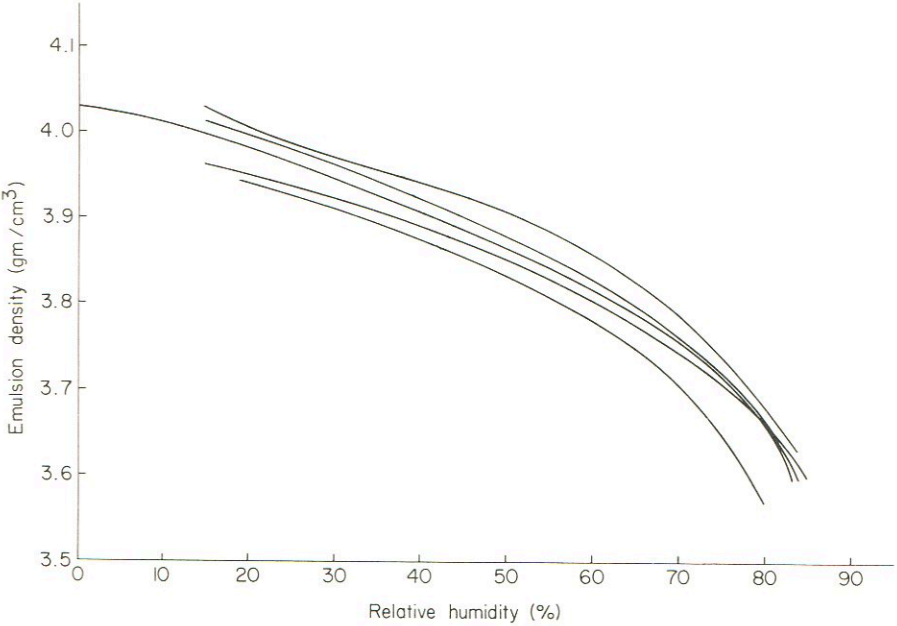

FIG. 3.8.2. Density of five samples of Ilford G.5 emulsion as functions of the relative humidity. Individual differences between them exist.

The weight of a sample of emulsion is the sum of the weights of all components:

$$W = W_e + W_h + W_w + W_l$$

where the subscripts e, h, w, and l refer to standard emulsion, halide, water, and loading, respectively. Similarly the volume, V, is:

$$V = V_e + V_h + \bar{r}V_w + V_l$$

so the density is:

$$\rho = \frac{W_e + W_h + W_l + W_w}{V_e + V_h + \bar{r}V_w + V_l}$$

Here $\bar{r}$ is an average value of the factor, r, previously defined. If it simplifies the particular problem, one may introduce the fact that the weight of any component is equal to its volume times its density. Of course, $W_w = V_w$ in the cgs system.

The density of the halide is taken to be 6.473 gm/ml by Swinnerton and Waller (SW 57) and the standard emulsion density is 3.815 gm/ml. Swinnerton and Waller found unexplained fluctuations in the apparent gel density. The mean gel density found in emulsion of mean density 3.8278 gm/ml, was 1.2932 gm/ml.

For some purposes the exact gel composition is of little importance. For example, when expressed in grams per cubic centimeter, the radiation length of any composition of C, N, and O is roughly the same as that of N. The stopping behavior of any gel composition can be well approximated by a combination of carbon and water, and even water alone is not far off. For this reason, if unloaded emulsion simply is described by its density and its halide content, one can already obtain a good estimate of its stopping power, the geometrical free path, or the radiation length. When the composition is altered in such a way that it cannot be described by a linear combination of a few components, one may consider lumping a number of small components into a fictitious "loading." When expressed in grams per cubic centimeter, the stopping power, for example, of this loading may be estimated well enough so that no appreciable over-all error is made.

3.9 Emulsion Statistics

Regardless of how thoroughly it is mixed, the composition of emulsion varies from one small volume to the next. This is an effect of the granularity. The silver halide is present in lumps of variable size that are randomly distributed throughout the matrix. Because of the finite non-penetrating volumes occupied by the crystals, and their various sizes, the statistics of the emulsion is complex. The crystals of halide, however, especially the smaller ones, are geometrically describable as spheres. This may be seen in the electron photomicrographs of Ilford emulsion shown in Fig. 3.2.1.

In this section we shall utilize a model of the emulsion that is slightly idealized, but closely resembles real emulsion. It consists of a diversity of crystals, but all are assumed to be spheres. No stratification or marbling that might destroy the randomness is present. The crystals are assumed not to interpenetrate, but it is supposed that they can come arbitrarily close together.

A particle trajectory through this emulsion is illustrated by Fig. 3.9.1. Some of the quantities studied in this and later sections are labeled.

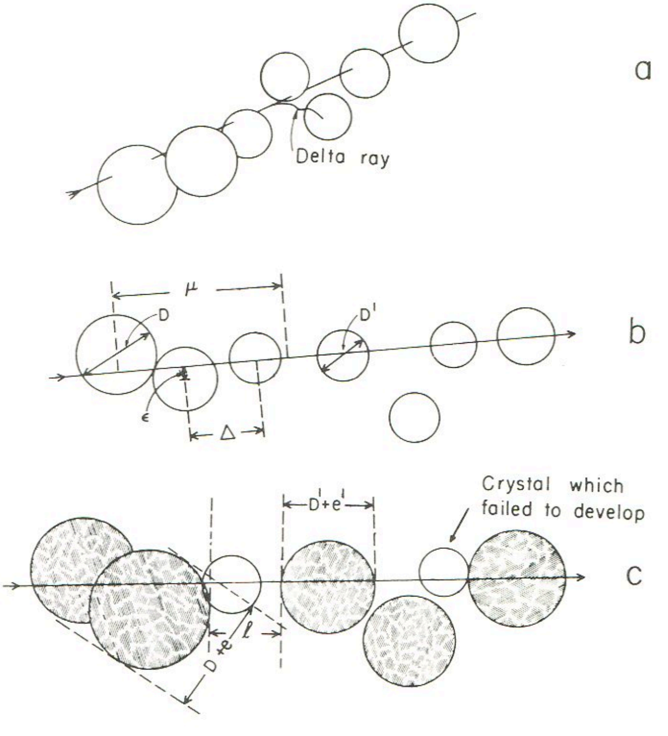

FIG. 3.9.1. (*a*) Illustrating the path of charged particle through emulsion crystals and the production of a delta ray. (*b*) Projection on plane containing particle path. (*c*) Projection of developed track grains on plane containing particle path (IDLRL).

3.9.1 Expectation Values

Let the fraction of crystals with diameters in the interval D to $D + dD$ be $F(D)dD$. Then the following mean values are defined.

The mean grain diameter:

$$\langle D \rangle = \int_0^\infty DF(D)\, dD \tag{3.9.1}$$

The mean square grain diameter:

$$\langle D^2\rangle = \langle D\rangle^2 + \sigma^2 = \int_0^\infty D^2 F(D)\, dD \tag{3.9.2}$$

The mean grain cross section:

$$\langle A\rangle = \frac{\pi}{4}\langle D^2\rangle = \frac{\pi}{4}\langle D\rangle^2 \left(1 + \frac{\sigma^2}{\langle D\rangle^2}\right) \tag{3.9.3}$$

The mean crystal volume:

$$\langle V\rangle = \frac{\pi}{6}\int_0^\infty D^3 F(D)\, dD = \frac{\pi}{6}\langle D\rangle^3 \left(1 + \frac{3\sigma^2}{\langle D\rangle^2} + \cdots\right) \tag{3.9.4}$$

Let a crystal be penetrated by a particle moving in a straight line. The segment of path in the crystal is of length δ. The joint probability, $u(\delta, D)$, that the crystal diameter should be in the interval dD and that the path segment should be in the interval $d\delta$ is given by

$$u(\delta, D) = \frac{2\delta\, d\delta}{\langle D^2\rangle} F(D)\, dD \tag{3.9.5}$$

The mean length, $\langle \delta \rangle$, of such path segments in crystals is:

$$\langle\delta\rangle = \frac{2}{\langle D^2\rangle}\int_0^\infty F(D)\, dD \int_0^D \delta^2\, d\delta = \frac{2}{3}\frac{\langle D^3\rangle}{\langle D^2\rangle} \approx \frac{2}{3}\langle D\rangle \left(1 + \frac{2\sigma^2}{\langle D\rangle^2}\right) \tag{3.9.6}$$

and their variance, $\langle \delta^2 \rangle - \langle \delta \rangle^2$, is:

$$\sigma_0^2 = \frac{9\langle D^2\rangle\langle D^4\rangle - 8\langle D^3\rangle^2}{18\langle D^2\rangle^2} \approx \frac{\langle D\rangle^2}{18}\left(1 + \frac{13\sigma^2}{\langle D\rangle^2}\right) \tag{3.9.7}$$

The average number, n, of grains encountered in unit path is:

$$n = N\langle A\rangle = \frac{\pi N\langle D^2\rangle}{4} = \frac{\pi N}{4}\langle D\rangle^2 \left(1 + \frac{\sigma^2}{\langle D\rangle^2}\right) \tag{3.9.8}$$

where, N, the average number of grains per unit volume is given by:

$$N = \frac{C}{\langle V\rangle} \approx \frac{6C}{\pi\langle D\rangle^3}\left(1 - \frac{3\sigma^2}{\langle D\rangle^2}\right) \tag{3.9.9}$$

Here C is the volume concentration of silver halide in the emulsion.

A formula known as "Jdanov's relation" has frequently been used incorrectly to calculate n. Since $N \langle V \rangle = C$, and $n = N \langle A \rangle$, one can write:

$$n = C \frac{\langle A \rangle}{\langle V \rangle} \approx \frac{3C}{2\langle D \rangle} \left(1 - \frac{2\sigma^2}{\langle D \rangle^2}\right) \tag{3.9.10}$$

The term $-2\sigma^2/\langle D \rangle^2$ is not present in Jdanov's relation, as usually employed, and this may result in a considerable error. For Ilford G.5 emulsion it is about 7 %. The reason for the discrepancy can be stated quite simply: the probability for encountering a grain is weighted by the square of its diameter; consequently the grains traversed by a particle are on the average larger than typical emulsion grains.

3.9.2 Distribution Functions

Let emulsion containing $N \int_D^\infty F(D)\, dD$ crystals per unit volume with diameters exceeding D be traversed by a particle moving in a straight line. The number of crystals whose centers lie at a distance between ρ and $\rho + d\rho$ from the particle trajectory, and whose diameters are contained in the interval dD is

$$2\pi N F(D)\, \rho\, dD\, d\rho \tag{3.9.11}$$

per unit path. A particular crystal of the above description is labeled "crystal C", and any other such crystal is of "type C."

Now one considers a different crystal, C′, the diameter of which lies in the interval dD', and the center of which is removed from the trajectory a distance in the interval, $d\rho'$. The center of C′ may be found anywhere with equal probability except in an excluded sphere of radius $(D + D')/2$ around crystal C. This statement requires justification. Dr. Robert G. Glasser has pointed out that under some circumstances it is not a valid assumption. For example, suppose all the crystals are equal spheres, and the tightest possible packing is assumed. Then the distance between neighbors is always the minimum distance D. The crystals of emulsion have a distribution of diameters, however. In addition, only half of the emulsion volume is occupied by silver halide. Two degrees of rotational freedom about the center of the excluded sphere exist. Unless the conditions are very unusual, therefore, these circumstances introduce enough degrees of freedom to destroy such a possible short-range order. Even were some ordering present, the average density of centers remains constant. Only some peaking of the probability then occurs at particular small distances. At larger distances, say two or three crystal diameters, ordering effects must disappear entirely. As we shall see in Chapter 9, the most important application of this assumption arises in the analysis of the gap structure of the developed track of a particle. It is then assumed that randomness exists beyond about $2\frac{1}{2}$ crystal diameters.

Now with respect to crystal C′ we shall assume that the probability that it is to be found in a volume element dV is $NF(D')\,dD'\,dV$.

In order that a segment of the particle path lie within crystal C, ρ must be less than $D/2$. Therefore, from Eq. (3.9.11), the mean number of crystals with diameters in the interval dD that are penetrated by the particle in unit path is:

$$\frac{\pi N D^2}{4} F(D)\,dD \tag{3.9.12}$$

and the probability that any particular crystal encountered belongs to class C is:

$$\frac{8\rho\,d\rho\,F(D)\,dD}{\langle D^2\rangle} \tag{3.9.13}$$

One measures distance, μ, along the particle trajectory from the point defined by the projection of the center of C on the trajectory. The "distance" between crystals C and C′ is the distance between the projections of their centers on the particle path.

The expected number of crystals satisfying the description of C′ to be found at a distance between μ and $\mu + d\mu$ from crystal C is:

$$\frac{16\pi N}{\langle D^2\rangle} \rho\rho' F(D) F(D')\,dD\,dD'\,d\rho\,d\rho'\,d\mu \tag{3.9.14}$$

One now asks for the probability, p, that the particle reach a distance μ after traversing C without penetrating a crystal of type C′. This is formulated as follows:

$$\frac{1}{p}\frac{dp}{d\mu} = -\frac{16\pi N}{\langle D^2\rangle} \rho\rho' F(D)\,F(D')\,dD\,dD'\,d\rho\,d\rho' \tag{3.9.15}$$

The next step is to calculate the probability, P_1, that the particle may go a distance exceeding μ without traversing a crystal of any description. This is found from:

$$\frac{1}{P_1}\frac{dP_1}{d\mu} = -\frac{16\pi N}{\langle D^2\rangle} \iiiint F(D)\,F(D')\,\rho\rho'\,d\rho\,d\rho'\,dD\,dD' \tag{3.9.16}$$

The projected distance, Δ, measured along the particle path, between centers of crystals successively traversed by a particle in emulsion, is the "elementary track cell." Equation (3.9.16) provides the fundamental statistical law describing how this quantity varies. From Eq. (3.9.16), the probability that the length of the elementary cell exceed μ is:

$$P_1 = \exp\left[-\frac{16\pi N}{\langle D^2\rangle}\iiiint F(D)\,F(D')\,\rho\rho'\,d\rho\,d\rho'\,dD\,dD'\,d\mu\right] \tag{3.9.17}$$

Because crystal C occupies a finite volume from which are excluded all other grains, a restriction on the limits of integration exists. The constraint requires that μ^2 can never be less than

$$\left(\frac{D+D'}{2}\right)^2 - (\rho+\rho')^2 \tag{3.9.18}$$

To evaluate Eq. (3.9.17) one distinguishes two regions of $(D + D')/2$ $(= \mathscr{D})$.

When $\mathscr{D}$ is less than μ, the excluded volume imposes no restriction on ρ and ρ', and the integral $\iint \rho\rho'\,d\rho\,d\rho'$ has the value $D^2/16\,[\mathscr{D} - (D/2)]^2$. The distances ρ and ρ' are constrained only to remain less than $D/2$ and $D'/2$ respectively.

When $\mathscr{D}$ is greater than μ, three subregions must be distinguished. For the moment one simply defines

$$\iint \rho\rho'\,d\rho\,d\rho' = \frac{1}{64}M(\mathscr{D}, D, \mu)$$

when $\mathscr{D} > \mu$, and Eq. (3.9.16) is written:

$$\frac{1}{P_1}\frac{dP_1}{d\mu} = \frac{\pi N}{4\langle D^2\rangle}\left[\int_0^{\mu} d\mathscr{D}\int_0^{2\mathscr{D}} F(D)\,F(2\mathscr{D}-D)\,D^2(2\mathscr{D}-D)^2\,dD\right.$$
$$\left. + \int_{\mu}^{\infty} d\mathscr{D}\int_0^{2\mathscr{D}} F(D)\,F(2\mathscr{D}-D)\,M(D,\mathscr{D},\mu)\,dD\right] \tag{3.9.19}$$

When $\mathscr{D}$ exceeds $\langle D \rangle$ by an amount comparable to the standard deviation of the grain diameter, the functions $F(D)$ and $F(2\mathscr{D} - D)$ fail to overlap. Therefore, the second integral of Eq. (3.9.19) becomes negligible when $\mu > \mu_0$; μ_0^2 is chosen as a quantity of the order of magnitude of $\langle D^2 \rangle$. For complete rigor, μ_0 must be set equal to the largest diameter occurring in the grain-size distribution.

In the first integral, when the maximum grain diameter is μ_0 and $\mu > \mu_0$, the integrations over D and D' are independent and unrestricted. Then the right side of Eq. (3.9.19) becomes a constant:

$$-\frac{\pi N}{4\langle D^2\rangle}\int_0^{\mu_0} D^2F(D)\,dD\int_0^{\mu_0} D'^2F(D')\,dD' = -\frac{\pi N\langle D^2\rangle}{4} = -n \qquad (3.9.20)$$

The probability, P_1, that a particle may traverse a distance exceeding $\mu(\mu > \mu_0)$ without encountering any crystal is therefore:

$$P_1 = W\exp(-n\mu) \qquad (3.9.21)$$

in which the constant factor W is given by:

$$W = \exp\left\{-\frac{\pi N}{4\langle D^2\rangle}\int_0^{\mu_0} d\mu\left[\int_0^{\mu} d\mathscr{D}\int_0^{2\mathscr{D}} F(D)\,F(2\mathscr{D}-D)\,D^2(2\mathscr{D}-D)^2\,dD\right.\right.$$
$$\left.\left.+\int_{\mu}^{\infty} d\mathscr{D}\int_0^{2\mathscr{D}} F(D)\,F(2\mathscr{D}-D)\,M(\mathscr{D}, D, \mu)\,dD\right] + n\mu_0\right\}$$

The function $M(D, \mathscr{D}, \mu)$ has a different value in each of three subregions in the integration range of D. The three functions $M_{\rm I}$, $M_{\rm II}$, and $M_{\rm III}$ have been evaluated, and are as follows:

(I) $0 < D < 2(\mathscr{D} - p)$; $p^2 = \mathscr{D}^2 - \mu^2$

$$M_{\rm I} = D^4 - 4\mathscr{D}D^3 + 4\mathscr{D}^2D^2 - (8/3)\,p^4$$

(II) $2(\mathscr{D} - p) < D < 2p$

$$\begin{aligned} M_{\rm II} = {} & \frac{D^4}{2} - (8/3)\,pD^3 + (4\mu^2 + 16p\mathscr{D} - 12\mathscr{D}^2)\,D^2 \\ & + (32\mathscr{D}^3 - 32p\mathscr{D}^2 - 16\mathscr{D}\mu^2)\,D \\ & + \left(16\mathscr{D}^2\mu^2 - 24\mathscr{D}^4 + \frac{64}{3}\,p\mathscr{D}^3\right) \end{aligned} \qquad (3.9.22)$$

(III) $2p < D < 2\mathscr{D}$

$$\begin{aligned} M_{\rm III} = {} & -8(\mathscr{D} - p)^2\,D^2 + 16\mathscr{D}(\mathscr{D} - p)^2\,D \\ & - (8/3)\,(\mathscr{D} - p)^3\,(3\mathscr{D} + p) \end{aligned}$$

Then in general, the probability P_1 that the elementary cell length, Δ, exceeds μ is:

$$
\begin{aligned}
P_1 = \exp\Big\{ -\frac{\pi N}{4\langle D^2\rangle} \int_0^\mu \Big[& \int_0^\mu d\mathscr{D} \int_0^{2\mathscr{D}} F(D)\,F(2\mathscr{D}-D)\,D^2(2\mathscr{D}-D)^2\,dD \\
& + \int_\mu^\infty d\mathscr{D} \int_0^{2(\mathscr{D}-p)} M_{\mathrm{I}} F(D)\,F(2\mathscr{D}-D)\,dD \\
& + \int_\mu^\infty d\mathscr{D} \int_{2(\mathscr{D}-p)}^{2p} M_{\mathrm{II}} F(D)\,F(2\mathscr{D}-D)\,dD \\
& + \int_\mu^\infty d\mathscr{D} \int_{2p}^{2\mathscr{D}} M_{\mathrm{III}} F(D)\,F(2\mathscr{D}-D)\,dD \Big]\,d\mu \Big\}
\end{aligned}
\qquad (3.9.23)
$$

with $p^2 = \mathscr{D}^2 - \mu^2$.

3.9.3 Fluctuation of Composition

When the volume concentration of halide is C, then in traversing a distance μ, the path length h in silver halide has a mean value $\langle h \rangle = C\mu$. Using the above theory it has been shown (B 59), that the variance, σ_h^2, of h is

$$\sigma_h^2 = \pi N \left[\frac{\langle D^4\rangle}{8} - \frac{\langle D^3\rangle^2}{9\langle D^2\rangle} \right] \mu \qquad (3.9.24)$$

(It is assumed that μ exceeds the diameter of the largest crystal present.)

For crystals all of the same size

$$\frac{\sigma_h}{\langle h\rangle} \approx \left(\frac{1}{8n\mu} \right)^{1/2} \qquad (3.9.25)$$

This means, for example, that in a 10 μ segment of path in G.5 emulsion the amount of silver halide penetrated fluctuates by at least 10 %.

CHAPTER 4

Preliminary Processing Procedures

The various processing treatments received by the emulsion can be broken down into two groups. One category consists of those operations carried out after the emulsion has been exposed to track-producing radiations and to the light used for grid printing. These are reserved for Chapter 5. A rather large number of operations and precautions already will have been initiated before the exposures are completed. This chapter treats these and related topics.

4.1 Conditions of Shipping, Handling, and Storage

Soon after nuclear-track emulsion is poured, it will commence to record the tracks of particles traversing it. These are of three sorts: (*a*) cosmic radiation, (*b*) natural radioactivity, (*c*) accidental or deliberate exposure to radiation from accelerators, X-rays, or artificial radioactive sources. There is also the possibility of exposure to light. General fog tends to increase with age. Except for those produced deliberately, all developable silver halide crystals in the plate are deleterious to its quality and ultimately make it unusable. For G.5 emulsion 600 μ thick, an accumulated radiation dosage of 20 milliroentgens (mr) is about as much as can be tolerated. Somewhat more is tolerable in thinner emulsion layers. One mr is equivalent to about 25,000 minimum ionizing tracks per square centimeter or about 10^5 quanta of annihilation radiation (energy 0.51 Mev) per square centimeter.

Ilford Ltd. recommends their S902 amber safelight to avoid light blackening of the emulsion in the dark room. With a 25 watt bulb behind this safelight, a 15 min exposure of G.5 emulsion at a distance of 3 ft does not increase the blackening. Any lighting arrangement should be tested with partially covered emulsion plates to determine if any blackening is caused by the light. For photography, nuclear-track emulsion would be classed as slow. Except for emulsions with extremely small grains, the silver halide content is so high that the emulsion is opaque. If the top surface of a plate is accidentally exposed to a moderate amount of light, the blackening on development is confined to a thin layer of the emulsion, and may be rubbed off with soft paper or cloth dipped in alcohol. Of course, some of the gelatin and segments of tracks are also removed by this process, but valuable exposures have been salvaged in this way. When ranges of particles entering the emulsion surface are to be measured, it is recommended not to wipe the emulsion surface. When light enters from the glass edge or from the glass side of a plate, it is usually ruined.

From Rosenfeld *et al.* (R 55) we obtain the estimates of the rate of accumulation of cosmic ray background radiation shown in the following tabulation.

Sea level	35 mr/year
5,000 ft	50 mr/year
10,000 ft	100 mr/year
15,000 ft	200 mr/year
25,000 ft	375 mr/year

Local radioactivity in soil, brick, etc., is about that from typical sedimentary rocks, about 40 mr per year, while that from granite is about 100 mr per year.

At sea level, an unshielded emulsion plate, therefore, may be expected to accumulate perhaps 100 mr per year. In terms of slow electron tracks, Orear and Waller (R 55) found 4250 ± 200 tracks per cubic centimeter per day at sea level.

Transporting the emulsion by air considerably increases the rate of accumulation of cosmic radiation, while the natural radioactivity from the ground is reduced by the air absorption. However, if the emulsion is stowed near radioactive cargo or near an instrument panel, which on many airplanes is liberally supplied with radioactive materials used to illuminate dials, the emulsion may be spoiled in a few hours from these sources of radiation. Irradiation by passage through radioactive clouds has also been suspected (HL 55). Emulsion should be handled in laboratory buildings which have never been used for research in radioactivity, because almost all old laboratories of this sort are contaminated. Bringing of radioactive sources into an area where emulsion is handled or stored should be strictly forbidden. For storage it is recommended that emulsion be kept in an iron box with walls several inches thick. The iron will shield out most local gamma radiation, and is itself, in contrast to lead, usually very free of radioactivity. If the box is stored underground, perhaps 50 ft or deeper, most of the cosmic radiation will be absorbed. It is a good idea to store the emulsion in the form of a large stack, because emulsion itself is very clean from the radioactive standpoint, and the self shielding of a large stack is remarkable. With these precautions, it is possible to store emulsion with little deterioration for several months.

Deep in the earth (600 meters) most of the cosmic ray effects are eliminated so that Walters (W 52) was able to analyze the various other kinds of background tracks affecting plates of 200 μ G.5 emulsion that were poured and kept beneath the ground. He observed that in a square centimeter these plates received per day 57 ± 5 thorium alpha particles from the glass, 100 ± 5 electrons from K^{40} in the glass, 83 ± 6 electrons of C^{14} from the gel, and 10 ± 2 tracks from the shielding.

The spectrum of typical background electrons found in the emulsion is peaked below 100 Kev, and ordinarily most of them originate in the emulsion, indicating that they are probably Compton or photoelectrons. High energy photons also produce pairs, and this component of the photon spectrum is easily analyzed. Slow neutrons captured in emulsion produce beta radiation chiefly from the radioactive silver and bromine isotopes formed, and also induce the reaction N^{14} (n, p) C^{14}. Fast neutrons produce proton recoils and neutron disintegration stars. The protons are easily recognizable, and the neutron stars are distinguishable from photon stars by the lack of tracks of accompanying electron pairs in the emulsion.

Tracks of fast protons, μ mesons, positron-electron pairs, and cosmic ray stars are found in the emulsion after exposure to the ubiquitous cosmic radiation. At sea level about 10 μ-e events per cubic centimeter per month may be observed. Pions, and even hyperons and *K*-mesons, also, are sometimes seen. These may constitute a serious background contamination which must be subtracted when rare events are being tabulated.

Alpha-particle background in the emulsion is not normally a serious problem, but may become so for studies in which one is employing emulsion as a detector for very weak alpha-emitting sources, as in the study of rock specimens. Deutsch and Dodd (DD 53) have observed the number of alpha-particle disintegrations in (a) standard Ilford C.2 emulsion, (*b*) C.2 emulsion prepared with deashed gelatin, (*c*) deashed gelatin additionally washed in an acid solution. The standard emulsion recorded 42 ± 4 events per cubic centimeter per year. In the sample containing the deashed gelatin washed in distilled water, this was reduced by about a factor of two, and the washing caused a pronounced reduction of the three- and four-prong alpha stars assignable to radium. The deashed gelatin reduced the number of single-prong events assignable to U, Th, and Po by nearly a factor of two, and also reduced the number of five-prong stars assignable to RdTh, although the total number of such events was always small. Further purification appeared not to bring about a further reduction in the alpha-particle track density. The concentration of thorium in the glass plate backing the emulsion was found to be $\approx 10^{-6}$ gm/gm. A small number of alpha particles also entered the emulsion surface and corresponded roughly with the normal radon content of the air.

Very occasionally a particle of dust occluded in the emulsion or detected at an emulsion surface is observed to be intensely alpha-or beta-active. Some of these objects doubtless are components of air contamination produced in fission reactions. A photomicrograph of such an event is shown in Fig. 4.1.1.

Very high pressures (≈ 1000 kg/cm^2) can cause blackening of the emulsion, but normally such pressures will be experienced by the emulsion only when abraded or scratched by a sharp point. Marks develop on the emulsion surface if it has been scratched or rubbed. Scratch marks may somewhat resemble tracks, and someone who is a complete novice might mistake them for particle tracks. They are, however, confined to the emulsion surface and differ from tracks in many other respects, so that they are never misinterpreted by an experienced scanner. If a surface layer of emulsion about a micron in thickness can be sacrificed, these surface blemishes like the blackening produced by light can be removed by wiping the surface with chamois skin impregnated with methyl or ethyl alcohol. The Eastman Kodak Company, if it is desired, will coat the surface of the emulsion with a thin layer of inert gelatin. Because of the damage caused by scratches and fingerprints, it is advised to handle pellicles carefully by the edges, and to lay them only on smooth, dust-free surfaces.

While many laboratories provide refrigerated, pre-exposure storage for emulsion, the writer has seen no clear evidence that low temperature storage prior to exposure is necessary or even better than storage at 15° - 20° C. The fog level is raised when the temperature is high, however. Ilford Ltd. recommend storage at 10° C and 50 % R.H. as "ideal." Hypersensitized emulsion must be refrigerated.

Storage at dry ice temperature has been proposed (D 49) to reduce the accumulation of background tracks, as it was thought that the emulsion sensitivity falls at low temperatures. The sensitivity of G.5 emulsion now, however, has been observed not to fall much as the temperature is reduced to this point.

Another technique was investigated by Samoylovich *et al.* (SBA 62). They lowered the pH of NIKFI type R emulsion so that it became very insensitive. Below pH 2 an irreversible loss of sensitivity was experienced, but between pH 2 and pH 3 the emulsion could be kept in an insensitive condition, and subsequently the sensitivity could be brought back by raising the pH to 7.

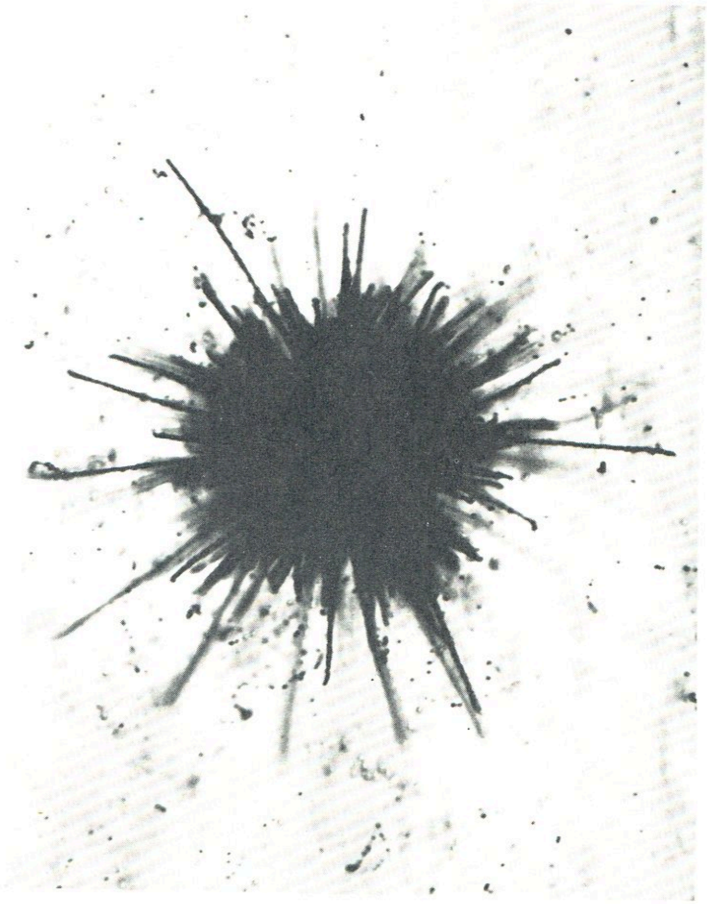

FIG. 4.1.1. Alpha particle tracks emerging from an occlusion found in K.5 Ilford emulsion. (Photomicrograph by C. Cole.)

Veprik *et al.* (V-C 62) arrested the sensitivity of emulsions carried in an earth satellite at the end of the optimum exposure interval. Later when the processing was completed on earth, tracks of singly charged, relativistic particles were visible.

Mere storage of G.5 emulsion hardly affects its sensitivity. A reduction of sensitivity, as measured by the blob density, of less than 5 %

after storing 450 days was observed by Barron and Wolfendale (BW 57). The emulsions of Demers increase in sensitivity over a period of months after manufacture. Further ripening probably occurs.

Emulsion as received from Ilford Ltd. is in equilibrium with a relative humidity of 55-60 %. If stored at this relative humidity, even open to the air, the emulsion will not lose or gain weight, and will remain close to the density 3.815 gm/ml for which the best information on the range-energy relation is available. If the humidity goes higher, the emulsion pellicles lose mechanical strength and are likely to stick together. At a lower relative humidity, they curl and become stiff.

Ilford Ltd. strongly recommend storing gel emulsion at just over 0° C, at which temperature it is expected to keep in good condition for about a month. Shipment of such emulsion must be rapid and it may be advisable to provide refrigeration en route.

4.2 Erasing of Unwanted Tracks

One can imagine a process that erases the latent images of old tracks in emulsion so that an exposure can start with a completely clean detector, and *a priori* this prospect can be contemplated with some enthusiasm. The ideal can be approached under some circumstances, but practical limitations exist, so that such a process of eradication is infrequently applied to thick emulsions, and even thin emulsions usually are treated only for problems requiring special freedom from background tracks. The following are some of the reasons why eradication of emulsion is employed less than might be anticipated:

(*a*) Emulsion gel can be procured so that when required a layer of emulsion can be poured that is completely free of tracks.

(*b*) Emulsion can be shipped and stored so that it is still in good condition for a reasonable time after manufacture.

(*c*) The eradication process is not completely controllable, or perhaps not all batches of emulsion are the same. There seem to be unknown variables which sometimes produce severe loss of sensitivity of the eradicated plate, cause failure of eradication, or perhaps leave a high single grain background in the eradicated plate.

(*d*) Eradication is tricky and requires close attention and good equipment. Therefore, it is regarded as a considerable nuisance.

Despite these less than optimistic introductory statements, we shall describe an eradication process which not infrequently has been found useful.

Many agents will destroy the latent image, but the problem is to weaken it only as much as necessary. Too violent treatment may cause the sensitivity to be lost for subsequent irradiation. Bathing the emulsion in weak chromic acid solution is effective in eradicating the latent image, but in the concentrations tested, too much sensitivity is lost for the method to be recommended. Yagoda and Kaplan (YK 47) have used hydrogen peroxide (which under some conditions of pH may blacken photographic emulsion) and have succeeded in eradicating alpha-particle tracks. Much of the sensitivity was restored after drying. A gentler treatment usually is better, however, and the combined elevated temperature-water vapor method also developed by Yagoda and Kaplan is now generally used.

It is a fact of experience that tracks of highly ionizing particles such as alpha particles probably cannot be eradicated from G.5 emulsion without damaging its sensitivity. Even in finer grain emulsions Wiener and Yagoda (WY 50) found it necessary to employ critical development, in addition to eradication, to obtain complete discrimination against old alpha tracks. The degree of eradication suffered by tracks of weakly ionizing particles is greater than that experienced by the tracks of intensely ionizing particles, so generally the aim is merely to erase the electron tracks, leaving perhaps a clump of grains where each electron stopped. Of course, if the emulsion had a heavy exposure to electrons, considerable non track background will remain in the emulsion.

An eradication procedure for thin layers of G.5 emulsion mounted on glass may be relatively simple. One requires a water tight vessel of stainless steel, glass, porcelain, or other material which does not react chemically with emulsion. In this the plates are mounted in a horizontal position. The vessel must be suitably weighted or secured so that it can be immersed in a water bath. In the vessel with the plates, a little water is placed—enough to permit the emulsion to come to equilibrium with a saturated atmosphere. No other material with an appreciable vapor pressure should be present. The water bath is stirred to prevent temperature gradients, and is heated to a temperature of 35° to 50°C or more. A conservative figure of 37°C produces satisfactory eradication of 200 μ emulsion in about 3 days. At higher temperatures the emulsion becomes quite liquid, and it is necessary that the plates be absolutely level and the emulsion not be too thick so that its surface tension will keep it on the plate. Albouy and Faraggi (AF 51, AF 51.1) have reported progressively more efficient eradication as the temperature was raised up to 95°C, but such high temperatures demand good technique in handling the plates.

If there are any temperature gradients, water will condense on the cooler areas of the surface inside the containing vessel and may drip on the emulsion. This must be guarded against. The plates are dried fairly rapidly before exposure. Using the procedure described above, the writer and his collaborators (BDGV 52) have removed all electron tracks from 200 μ G.5 emulsion and retained sufficient sensitivity to record 41 grains per 100 μ in the tracks of electrons on the plateau of ionization. Apparatus used for the eradication of electron tracks from emulsion used in a beta-ray spectrometer in the Lawrence Radiation Laboratory is shown in Fig. 4.2.1.

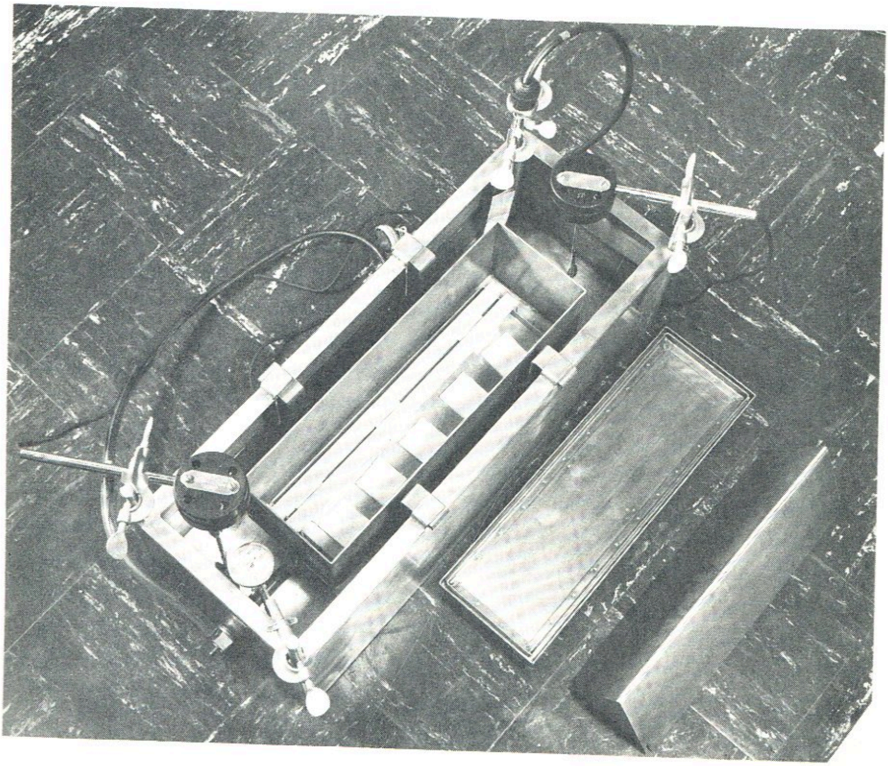

FIG. 4.2.1. Water vapor thermostat for emulsion eradication. The water bath is stirred and maintained at a constant temperature. The inner tank also contains a little water. The plates are supported above the liquid level. The "roof" shown is placed above the plates to prevent dripping on them, and a light-tight cover maintains the space within at a relative humidity of 100% (IDLRL).

Eradication in thick layers of unmounted emulsion is generally more difficult and less complete. Stiller, Shapiro, and O'Dell (SSO 54) have succeeded in eradicating layers of G.5 emulsion 400 μ in thickness, and have shown that when the emulsion was subsequently exposed, the procedure had reduced the track distortion. This reduction is what one would expect if strains that are relieved by the eradication exist in the emulsion.

Eradication of 600 μ pellicles has been carried out by Birge *et al.* (B-W 54) at a temperature of 33°-36°C for 5 to 7 days, but they find a reduction of the grain count of minimum tracks to half the value it has in uneradicated emulsion.

Lord *et al.* (LBCG 62) have reinvestigated the eradication of thick emulsions using both chromic acid solutions and high humidity at temperatures near the emulsion melting point. Good results were obtained with emulsions as thick as 600 μ.

Many emulsions, especially those with small grains, are easily eradicated. In such emulsions the latent image has low natural stability. In some fine grain emulsions the buildup of background in old plates is not usually a problem because the older tracks fade as rapidly as new ones are produced. Very simple eradication procedures suffice for accelerating the fading of such emulsions. The emulsions of Demers, and the NIKFI type T-1 emulsions have been observed to behave in this way.

It is probable that the processes of fading and eradication are really quite complicated, and the present uncertainty of success in controlling the results can be attributed to this. Experiments of Winand and Falla (WF 49, WF 49.1) may be cited to suggest that the bromine released in the photolytic process may itself be instrumental in fading the latent image when conditions are favorable for it to recombine with the silver (rehalogenation).

Controlled fading and discrimination are closely related topics. The emulsion T-1 was developed by NIKFI to provide good discrimination between the tracks of alpha particles and those of protons recoiling from an intense flux of neutrons. A very sharp discrimination was obtained by the use of sodium rhodanide which desensitizes and increases the abruptness of the threshold of response of the photomaterial. The desensitization is brought about by dissolution of the small centers of sensitivity. A 1 % solution of rhodanide is added to the emulsion—some 5.6 to 8.4 ml per 100 ml of emulsion.

The presence of the rhodanide in the emulsion causes very rapid regression of the latent image. Silver of the latent image forms a complex with the rhodanide ions and is lost. The plates can be stored cold for a maximum of 24 hr after exposure. The presence of the rhodanide is of course the reason that T-1 plates are easily eradicated prior to exposure.

For eradication it is recommended that they simply be stored for 3 hr at 25°C in a relative humidity of 100 %.

4.3 Preparation of Plates for Exposure

As many operations as possible should be carried out prior to exposure. Then the emulsion can be developed without delay once the exposure has been made. It is essential that the plates be marked so that they can be identified. If one writes with a sharp, medium-hard pencil on the emulsion surface, the writing can be read on the emulsion surface under a safelight; and after development, the pressure-blackening effect makes the pencil marks clearly readable. Plates are usually marked with small numbers and letters on an unimportant area of the emulsion surface. Glass-backed emulsion also can be marked by writing with a diamond stylus on the glass backing.

If the plates are to be exposed in light, they must be wrapped. The type of black paper used for X-ray film has been found good for wrapping test plates, but since sheets of such paper often contain tiny holes, two layers of paper are strongly recommended. Photographic or cellophane tape are useful for binding the wrapped plate. If the ranges of low energy particles are to be accurately measured, no covering of the emulsion can be tolerated, and darkness during exposure must be provided by other means. When the energy is not too low, an opaque foil window of known stopping power may replace one side of a box in which the plates are mounted. Particles also may be bent in vacuum through curved, internally blackened channels (BBS 56) by a magnetic field, thus excluding light.

Emulsion which is to be used for track measurements requires a known emulsion density and a shrinkage factor that does not vary with depth in the emulsion. It must be conditioned by storage at the relative humidity to be employed in the exposure, or the exposure must be accomplished so quickly that no appreciable change in emulsion water content can take place (see Section 4.7).

4.4 Preparation of an Emulsion Stack for Exposure

The emulsion stack is an invention that has greatly enlarged the scope of research with emulsion (PFP 59, D 52, SSO 54, LPP 53). The stack is made up of many emulsion layers called pellicles. These sheets of emulsion are clamped tightly together so as to eliminate the spaces between layers. It is essentially a block of emulsion. No foreign matter, such as tissue paper, should be used between layers because of the serious errors introduced into range measurements and the added difficulty of tracing particle tracks from one pellicle to the next. Such spacers also introduce errors in decay-lifetime measurements and in interaction free paths.

During assembly of a stack, each pellicle should be examined to detect surface defects by reflecting the safelight from each surface at grazing incidence. Pellicles of normal Ilford emulsion will not adhere to each other at room temperature if the relative humidity with which the emulsion is in equilibrium is below about 60 %. When quantitative measurements are planned, the emulsion to be made up into a stack should be brought to a uniform condition of humidity equilibrium by storing it in a closed container for 2 weeks or more.

For mechanical manipulations of emulsion pellicles some rules should be formulated. Among these are the following:

(*a*) Hold pellicles only by the edges or support on a clean smooth surface. Fingerprints and other grease spots are strongly suspected as sites of blisters on the processed plate.

(*b*) Never slide a pellicle on a hard surface. Tiny grains of dust or points on the surface make developable scratch marks.

(*c*) Never bend a pellicle sharply. Even a radius of curvature of 5 cm or so may cause plastic flow at the surface where the stress is greatest and result in a permanent distortion.

(*d*) Never apply a high local stress, and especially no tangential stresses to emulsion pellicles. Rolling as a means of mounting pellicles on glass is a completely unacceptable procedure if distortions are to be avoided.

(*e*) Always store a pellicle supported over its whole area. If unevenly supported, slow plastic flow of the emulsion will take place and large distortions will result.

(*f*) Carry out mechanical operations on pellicles when they are cold and as dry as they can be consistent with other requirements.

(*g*) When machining operations are carried out on an emulsion block, a sharp high-speed tool should be used and the cutting speed controlled so that the temperature remains low. Adjacent pellicles tend to stick to each other at points where they have been machined. Rather than to pull them apart, a thin sharp instrument should be inserted between them to the depth of a few thousandths of an inch necessary to separate them.

Clamping of the stack without penetrating the emulsion with bolts or rods requires some thought in order to distribute the pressure evenly. If the top and bottom of the clamp are not sufficiently rigid, they will bend, and pressure will be applied only at the sides or ends. The pellicles, too, are often not uniform in thickness, and they may contain waves or be wedge-shaped. To distribute the load over the pellicles, it is wise to place about a 3/16 inch of rubber beneath the lowest pellicle or above the top pellicle. The clamping members may be strong and rigid as illustrated in Fig. 4.4.1, or be especially designed to distribute the load as shown in Fig. 4.4.2. Even with a well-designed clamp for the stack, bumps on the surfaces still tend to keep the pellicles from intimate contact and 2 or 3 μ average air space may be expected. A clamping pressure of 44 lb per square inch has been used and seems satisfactory.

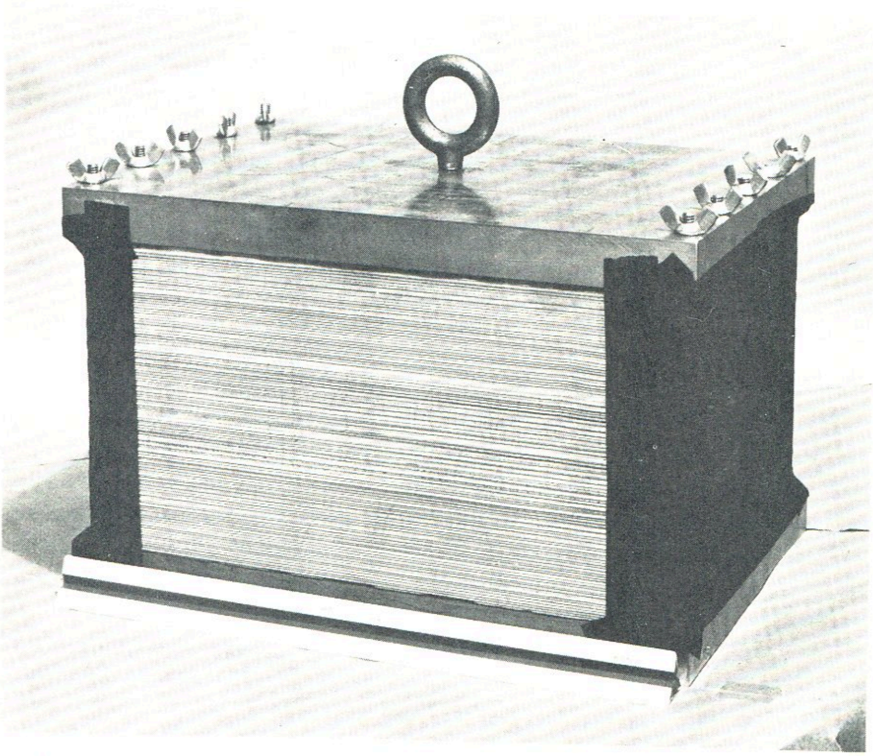

FIG. 4.4.1. An assembled stack consisting of 240 sheets of 9 × 12 inch × 600 μ Ilford G.5 emulsion. The top and bottom clamping members are 1 inch brass plates. The eye bolt provides a means for lifting the large weight of the stack (IDLRL).

The optimum clamping force varies rapidly with the dryness of the emulsion. If a stack is permitted to remain under pressure for a day or more after clamping, it will be found that the pellicles have loosened in the stack, and that the emulsion has flowed slightly in the lateral directions. On relieving the pressure completely, the emulsion tends to return to its original dimensions with a relaxation time of many hours.

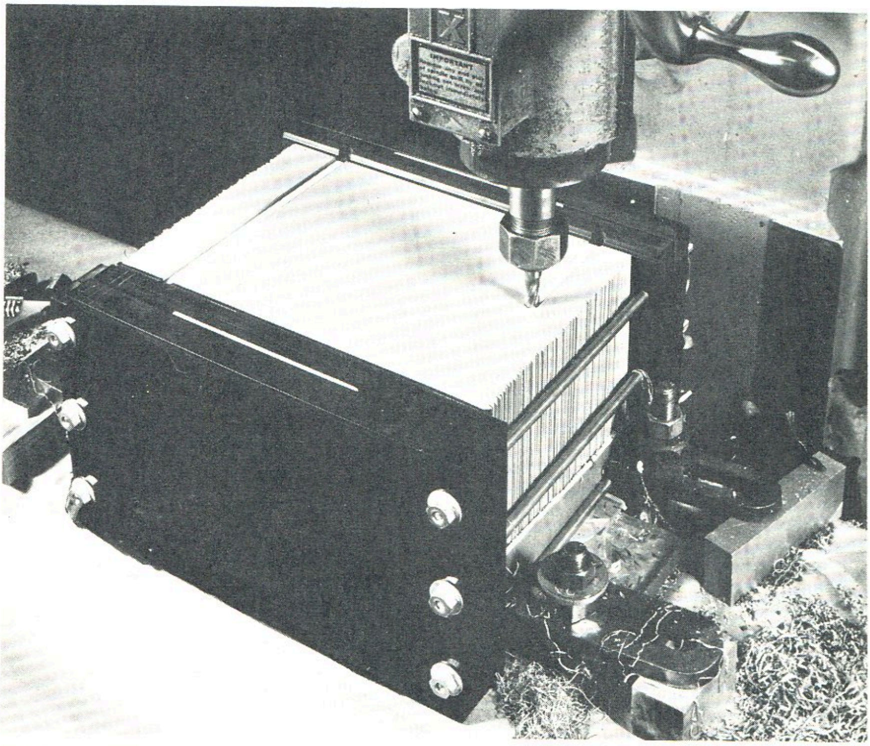

FIG. 4.4.2. Milling of an emulsion stack. The face is machined flat and notches are cut for preliminary alignment of the pellicles. Both the top and bottom clamping members are double with a third thin sheet between them in the middle portions. This design distributes the load over the pellicle surfaces when the clamping force is applied at the ends of the outer members (IDLRL).

The dimensional changes usually are not serious ($< 0.3\ \%$) unless excessive clamping forces are employed or the emulsion is quite soft.

In order to trace tracks through the stack, one needs a system by which the points on the top surface of each pellicle can be put into a one-to-one correspondence with the points on the bottom of the pellicle above and adjacent to it. Several systems of alignment that are reasonably convenient for small pellicles are in use, but a really satisfactory system for large pellicles is hard to devise. The most fundamental difficulty is that the emulsion pellicle is insufficiently rigid for two points in a large pellicle to define the positions of all the rest with sufficient accuracy. Another difficulty, which is quite general, is that the points on one surface of a sheet do not determine the positions of those on the other surface. Shear-type distortion is quite common after the pellicle is mounted on glass and processed.

An important requirement for working rapidly in track tracing is that, when a plate is taken off the microscope stage, the next one can be easily inserted into the same position (within a few microns). The track being traced should be found in the field of view in a predictable position and unaltered in direction. The location of an event in a pellicle also must be describable in a way that is independent of any particular microscope. This need is generally met by photographing a coordinate grid directly on the surface of the pellicle. Depending on the alignment system employed, this may be done while the stack is being prepared for exposure or upon disassembly after exposure.

A simple, if rather primitive, alignment system was used successfully by the writer in early experiments with stacks of a few pellicles. It was simply to drill two small holes through the emulsion stack, using a template for the hole positioning. The holes were then reamed slightly with a template to a diameter that was known to 10^{-4} inch. Identical holes were also drilled by means of the template in a flat, heavy brass plate. In the holes close-fitting dowel pins were fitted which, when passed through the emulsion holes, held the pellicle in a precise position on the brass plate. A glass-backed photographic negative of a grid known as the Berkeley grid, comprising numbered millimeter squares, was prepared by Mr. Philip Carnahan. This was placed on top of the pellicle in a reproducible position and located by mechanical stops. The grid was then contact-printed on the shiny surface of each pellicle (the surface which was later to be placed against the glass in mounting). The optical image of the grid develops simultaneously with the tracks in the emulsion. The grid printed on the processed pellicles provided a convenient x and y coordinate frame in the stack. Unfortunately, thick stacks were found difficult to drill. For large stacks, H. H. Heckman and Conrad Mason improved the procedure as follows: instead of drilling through the stack, two notches are unsymmetrically milled on the side of the stack, cutting into every pellicle as shown in Fig. 4.4.2. Under the microscope, the corners of the notch are somewhat rounded, but the crossing of the extensions of two adjacent sides of the notch define a point in the plane of the emulsion sheet. By translating the pellicle until this point coincides with a fixed point on the negative of a grid beneath the pellicle, and subsequently rotating the pellicle in its own plane around the point until a point of the other notch comes to a prescribed position on the grid, the pellicle can be brought into an accurately defined position on the grid. The adjustment for coincidence of the points is carried out under microscopes using red light that does not fog the emulsion.

The Berkeley grid is used extensively, and has been of great value, but some improvements are desirable. It is not quite accurate enough for range measurements to be based on it; the numbers are too large to read well under high magnification, and it is said to intercept too large a fraction of the light incident on it. A number of other grids perhaps superior to it in some respect have been made, but none give promise of superseding all others. The Chicago grid, for example, obscures only 1/2 % of the bottom surface of the emulsion (SS 56) but may be criticized for being too abbreviated. A small portion of the Brussels grid is shown in Fig. 4.4.3 along with the Berkeley grid at the same magnification.

For rapid track tracing the grid should be supplemented by other devices. Under the microscope, time is wasted finding the grid square containing the track, and some grid shift always exists. Microscope stage accessories must therefore also be devised. Many research groups working with large emulsion stacks have developed their own systems of alignment, a few of which may be mentioned.

A common scheme utilizes X-ray markings in the edge of the stack. This marking may be done by the manufacturer or in the laboratory by setting up an X-ray tube of 50-150 Kev, and a narrow slit in a heavy element shield. The X-ray can be passed between two heavy-element cylinders in contact (P 53). The X-rays make a blackened line running into the stack for perhaps 1/4 inch, if the stack is placed behind the slit. Several such lines can be put on the edge of the stack. The lines are used to position the glass plate on which the pellicle is mounted in a glass-cutting machine. The glass is then cut and its edges used to position the plate on a microscope stage. By this means the plates are located to perhaps $\pm$ 200 μ. Birge *et al.* (B-W 54) have improved this to $\pm$ 15 μ by cementing brass tabs on the edge of the glass and filing the brass for precise positioning. They also used a grid, but it is located only approximately with respect to the pellicle. Their pellicles are mounted on glass plates coated with a 10 μ layer of emulsion on which the grid previously has been printed.

Vanderhaeghe (V 54) cements a thin sheet of emulsion on which a grid is printed on top of the pellicle. Goldhaber *et al.* (GGL 55) punch holes in the emulsion and stack the emulsion on pins fitting the holes accurately. The grid is then printed on the emulsion using the same holes for alignment. The main disadvantages of this method are that

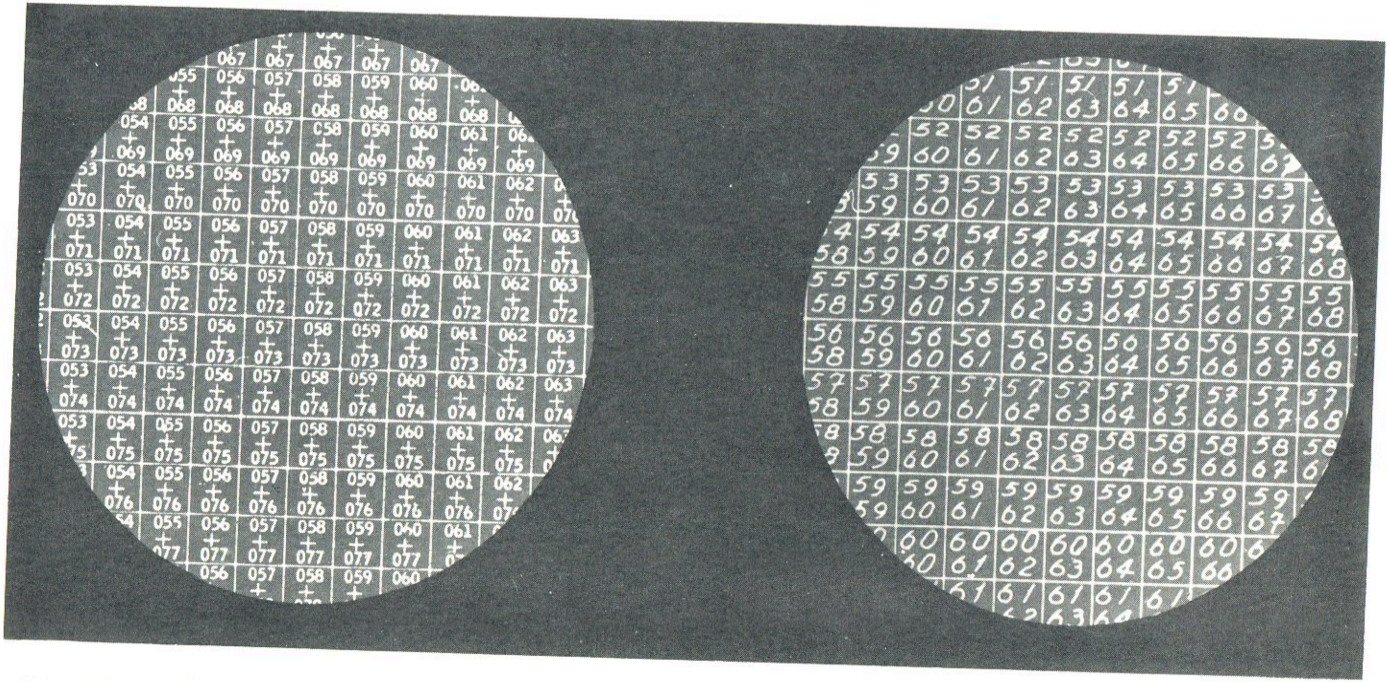

FIG. 4.4.3. Small portions of the Berkeley grid (three-digit numbers) and the Brussels grid (two-digit numbers). The side of a grid square is 1 mm. (Photomicrograph by C. Cole.)

holes remain in possibly useful parts of the stack, and a local thickening of the emulsion occurs where the holes are punched.

The press with which the holes are punched also makes four pin pricks in the pellicle. When the pellicle is developed these are seen as blackened areas about 100 μ in diameter. By means of four low power microscopes these marks are used in cementing corner brackets of brass on the plates. The brackets are used to hold each plate on the microscope stage in accurately the same position.

A method once used at Columbia University is to place thin nylon threads between the pellicles when the stack is assembled. They cross at two or more well-separated points. When the stack is clamped, the pressure at the points where the threads cross is sufficient so that pressure marks develop on the emulsion surfaces—thus directly relating the top surface of each pellicle to the bottom of the pellicle above it. Another form of this technique is to use thread soaked in a solution of an alpha-particle-emitting isotope. The rows of alpha-particle tracks then align the top of one pellicle with the bottom of the one above it.

A further feature of the Columbia system is the use of an iron strip cemented to the edge of each plate in the same position relative to each pellicle. The strips have V-shaped notches machined in them. An alnico magnet on the microscope stage is fabricated to fit into a notch and hold each successive pellicle in the same relative position.

Often stacks are aligned by mounting each plate in a light metal frame. The frame is held by pins or clamps on the microscope stage and the position of the plate in the frame is adjustable by set screws or otherwise. In another system the glass plates are cemented to underplates of transparent plastic which have been machined or drilled to fit on the microscope stage in certain positions. This alternative and that of the metal frame may raise the emulsion above the condenser lens so far that a good optical image of the tracks in the emulsion is unobtainable under high magnification.

Samman and Vanderhaeghe (SV 59) make up stacks in which between each pair of pellicles, U and L, a thin film is inserted. A negative grid is printed on it. During assembly of the stack an exposure to light through the grid is given to each lower pellicle, L. On disassembly, the stack is turned over. Then, as each new grid is exposed, an exposure to light through the grid is given the pellicle U, which was originally uppermost. In this way adjacent interfaces are put in accurate registration. The dead volume between pellicles, the danger of moving the grid between exposures of pellicle L and U, and the presence of two light-absorbing grids on each pellicle are the chief objections to this method.

The most precise means for alignment are steep heavily ionizing tracks that pass from one pellicle to the next. By making careful measurements with respect to such tracks one has a completely reliable method of tracing difficult tracks through from one pellicle to the next. A track which does not pass continuously from pellicle to pellicle may be rejected as background which was produced in the emulsion at a time when the stack was not in its final assembled condition. (It is a good idea to shuffle the pellicles and assemble the stack just prior to exposure for this reason.)

A complete system of alignment that the writer currently employs and can recommend is based on the combined use of steep tracks passing from one pellicle into the next, and a grid printed on the emulsion surface. The grid is printed on the pellicles, using the notches milled in the edge of the stack as illustrated in Fig. 4.4.2.

The pellicles are mounted on glass plates leaving about a 1/2 inch margin at the edges. After processing they are cemented on a steel frame, the notches in the pellicles being adjusted to fixed positions relative to the frame with the aid of a jig. (Alternatively a steel strip, B, is cemented to the glass. It contains bolt holes that are in definite positions relative to the milled notches.) The accuracy required in the cementing operation is not high—a few hundred microns.

A strip, A, of steel is bolted to the frame (or to the strip B) through oversize holes, so that adjustment of the position of strip A with respect to the frame is possible. In this strip holes are drilled and reamed, accurate in position and diameter to about a micron. Drill-rod dowl pins pass through the holes into jig-bored holes in the microscope stage. Strip A is held with respect to the microscope stage to within a few microns. By adjusting the position of strip A with respect to the frame the pellicle can be placed precisely on the stage. When the adjustment is optimum, the bolts holding strip A to the frame are tightened.

The adjustment is made on an instrument designed for this purpose (B 58.2). It is a type of jig that utilizes two microscopes. In this the exit points of tracks emerging from the surface of a pellicle are matched to the entrance points of the same tracks in the bottom of the pellicle that was above it in the stack. The adjustment process is started with the first plate of the stack and carried through the entire stack. In this way each pellicle is aligned with the one below it.

It is most convenient if strip B or the frame is initially cemented on with sufficient accuracy so that the continuation of a track can be found in the same field of view when an unaligned plate is put on the microscope stage.

The sequence of emulsion sheets in a stack should be established by numbering the pellicles. This is done best while the stack is being assembled. It is sufficient merely to write the numbers on the emulsion surface as described in Section 4.3, pressing the pencil firmly so that a pressure image will develop. As the glossy side of the pellicle must be against the glass when the pellicle is mounted, it is best to put the number on the dull surface of the pellicle.

Since the surface fogging of the pellicles increases with time under even a good safelamp, it is wise to keep the emulsion covered in the darkroom except when work is actually being performed on it. The emulsion should also be kept sealed as much as possible to reduce the rate at which water is lost or gained when the disassembled stack must be left for a period. A simple expedient is to sandwich pellicles between glass or plastic sheets and tape them together around the edges. If practical, the relative humidity in rooms where emulsion, either unprocessed or processed, is stored should be kept close to 60 %.

If there is any tendency for the pellicles to stick together, there is danger of distorting them upon disassembling the stack. Rather than attempting to pry them apart, Rosenfeld *et al.* (R 55) recommend freezing the stack. This not only makes the individual pellicles stiff and strong but they tend to come apart spontaneously. When it is not practical to freeze the emulsion, a moderate, steady, and long sustained force will often be successful in separating emulsion from another surface to which it adheres. For example, two plates stuck face to face can sometimes be separated simply by inserting a wedge between them, and gradually over a period of hours, pushing it in farther. Unfortunately the distortion resulting from this operation is usually serious. Only the characteristic of gelatin to remember its original form and to tend to return to it, helps to undo the injury to emulsion.

An emulsion chamber is any of a variety of combinations of emulsion interleaved with other materials that is used to analyze radiations. For example, layers of a heavy element can be placed between emulsion pellicles or plates as an analyzer of high energy photons. The principles of emulsion handling that should be observed in these special applications are much the same as those applicable to stacked emulsion. Items of equipment used for storage and manipulation of emulsion at the Lawrence Radiation Laboratory are illustrated in references (B 58.2) and (B 60.1).

4.5 Pouring Emulsion Received in Gel Form

Emulsion in gel form is sold by Ilford, Ltd. for applications in which no old background tracks may be tolerated in the emulsion, or when the emulsion must make intimate contact with the source of radiation. It has many possible applications in biological research, in geology, in chemistry, as well as in physics.

Because pouring of the emulsion involves additional critical operations, it is recommended that it not be undertaken unless it provides a considerable advantage over plates or pellicles.

In one well-known experiment by George and Evans (GE 50) the manufacturer transferred part of the factory operation to the site of an underground experiment, and poured the emulsion on the spot.

If reasonable care is taken, satisfactory pouring results are usual. Some of the pouring procedures adopted at the University of Chicago (R 55) with the advice of Mr. C. Waller of Ilford, Ltd. will be described.

Until needed, the emulsion gel, which comes in 50 or 100 ml brown bottles, is stored at a temperature just above 0°C. Stored thus, it will keep for about a month. Even in the bottle it should not be exposed to any light except that from a safelight of the type employed for noncolor sensitive photographic materials, and all illumination should, of course, be minimized. Ilford, Ltd., recommend type "S" safelight No. 902, but others can be used if tested first.

For pouring, the gel is heated to a temperature between 45° and 50°C. To make a thin layer, the higher temperature is required, and the glass backing is preferably preheated. A glass rod may be employed to help in obtaining uniform coverage of the glass plate. It is important to avoid keeping the emulsion warm longer than necessary—not longer than 1 hr at 50°C, or 2 to 3 hr at 40°C. The effects of reheatings on the gel also are probably cumulative.

The volume of the gel is about 8 times that of the dried emulsion, and its specific gravity is 1.37. One calculates the desired volume or weight of gel to be transferred by spatula to a large beaker which has sufficient bottom area so that the gel can heat quickly when placed in an oven. With the oven at 60-65°C it is found that the desired temperature is attained in 20 min.

The emulsion should be stirred, but bubbles must be avoided. While heating the emulsion all the glassware to be used in handling it should also be heated. A pipette is useful for measuring and pouring small emulsion volumes, but the discharge hole must be enlarged. Otherwise, a small calibrated beaker may be used to measure out the correct volume of emulsion and to pour it.

A thick glass plate, level to 0.04° or better, and with a clean, flat, smooth surface is employed for pouring. Pellicles are poured directly on the glass surface, but for thick layers dams confining the liquid gel to the desired pellicle dimensions are required. The dams can be made of glass or lucite rods held in place by gelatin or paraffin used as a glue. Emulsion plates are poured on special Ilford glass prepared for coating which has been laid out on the large, level glass plate. This plate should be oiled to prevent adherence of glass to glass, if some emulsion should leak between the plates.

Before pouring, bubbles should be removed from the gel with a glass rod, and the gel can be drawn from the bottom by means of a pipette. In addition, after pouring, the liquid surface should be examined for bubbles which can be pushed to the dam with a glass rod.

The emulsion is dried in darkness with clean warm air or nitrogen circulating over it. It should remain in the horizontal plane. A temperature of 80°F is suggested. The drying should not be so rapid, however, that a "skin" forms on the emulsion surface. The reason for this is a somewhat paradoxical one; namely that too rapid drying results in a great retardation of the drying rate. Such an effect is explained by pointing out that if a "skin" of dry emulsion forms at the surface, the diffusivity in this layer is so low that the water from the semiliquid emulsion below can penetrate it only very slowly. Usually in 24-28 hr 600 μ emulsion will have dried enough to come off the glass easily. and the damp underside of the pellicle will then dry off rather quickly.

Usually a special box is constructed for the drying operation. The type of box used at the University of Strasbourg is shown in Fig. 4.5.1.

The use of liquid emulsion has also been developed in the Soviet Union. In order to obtain good adherence of the emulsion to the surface on which it is to be poured, or to prevent reactions between the base material and the emulsion, Bogomolov *et al.* (BDSU 57) recommend a special undercoating. This consists of 90 ml of water, 0.33 gm of gelatin, 2.5 ml of a 3 % solution of chrome alum, and 10 ml of a 3 % solution of thymol. The gelatin is melted in the water at 35°C. The alum is then added slowly with mixing. Subsequently the thymol is added in the same way, and the solution is filtered. It is prepared in small quantities because the solution will not keep.

In the event that the speciman on which emulsion is to be poured contains components such as active metals or oxidizing agents that react with the emulsion, it is necessary to coat it with this undercoat to a thickness of 1 or 2 μ. It is applied at a temperature of 25°-27°C preferably by immersion. The surface coated with emulsion should be dried in a vertical position.

NIKFI emulsion gel is prepared for coating by the addition of a chromium acetate toughening agent (BDSU 57). The emulsion is applied by bathing the experimental surface with the diluted emulsion at a temperature of 30°-35°C. Repeated immersions are made to build up the thickness of the layer. The liquid emulsion gels in 20-35 min after which the coated object is placed in a drying cabinet. In the cabinet filtered air moves about 0.5 meters per second and is at a temperature of 18°-20°C.

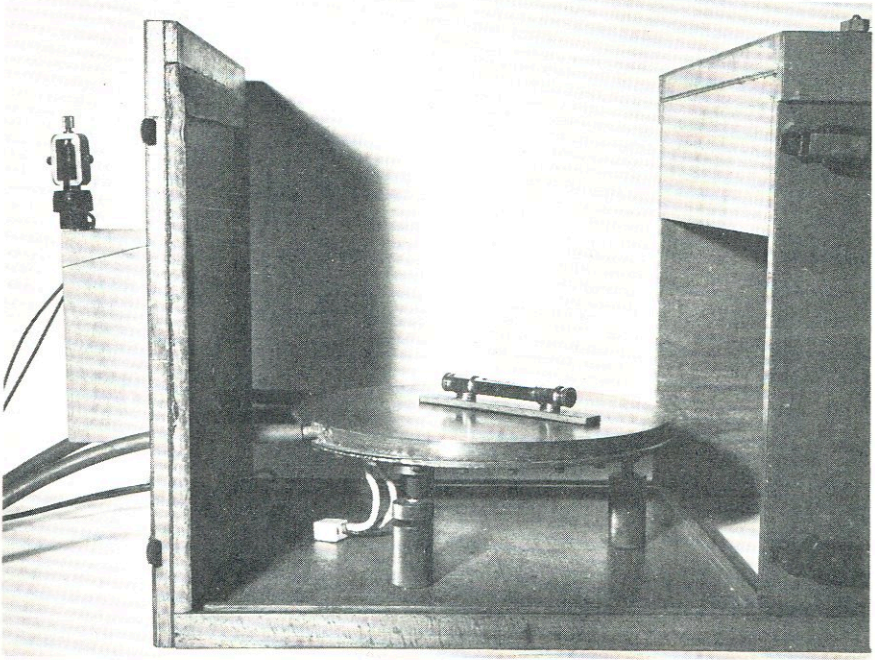

FIG. 4.5.1. A drying box developed at Strasbourg. The smooth stainless-steel plate is accurately level. Its temperature is controlled by water flow through it. The box on the right rolls into position to exclude light and dust. It is partitioned; the upper part can contain ice or dry ice. The bottom of this portion is sloped to prevent dripping on the emulsion. A flow of filtered gas such as nitrogen can be maintained in such a box, and its water content can also be controlled. (Courtesy R. Rechenmann.)

It is sometimes desired to prepare very thin (approximately 10 μ) emulsion films free of support. Dr. C. Mason at the Lawrence Radiation Laboratory prepared such films. His method is summarized as follows: if a film of area A and thickness t was to be made, a solution was prepared consisting of emulsion in gel form (volume $10A't$), glycerin (volume $A't/4$), and water (volume $20A't$). The solution was raised to a temperature somewhat above 60°C using glassware warmed to the same temperature. It was poured on a glass plate of area A' surrounded by a dam of tape. The area A' differed from A because of wastage at the edges and spoiling of the film where it was grasped in stripping it from the glass after drying. Mason found that A = length, l, times width, w, was related to A' by the relation $A' = (l + 4 \text{ cm})(w + 1 \text{ cm})$.

The glass plate on which he poured the gelatin-halide solution was prepared by cleaning it carefully with water and alcohol. This was followed by rubbing a thin film of immersion oil into the surface. Any oil adhering to the film was removed at a later stage with xylene.

The glass plate rested on a flat brass table, the temperature of which was adjustable, and which was provided with leveling screws. It was leveled so that the angle of inclination was less than 10^{-4} radians. The table was hollow to permit flow of water through it, thus controlling its temperature. Froth and bubbles were avoided in pouring.

After pouring, the gel was spread with a piece of glass or a spatula while maintaining the temperature somewhat above 60°C. After inequalities in thickness disappeared the emulsion was chilled so that it jelled. It was dried in a flow of air.

To remove the film from the glass Mason cut away 1/2 cm at the edges. Then cellophane tape—with a length equal to the film width—was attached across the end of the film. The tape was also attached to a cylinder of approximately 1 inch diameter. The film was rolled on the cylinder with a layer of paper under it. Then, while gently preventing the assembly from unrolling, it was immersed in xylene. The xylene dissolved the cellophane tape adhesive and left the film dry and easily handled on the paper. (Note that xylene is toxic.)

4.6 Humidity Measurement and Control

The properties of gelatin vary considerably with the ambient relative humidity, which should stay within a limited range for emulsion work. The measurement and control of the R.H. are therefore of some importance to the emulsion worker. There are many methods of estimating the R.H.: from observation of the dew point on a mirror surface, from the wet-bulb thermometer, from calibrated mechanical properties of moisture-sensitive hairs, foils, etc. The best for a control system is a stable humidity-sensitive electrical property that can be calibrated and employed to control an electrical current.

A complete air conditioning system installed in the scanning and plate storage rooms to maintain the temperature and humidity at constant nominal values is desirable for emulsion work, but it is expensive and not a strict requirement in many localities. It is less expensive just to install air humidifiers that keep the air from becoming too dry in cold weather or in dry climates. In some places such equipment is necessary. Merely to store plates, one can simply maintain the air in a limited volume at a known constant relative humidity. The simplest and most flexible method for doing this is to keep the air in such a closed space in contact with a water solution of glycerol of sufficient volume so that the water content of the air in the enclosure is small compared to the amount of water in the glycerol solution. The glycerol concentration to bring the air to various equilibrium relative humidities is given in Table 4.6.1.

TABLE 4.6.1

RELATIVE HUMIDITY OBTAINED FROM WATER-GLYCERIN MIXTURES AT 25°C

Per cent R. H.	Per cent glycerin by weight	Specific gravity
10	95	1.245
20	92.5	1.237
30	89	1.229
40	84.5	1.216
50	79	1.203
60	72	1.184
70	64	1.162
80	51	1.127
90	33	1.079

Another frequently used method is to maintain the humidity by establishing an equilibrium between the water content of the air and a saturated solution with an excess of solid of a suitable salt. Table 4.6.2 lists a number of salts that have been employed, and the relative humidity that each maintains. The water content of emulsion at a constant relative

TABLE 4.6.2

CONTROL OF HUMIDITY BY SATURATED SALT SOLUTIONS

Salt	2°C	10°C	15°C	20°C	25°C
H_2O	100	100	100	100	100
KCl	88	88	87	86	84.5–89
NaCl	75	77	77.5	77	75
$MgCl_2 \cdot 6H_2O$	35	34	34	34	33
Fresh P_2O_5	0	0	0	0	0
$ZnCl \cdot H_2O$	10	10	10	10	10
$NaBr \cdot 2H_2O$	—	—	—	58	57

humidity, but widely varying temperature, has not been fully studied. Oliver (O 53) found, using a solution of $Mg(NO_3)_2 \cdot 6H^2O$ to stabilize the R.H., that C.2 emulsion density increased 1 % when the temperature was changed by 23°C while Leide (L 56) observed that a temperature increase of 10°C at 75-50 % R.H. causes a loss in G.5 emulsion weight equivalent to a decrease in relative humidity of 2-4 %. Leide mentions excellent agreement between his measurements and those of Oliver.

4.7 Theory of Diffusion in Emulsion

Nearly all the emulsion processing operations are concerned with the diffusion of water solutions into or out of the emulsion. At least an approximate treatment of the problem of water diffusion is therefore demanded in order that one properly understand and control operations that depend on it. The general problem is mathematically very complicated because heat of reaction between gelatin and water exists, so it is not really a simple diffusion problem of one medium penetrating another without interaction. Furthermore, it is not possible, in general, to define a constant of diffusion, because an increase of water concentration in the emulsion greatly increases the ease with which solutions can diffuse through it, and a change in pH changes the equilibrium swelling suffered by the emulsion. Glycerin, which is often present in the emulsion, and chemically induced changes in the gelatin, also complicate the behavior of the emulsion-water system.

Because of these difficult and/or unanalyzed effects, we first follow the writer's earlier treatment (O 53) of the theoretical problem in limiting the mathematical analysis to low water concentrations. Then the volume changes are negligible and a constant of diffusion may be assumed.

Let the glass-emulsion interface of an emulsion plate be the x, y plane. Then we take the free surface of the emulsion to be the plane $z = z_0$. The diffusion of water or a soluble material in water through the emulsion is described by the partial differential equation

$$\frac{\partial c}{\partial t} = K \frac{\partial^2 c}{\partial z^2} \tag{4.7.1}$$

Here $c = c(z, t)$ is the concentration of the water or dissolved material, z is the coordinate of the layer of emulsion in which the concentration is c, t is the time, and K is the diffusivity.

Suppose the initial concentration of the material in the emulsion is c_0 and the concentration at the surface is suddenly changed to, and remains at, c_1. The solution of Eq. (4.7.1) subject to this boundary condition is found by the method of the Laplace transform:

$$\frac{c - c_1}{c_0 - c_1} = 1 - \frac{2}{\pi^{1/2}} \sum_{n=0}^{\infty} (-1)^n \cdot \left[\int_{u_n - v}^{\infty} e^{-x^2}\, dx + \int_{u_n + v}^{\infty} e^{-x^2}\, dx \right] \qquad (4.7.2)$$

where $u_n = [(2n + 1)/2] \ (z_0^2/Kt)^{1/2}$ and $v = (1/2) \ (z_0^2/Kt)^{1/2} \ (z/z_0)$. In units of the distance z_0 and a characteristic time, $T = z_0^2/K$, this expression is universal.

Equation (4.7.2) is the general expression for the concentration, c, as a function of time and position. Then

$$\phi = K \partial c / \partial z \,|_{z=z_0}$$

is the flux of the material into the emulsion. From Eq. (4.7.2) this can be written:

$$\phi = (c_1 - c_0) \left(\frac{K}{\pi t} \right)^{1/2} \left[1 + \sum_{n=1}^{\infty} (-1)^n \, e^{-n^2 T/t} \right] \qquad (4.7.3)$$

In Fig. 4.7.1 the behavior of the water concentration in emulsion that has suddenly been plunged into vacuum is graphed.

For t small compared to the characteristic time, T,

$$\phi = (c_1 - c_0) \left(\frac{K}{\pi t} \right)^{1/2}$$

so that the initial gain or loss of the diffusing material obeys the law:

$$w = \int_0^t \phi \, dt \approx 2(c_1 - c_0) \left(\frac{Kt}{\pi} \right)^{1/2}$$

The weight varies with the square root of the time. The relationship has been observed to hold for the change in weight accompanying loss or gain of water in pellicles suddenly subjected to a change in ambient relative humidity.

Oliver (O 53) measured the weight of a plate of G.5 emulsion, initially at 50 % R.H. and 884 μ in thickness, as a function of the time after it was placed in an evacuated chamber. The observed loss is compared in Table 4.7.1 with that predicted from Eq. (4.7.2).

TABLE 4.7.1
CALCULATED AND OBSERVED WEIGHT LOSSES[a]

t (days)	Observed loss (gm)	Calculated loss (gm)
0.0417	0.0055	0.0059
0.0833	0.0081	0.0083
0.1667	0.0117	0.0118
0.941	0.0280	0.0280
2.94	0.0500	0.0490
8.96	0.0743	0.0770
17.96	0.0878	0.0864
27.95	0.0962[b]	0.0880

[a] Weight loss calculated from:

$$W = 0.100 \int_0^t \frac{\left[1/2 + \sum_{n=1}^{\infty} (-1)^n \exp(-12\, n^2/x)\right]}{(x/12)^{1/2}} (dx/12)$$

[b] It is believed that this number is erroneous. Some chips of splintered glass were unaccounted for in the weighing.

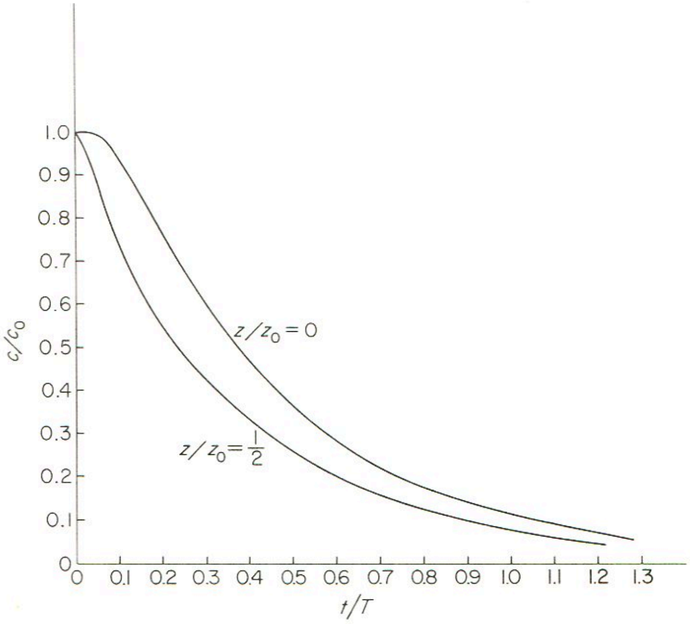

FIG. 4.7.1. The ratio c/c_0 as a function of t/T. The curve designated $z/z_0 = 0$ corresponds to the emulsion-glass interface, and the one labeled $z/z_0 = \frac{1}{2}$ corresponds to a point halfway through the emulsion layer. At $t = 0$ the plate was suddenly plunged into a vacuum ($c_1 = 0$).

The agreement is seen to be good if one takes the characteristic diffusion time to be $T = z_0^2/6.5$ days, where z_0 is expressed in units of 100 μ, and if one assumes that the diffusible water content of G.5 emulsion at 50 % relative humidity is 53.7 mg/ml.

While this comparison has been made for the loss of water in a vacuum, the characteristic time should be the same for loss or gain of water in circulating air, if no hysteresis exists. The hysteresis effect is reported to be small by Swinnerton and Waller (SW 57).

At high water concentrations, the characteristic time is reduced, but it still varies directly with the square of the emulsion thickness. The differential equation is then

$$\frac{\partial c}{\partial t} = K(c)\,\frac{\partial^2 c}{\partial z^2} \qquad (4.7.4)$$

In this equation z is the coordinate of a plane in the swollen emulsion, and $K(c)$ is the unknown functional dependence of the diffusivity on the water concentration.

Some important problems do not require solution of Eq. (4.7.4) for their approximate treatment.

The problem of fixation, for example, is treated as follows: The concentration of hypo varies from a maximum at a point outside the emulsion to near zero at the boundary plane existing in the emulsion between dissolved and undissolved halide. Similarly the dissolved silver concentration varies from a maximum at the halide boundary to a low value in the solution exterior to the emulsion layer. The rate of solution of halide, or the velocity of motion of the boundary, is written:

$$\frac{dx}{dt} = \frac{B}{x + l} \qquad (4.7.5)$$

This formula is due to Demers (D 58). The meaning of the symbols are as follows: x is the distance from the surface to the moving boundary of undissolved halide. It can be assumed to be the actual thickness of this swollen emulsion layer. The time is symbolized by t, and l is a characteristic length that describes the fixation process. For example, if the fixer is stagnant l is large; if well stirred, l is reduced. If the halide is difficult to dissolve, l is increased. The quantity B is a constant for a given type of emulsion and fixing solution. Equation (4.7.5) merely states that the rate at which dissolved silver leaves the emulsion or fresh hypo enters the emulsion is proportional to their concentration gradients in the emulsion. The time for all the halide to dissolve then is $L(L + l)/B$, where L is the value of x at which all the halide is dissolved. (Demers has assumed B to be proportional to the hypo concentration, but this assumption is not valid at high concentrations.) Because the nature of the outer layer of emulsion in which the halide has already been dissolved probably does not change with time, the diffusivity can reasonably be taken as constant. Demers has found that various experimental data are fitted with l between 25 and 100 μ. He has also applied Eqs. (4.7.2) and (4.7.3) to the processes of developer penetration, stop solution penetration, and to the washing and drying of emulsion layers by substituting for the emulsion thickness L, an equivalent thickness $L + l$.

In some of these applications this seems to be an oversimplification, because the diffusivity can change by orders of magnitude depending on the water concentration in the emulsion. Experimentally the penetration time of developer, and presumably other solutions in emulsion, follows a "thickness squared" law (O 53).

The long characteristic diffusion time for water in emulsion at low concentrations is doubtless a consequence of the very close packing of the silver halide crystals in nuclear-track emulsion. The closest periodic packing of spheres of uniform size gives a volume concentration equal to $\pi/3\sqrt{2}$, or 74 %, whereas in standard emulsion the halide concentration is already about 50 %.

The large diffusion time has caused various experimental difficulties and falsified much data, because in many experiments it was thought that the emulsion density was uniform when it actually was not. When shipped by the manufacturer, thick emulsion is unlikely to have reached complete equilibrium throughout its volume, and when emulsion is placed in vacuum for an experiment, uniform dryness is seldom attained. Further experimental and theoretical work on this problem probably would be justified because of the rapid change of diffusivity of concentrated emulsions with water content. Whereas for small water concentrations the characteristic time was found to be $z_0^2/6.5$ days, in emulsion swollen with neutral water it is only about 1 % of this figure. The salt content of the solution as well as its hydrogen ion concentration and temperature also affects the swelling and diffusivity of the emulsion, with practical consequences.

It is also known that the behavior of gelatin toward water varies with the gelatin sample, but the importance of this variability for the processing of nuclear-track emulsions has not been evaluated.

The increased diffusivity of swollen emulsion is utilized practically in obtaining more uniform development of thick emulsion layers. The emulsion is presoaked in distilled water before immersion in the developing solution in order to obtain more rapid penetration of the developer into the deeper layers of emulsion.

4.8 Thermal Lags in Processing

During processing the temperature of the emulsion often must be changed, and if the time for the change to be effected throughout the emulsion volume is not small compared to the time it is maintained at the new temperature, there is danger of nonuniformity. The changes in temperature are brought about either by immersing the plate in a solution, or by putting the glass in contact with a metal plate maintained at the desired temperature. It is to be noted that if a hot plate is used, the cover over the emulsion should be at the same temperature as the plate, or the surface of the emulsion will tend to come to an equilibrium temperature determined by radiation exchange with the cover. It is not determined solely by the metal temperature because heat is conducted through the glass and emulsion relatively slowly.

Heat must penetrate into or out of emulsion from one or both surfaces; edge effects usually are small. The diffusion equation now is: $S(\partial T/\partial t) = K(\partial^2 T/\partial z^2)$. Here S is the specific heat, T the local temperature, t the time in seconds, K the thermal conductivity, and z the depth of penetration. The solutions of this equation are found by a Laplace transform. As in the problem of water diffusion (Section 4.7), there exists a characteristic time $z_0^2 S/K$, where z_0 is the thickness.

Taking the specific heat, S, as 0.2 for glass and its thermal conductivity as 0.002 cal cm deg^{-1} sec^{-1}, for a thickness of 1 mm this time is about 1 sec. In a few seconds, therefore, the glass plate on which the emulsion is mounted will be heated uniformly to the temperature of any surface with which it has good contact or to that of a liquid in which it is immersed. Water-soaked emulsion will have much the same constants as water itself. If one takes $S = 1$ and $K = 0.0014$ cal cm deg^{-1} sec^{-1} for a swollen thickness of 1 mm, the calculated characteristic time of the emulsion is about 7 sec. Therefore, no appreciable delay is involved in changing the emulsion temperature when it is immersed in a solution at a new temperature, but if heated only from the bottom, a thermal gradiant through the emulsion may exist.

4.9 Measurement of Emulsion Density and Pellicle Thickness

An emulsion stack should be taken apart as soon as feasible after an exposure, and during the interim should be protected against loss or gain of water. As soon as the pellicles are exposed to the air their density should be measured, and if any delay is encountered, the pellicles that are to be measured should be clamped between sheets of glass or otherwise protected from loss or gain of water. The need for this precaution will be realized if one observes the changing weight of a pellicle freshly exposed to air with which it is not in humidity equilibrium.

The *average* stack density can be determined by weighing it and by measuring its external dimensions. If the edges are irregular, external machining of the stack or measurement of the area of each sheet is recommended. If mounted carefully on glass, the area measurement can be made after processing. Since the pellicles are not in perfect contact with each other, the average stack density will always be less than the emulsion density, and no indication is obtained from the average density of the corrections required for the air spaces within the stack. If one does not know the actual emulsion density, he has no satisfactory method for adjusting measured ranges to standard conditions.

It is best to determine the density of samples of the emulsion by Archimedes' principle. The weighing is done in air and in a suitable liquid. The method developed by the writer's group employs carbon tetrachloride, but methyl salycilate, used by Ilford, Ltd., has been tried and found to give the same results. It is best to carry out the density determination in air that is close in relative humidity and temperature to that with which the emulsion is in equilibrium. Then, when a piece of emulsion is suspended from the arm of the chemical balance used in the weighing, the weight will not be observed to change with time. A small rate of loss or gain of weight can be corrected by utilizing the fact that the initial part of the absorption or desorption curve is linear if one plots the pellicle weight against the square root of the time elapsed since the ambient humidity was changed (Section 4.7). If the temperature of the emulsion is below the dew point, water will condense out on it, and the weight will increase. The cooling caused by the evaporation of carbon tetrachloride may be sufficient to cause this effect.

For maximum accuracy, one not only allows for the tares in air and submerged, but allows for the buoyancy of the air. The vapor of CCl_4 is heavy and unless wafted away will interfere with the measurement. It is also toxic, so it must be kept off the skin, and the weighing in CCl_4 *must* be done under some type of hood.

The weighing is first carried out in air and the tare subtracted. Then it is weighed in the liquid, the temperature of which is carefully recorded, and the corresponding tare subtracted. Finally, another weighing is made in air. The weight is followed until it reaches a minimum corresponding to the evaporation of all the CCl_4. This normally will check the original weight to a part in 10^4 or so. If the CCl_4 contains dissolved impurities, these will be left on the pellicle when the CCl_4 evaporates, and the weight will not return to the original value.

For weighing small samples (~ 1 gm) of emulsion, a hole is punched in the piece and it is suspended by a 1 mil wire. Larger pieces can be gripped with paper clips and the use of whole-pellicle samples is recommended. The CCl_4 has no effect on the latent image.

The density, ρ, of the emulsion is found simply from the following formula.

$$\rho = \frac{W_a \rho_l}{W_a - W_l}$$

Here W_a is the corrected weight in air, ρ_l is the density of the liquid at the temperature of weighing and W_l is the corrected weight in the liquid. To a sufficient approximation, the density ρ_{CCl_4} of CCl_4 is $\rho_{CCl_4} = 1.63255 - 1.9110 \times 10^{-3}\theta - 0.690 \times 10^{-6}\,\theta^2$ gm/cm^3, where θ is the centigrade temperature.

If the area of the pellicle is A, its average thickness is $t = (W_a/A\rho)$. This is the best way to determine the average pellicle thickness. For accurate emulsion work, each pellicle of the stack should be routinely weighed and its area and thickness measured. An estimate of the thickness of the average pellicle is obtained by dividing the total stack thickness by the number of pellicles. When this is compared with the average of the individual thickness, it is normal to find an excess of a few microns per pellicle, attributable to air spaces between the layers. By equalizing the clamping force over the stack, and clamping firmly (about 44 lb per square inch), the air spaces can be minimized. Still higher clamping forces are not advisable because then the emulsion tends to flow laterally (B-T 58) and the pellicles may fuse together.

A less satisfactory method of determining the emulsion thickness is simply to measure the pellicle thickness with a micrometer caliper or a dial gauge. The measurements, if made this way, should be carried out at several identifiable points on each pellicle. Individual measurements of this sort are seldom accurate to better than 1 %, but may usually be improved by averaging. After processing, when the pellicles have come to an equilibrium condition of dryness, the thickness of each pellicle should be measured again at the preselected points. One uses the fine focussing motion of a microscope for the measurement if the relative plate-objective separation varies linearly with the rotation of the knob. Then, whatever the units in which this measurement is made, and whatever the emulsion index of refraction, the effective shrinkage factor can be obtained if the all-important original thickness was recorded. Subsequently, microscope measurements of the thickness should be taken whenever angle or range measurements are made, so that the applicable shrinkage can be used.

In the writer's experience the shrinkage depends almost entirely on the processing to which the emulsion is subjected, batch-to-batch variations in emulsion of standard nominal composition being small.

A method for permanently recording the original emulsion thickness on a plate was developed by the writer (B 51). It consisted (Fig. 4.9.1) of casting the X-ray shadow of a sharp-edged heavy-element absorber through the emulsion at precisely 45° to the surface. The blackened wedge of emulsion after processing then will be equal in length to the original thickness of the emulsion if there is no shear distortion. Such a procedure may occasionally be found useful. A better method, if the facilities are available, is to expose the plate to a beam of high energy particles which scatter negligibly, the beam being inclined at $\pm 45°$ to the perpendicular (GM 57). Then, after processing, the average projected track length will be accurately the original emulsion thickness.

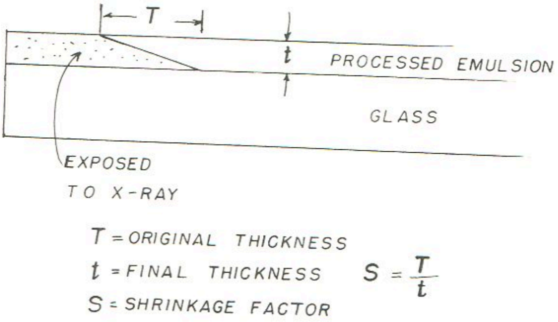

FIG. 4.9.1. Illustration of a means for determining the original thickness T of a layer of emulsion mounted on glass (IDLRL).

M. Blau and J. N. Smith (BS 56) have measured the absorption coefficient of 40 Kev X-rays in emulsion and have found that it is large enough to be used to measure the thickness, or at least the heavy element areal density of emulsion.

Direct measurements of emulsion thickness are subject to errors because mechanical gauges have a tendency to make depressions in the emulsion surface, especially when it is soft. This source of error can be avoided by approaching the surface with a gentle stream of air to determine the position of the surface. The resistance to the air flow out of a nozzle varies rapidly as the surface is approached, and the relative surface position can be defined by selecting a particular point on this curve of resistance versus distance.

If care is taken to make very accurate weighings, even the density and thickness of emulsion layers mounted on glass plates can be determined, as demonstrated by Oliver (O 53), but since the weight and volume of the glass must be separately determined, some emulsion on the plate must ultimately be sacrificed.

4.10 Emulsion Loading

The complex constitution of emulsion, which dismays the amateur, is revealed, with deeper understanding, as an important reason for its extreme versatility.

By far the most abundant nucleus in emulsion is the free proton. The high density of hydrogen permits one to study elementary particle-proton interactions and gain some results, which in dollars of cost per unit of information are competitive with any instrument. The nuclei of the CNO group also are abundant. In them typical light-nuclear reactions occur, and often the specific reaction can be identified. The small amount of sulfur in emulsion is usually neglected. The reactions with the Ag, Br, I group of elements are typical of reactions with complex nuclei. The fission region is not represented in the atomic numbers of normal emulsion constituents, but heavy nuclei can be added as loading materials. This possibility of altering the proportion of any element present and incorporating into emulsion almost any desired element is one of the most powerful techniques available to the emulsion experimenter.

General procedures for finding the stopping power of an emulsion of arbitrary composition are given in Chapters 9 and 10. Some considerations that determine how much and which materials can be used for loading are the following: (*a*) The additive must either be transparent, present in a sufficiently low concentration to be tolerable, or be removed in processing, in order that the optical transparency of the gelatin not be destroyed. (*b*) It must be in a chemical form neither to act as an oxidizing agent nor as a reducing agent; it must neither destroy the latent image nor fog the emulsion. (*c*) It must behave in no way that is incompatible with the primary functioning of the emulsion as a track recording instrument. (*d*) If not homogeneously distributed, it usually must be finely divided.

Nuclear-track emulsion is supplied by the Eastman Kodak Company and by Ilford, Ltd. loaded with lithium or boron; either natural elements or separated isotopes may be introduced into the emulsion. Lithium has also been loaded in the form of lithium glass dust (RK 57). Other alkali metals presumably can be loaded in the same way as lithium is loaded. Ilford, Ltd. have supplied C.2 and E.1 emulsion normally loaded with 16 mg/ml of lithium or 23 mg/ml of boron, and have also loaded emulsion with separated Li^6, Li^7, B^{10}, or B^{11} supplied by the purchaser in the form of lithium sulfate and sodium borate. Boron-loaded emulsions are more sensitive and fade less than the unloaded emulsion. Wilkins (W 51) has given corrections to the range-energy relation for boron-loaded emulsion.

According to Labew and Nikolae (LN 59) NIKFI emulsions are impregnated with 10-120 mg of boron per cubic centimeter or 3.5-20 mg of lithium.

Emulsion loaded with water has been extensively employed by Goldhaber *et al.* (GBFG 53). It is often desirable to introduce deuterium into emulsion or to increase the normal hydrogen or oxygen content. For this, water loading is practical. The actual hydrogen density in water-soaked emulsion of density ρ in grams per milliliter (the volume of water in this case is surely closely additive with the volume of standard emulsion) can be found from:

$$\rho_H = 0.1325 - 0.0206\rho \text{ gm/ml} \tag{4.10.1}$$

The volume is greater by the factor σ [which $= 2.815/(\rho - 1)$] than the unswelled emulsion.

The density ρ_H is the concentration of the *light* hydrogen isotope after soaking in ordinary water. For heavy water the density of deuterium approaches nearly twice this figure when 100 % heavy water is used and when the swelling is sufficient that the light hydrogen (which may partially exchange with deuterium) content can be considered small compared to the concentration.

There is an advantage in the use of water-loaded emulsion for observing hydrogen interactions. The areal density of events observable on the emulsion plate is increased by the factor $2.08\sigma - 1.08$ as compared to standard emulsion, while the background events from the emulsion do not increase at all. The increase in background from the additional oxygen varies linearly with the density of hydrogen events.

Not only glass backed plates, but also stacks of water-loaded emulsion have been used successfully. Ascoli, Hill, and Yoon (AHY 58) swelled 600 and 1000 μ G.5 pellicles to 3.1 times their original volume and 2.1 times their original thickness by soaking in water at 4°C for 12 hr.

The wet pellicles were kept chilled to 5°C in a box, the inside dimensions of which were the same as the pellicle dimensions. This prevented the pellicles, which are slippery under these conditions, from sliding with respect to each other. The stack was X-ray-gridded before disassembly. The pellicles were processed off the glass and mounted after processing on glass plates using a thermo-setting plastic. The grain density at minimum obtained in various tests was 16-22 grains/100 μ. Despite the advantageous features this method possesses, Ascoli *et al.* found that the additional complications of preparation, the lower grain density, the larger shrinkage, the larger distortions, the higher fog level observed, the reduced accuracy of the range-energy relation, all made the water loading of emulsion for the study of negative K meson-proton interactions unjustified.

On the other hand, Kriventsova *et al.* (KLS 60) reported good results using emulsion pellicles loaded with water so that their shrinkage factor was about 6. Then perpendicular exposure to 8.2 Bev protons permitted the efficient study of small angle p-p scattering. There are several obvious advantages for the method of normal incidence apart from the augmented hydrogen content derived from the loading. The grain density is increased by the factor of the shrinkage, and the scattering angle is increased by the same factor as compared to tracks in the emulsion plane. A further advantage for the method of normal incidence is that the tangent of the scattering angle itself is directly measured. When the dip angle and azimuth angle are separately obtained, the derivation of the space angle involves both tedium and error.

Tracks parallel to the emulsion plane that are produced while the emulsion is swollen will have a lowered grain density even if the emulsion is not desensitized by oxidation of the latent image. This is because a reduced density, n, of emulsion grains is traversed by a particle in penetrating the emulsion.

If an Ilford emulsion is soaked in water and then allowed to dry, about 1 % of its weight disappears. This loss is ascribed to soluble glycerin which is believed to be used as a plasticizer in the emulsion manufacture.

Vigneron, Genin, and Chastel (VGC 55) have observed a peculiar behavior of Ilford C.2 emulsion. After swelling by water, if it is dried by a hydrophilic liquid such as acetone or alcohol, the emulsion seems to be porous; the density may be lowered to 3/4 of normal. The effect disappears if the emulsion is resoaked and dried in air.

Usually the emulsion sensitivity is decreased by water-loading, perhaps through oxidation of the latent image, but often merely because the pH is lowered. This problem has been little studied, however, and the optimum conditions of dissolved gas content, pH, and the temperature of the water for stability of the latent image are not known. General experience advises keeping the emulsion cold, and developing as soon as possible after exposure. The presence of even trace amounts of oxidizing agents is known to hasten the destruction of the latent image, and low pH values are deleterious to the emulsion sensitivity. Wet emulsion can be kept firm and reticulation avoided if it is cold. The ice point is a convenient one for storage of water-soaked emulsion.

Three parameters which affect the diffusion of water solutions into emulsion are the (*a*) hydrogen-ion concentration, (*b*) temperature, and (*c*) dissolved salts in the water. Most of the studies that have been made of the swelling of emulsion when soaked in water are of little value because one or more of these variables has not been controlled or recorded. For this reason Miss F. Smith, Mr. Carl Cole, and Mr. Robert Blinkenberg have made further measurements at the Lawrence Radiation Laboratory. The water absorbed in unmounted 600 μ pellicles of G.5 emulsion was studied for solution pH values in the range 1-12 and for temperatures of 0°-30°C. The desired pH was obtained by adding hydrochloric acid or sodium hydroxide to a large volume of water in which the pellicle was immersed. The buffering action of the emulsion itself then had little influence on the solution pH. [Picciotto (P49) states that C.2 emulsion buffers a solution to pH 5.9-6.3 and boron-loaded B.2 emulsion maintains pH 9.0.]

On immersion in water the emulsion swelled for a period of hours approaching asymptotically an absorption of W_0 grams of water for each original cubic centimeter of standard emulsion. The quantity W_0 is a function of the temperature and pH.

In the region 0-30°C at a pH of 7.0, unit volume of dry G.5 emulsion absorbed an ultimate mass of water, W_0, equal to $24.4\ (33\text{-}T)^{-0.7}$ gm. Here T is the centigrade temperature. When the pH was varied while keeping the temperature close to 25°C, the ultimate absorbed mass of water per unit volume of standard emulsion was found to be given by $W_0 = 2.42 + 0.44\,(\text{pH})$ gm—essentially linear with pH between pH 1 and pH 12.

The emulsion swells without limit when a critical temperature, T_c, is reached, This is given in a linear approximation by:

$$T_c = \frac{87 - 3(\text{pH})}{2}\ {}^\circ\text{C}$$

The amount of water absorbed per milliliter of emulsion in the vicinity of 25°C and pH 7 was found to be given by:

$$W_0 = \frac{10.7[1 + 0.182(\text{pH})]}{[43.5 - 1.5(\text{pH}) - T]^{0.7}}\ \text{gm/ml} \qquad (4.10.2)$$

It is well established that when emulsion is soaked in ordinary water that the volumes are quite exactly additive. See, for example, S. Goldhaber *et al.* (GBFG 53).

The transient swelling behavior of emulsion has been incompletely studied. The water absorption is rapid at first, subsequently leveling off at the saturation value, W_0, per unit initial volume. The later stages of the swelling are reasonably well represented by the formula

$$W = W_0(1 - e^{-t/\tau}) \tag{4.10.3}$$

but neither an exact theory nor precise experimental data on the transient behavior of emulsion as a function of temperature and hydrogen ion concentration is available.

When emulsion is swollen by immersion in water it does not behave isotropically. The molecular structure evidently has an axis perpendicular to the plane of the emulsion along which the emulsion contracts and expands more readily than in other directions. Demers (D 58) has compared the molecules to coiled helical springs with axes parallel to the direction of gravity at the time the emulsion was poured. To be specific, at pH 7 and at a temperature of 0°C, the thickness of an unmounted pellicle was observed to increase 114 % while the lateral dimension increased by only 22 %.

At 25°C a crude estimate of τ is $9/(\mathrm{pH})^{1/2}$ hr for 600 μ pellicles. The time to gain half of its ultimate weight increment increases with the plate or half-pellicle thickness, but very little with temperature, and only slowly as the pH falls. For G.5 pellicles 600 μ in thickness, the time required for half the limiting quantity of water to be absorbed at pH 7 is about 1.1 hr. Beyond the half-saturation point the behavior of the emulsion is quite well described by Eq. (4.10.3). In order to avoid mistakes in estimating the water content of emulsion, it must be protected from evaporation when stored.

An unanticipated result of these measurements is the form of the temperature-dependence of the swelling. Whereas pure gelatin is stated (M 54) to swell more at reduced temperatures, the opposite is true here, indicating rather profound changes in the gelatin during emulsion manufacture. No minimum swelling near the isoelectric point of gelatin was observed either, although DeCarvalho and DaSilva (DD 58) report that it occurs at a pH of 4.25 ± 0.1.

The methods used for concentrating deuterium usually also concentrate tritium, so that in a sample of deuterium the amount of tritium, compared to that in normal hydrogen, may be greatly increased. It is necessary in loading emulsion with heavy water, or other deuterium compound, to check the tritium content. The effect of tritium on a deuterium-loaded plate is to increase the single grain background or fog in the emulsion. Heavy hydrogen, low in tritium, can be obtained if this specification is made. If low tritium concentration is not certain, heavy water-loaded emulsion must be developed as soon as possible after loading. One can at the same time conserve heavy water and insure that no error in the estimate of the deuterium content is introduced by proton-deuteron exchange, by adding to the dry emulsion just the quantity of heavy water required to swell it the desired amount. Although ND_4Cl (P 40), calcium nitrate containing heavy water of crystallization (GGL 47), and hexadeuterodiacetin (MCG 50), have been tried, no other known loading material is as satisfactory as heavy water itself.

Loading emulsion by soaking it in a solution of the desired additive is one of wide but not universal applicability. The gel combines with many salts and prevents their deep penetration in the emulsion. This can often be avoided by complexing the material, but as a general rule some tests for uniformity of loading in the emulsion should be made. Often the most direct method for finding the amount of material loaded is to measure the change in the net content of the solution in which the emulsion was soaked before and after impregnation. Another, which however does not produce uniform loading, is to put a puddle of known volume of solution on the emulsion surface and permit it to soak into the emulsion.

Loading emulsion with uranium is important, and many procedures for impregnating emulsion with it have been employed. They have had various degrees of success. The impregnating material used has most frequently been a uranyl salt. It was thought at first that the uranium had a specific desensitizing effect on emulsion, but it was pointed out by Picciotto (P 49) that the effect was probably caused by the low pH values of the solutions employed.

Burdet and Philbert (BP 62) were able to load emulsion homogeneously to a concentration of 2.4×10^{21} uranium atoms per milliliter of dry emulsion. It was impregnated with an optimized uranyl acetate solution. It was found, however, that when the uranyl acetate concentration was made as high as 2 % it affected the photographic mechanism, and the tracks were partly destroyed.

Rochat (R 49) has given details of a process that successfully impregnates emulsion at pH 4.89 with a large quantity of uranium, while at the same time maintains a useful emulsion sensitivity.

In a total volume of 100 cm³, 30 gm of uranyl nitrate, $UO_2(NO_3)_2 \cdot 6H_2O$, first are dissolved in a 0.4 M sodium citrate solution. (The sodium citrate solution is prepared by dissolving 35.712 gm of the salt in 250 cm^3 of solution.) The uranium solution prepared in this way has a pH of 1.84. Sixty cm^3 of normal sodium hydroxide solution is divided into four parts. These are successively added to the uranium solution, stirring the yellow-orange precipitate that forms each time until it dissolves. Plates coated with Ilford D.1 emulsion 50 μ in thickness are soaked in this solution at 17° ± 1°C for 2 hr with slow agitation, the plates being horizontal with the emulsion side up. In order to obtain reproducible loading it was found useful to withdraw the plate from the solution on an inclined plane at a uniform rate of about 3 inches per minute. The plate is then placed in a partial vacuum of air dried with $CaCl_2$. The pressure in the drying chamber is maintained at a few millimeters of mercury and the relative humidity is close to zero.

Rochat's plates were developed by a temperature cycle process in D-19 developer diluted with three parts of water. Temperature cycle development is hardly necessary for 50 μ emulsion, however. At the Lawrence Radiation Laboratory 200 μ plates have been successfully loaded by this method.

The amount of uranium taken up, and its distribution in the emulsion, may easily be determined for quantitative work. After waiting a known time, ΔT, between loading and processing, the density ΔN of alpha-particle tracks in the emulsion yields the density N of radioactive nuclei in the emulsion: $N \approx (1/\lambda)\,(\Delta N/\Delta T)$, where λ is the decay constant of the isotope yielding the alpha-particle tracks observed. While the plates are wet between the time they are loaded and dried, the alpha particles are either not recorded at all, or the tracks may appear very thin. This effect must be allowed for in the calculation; the drying should be accomplished promptly, and the time between loading and processing made large compared to the drying time.

There is reason to believe that uranyl loading deposits uranium on the silver bromide. An unusually large amount of uranium relative to other elements is taken up by the emulsion, and tracks in uranyl-loaded plates may be intensified. (See Section 5.11.)

A new method (LS 59) for loading emulsion with uranium has been developed at the Enrico Fermi Institute:

A 10 % uranyl nitrate solution is prepared using nitrate which has been separated from the products of uranium decay by ether extraction. This solution is neutralized with Versene $[Na(C_2H_3O_2)]_4C_2H_8N_2$: about one Versene molecule per atom of uranium. There is a precipitate at this stage which is dissolved by acetic acid. The solution is then returned to pH 7 with ammonia and no precipitate should persist. The pellicles separated by paper sheets are immersed in this solution for

8 hr at 5°C. The sheets are then removed and the stack is frozen at dry ice temperature. This reduces the fading which is further reduced by boiling all liquids in which the pellicles are to be soaked. The solutions are also saturated with nitrogen before use. The emulsion can be transported and exposed at the dry ice temperature. Before processing the emulsion is immersed in cold, boiled, and nitrogenated water. The pellicles come apart readily in the water. After brief rinsing to remove some of the uranium they are processed off the glass. By this procedure the uranium concentration obtained was 2.8×10^{20} nuclei/cm^3.

Demers (D 58) has reviewed exhaustively the experimental results on uranium fission in emulsion.

Juric *et al.* (JWD 57) described their experiments on loading of emulsions with uranium and other materials before pouring. They put Ilford gel into a solution of the loading material. The two are thoroughly mixed at 40°C. The pH cannot be lower than 6 or the gel coagulates. For the same reason alcohol is not used as a solvent for the loading material.

Very recently it has been reported that concentrations of such heavy elements as lead and tungsten comparable to that of the AgBr itself have been loaded in emulsion layers (BZNP 60). Lead sulfate and silver halide, for example, have been coprecipitated in a water solution of gelatin during emulsification. Tungsten, on the other hand, was prepared as a gelatin-protected emulsion of $CoWO_4$. This was mixed with the silver halide gel. Presumably these compounds can be dissolved out of the gelatin during processing as the silver halide is.

To overcome the problem of opacity of the loading material, transparent forms of the material may be used. Xuan and Chastel (XC 58) and the laboratories of Kodak Ltd. and Ilford, Ltd. have found it practical in order to load an emulsion with carbon to introduce it in the form of diamond dust. Particles 0.3 to 1.1 μ in diameter are placed in a thin layer between two sheets of emulsion. By this means they were able without ambiguity to study the disintegrations produced by fast neutrons in carbon.

The carbon content of emulsion relative to silver halide has also been increased by a factor of up to 2.9 using glycerin loading (GBFG 53). The emulsion is swelled with water before immersion in glycerin.

There are not many elements with atomic numbers near that of chlorine that are easily loaded in emulsion. It has been found possible, however, to manufacture silver chloride emulsions that record the tracks of charged particles. H. Gauvin and W. Sebaoun (GS 57.1) have investigated such emulsions while controlling the pAg potentiometrically, and varying the pH, and also varying both during precipitation. Grain size, fog, and sensitivity were found to be controllable. Square crystals 0.1 μ on a side were produced in some pourings. It was found that good discrimination between the tracks of alpha particles and protons was obtainable if the pH and pAg were regulated during the second ripening. Other silver halides such as the fluoride are also possible additions as either active or inactive emulsion constituents.

As described in Chapter 3 diluted emulsions are available in which the silver halide concentration is reduced relative to the H and CNO concentrations. The grain density at the minimum of ionization in diluted G.5 emulsion can be maintained high enough for recognition of such tracks in four-times diluted emulsion. The composition and range-energy relation for such emulsions are known (see Chapters 3 and 10).

Attempts have been made to avoid nitrogen as a constituent in emulsion by replacing the gelatin with polyvinyl alcohol (D 57.1, F 61).

Plates with alternate thin layers of gel and emulsion or mixed layers of varying composition are sometimes used as an aid to analyzing separately nuclear reactions in light and heavy elements. Special loadings with specks of a large number of materials can be made, but the concentrations obtainable in most cases are very low. Heavy elements such as bismuth, thorium, and uranium, however, are reported to be readily complexed by ethylenediaminetetraacetic acid (Versene) and can in this form be loaded into emulsions. H. G. DeCarvalho and A. G. DaSilva (DD 58) have made successful loadings of emulsions with these materials and have subsequently developed them with ferro-Sequestrene complex at a pH as low as 2.0.

Loading of emulsions with fine wires or fibers has also been described (DY 51, MOV 51), but results of only limited value have been obtained by this method. One problem encountered is that the wires do not swell and shrink with the emulsion so that serious distortions near the wires are unavoidable. Wires of such active elements as aluminum also must be coated to prevent chemical action. The principal advantage of this method is that the scanning need be only linear—along the wires.

Meulemans *et al.* also tried loading emulsion with glass capillary tubes. Radioactive liquids or gases in extremely small quantities can be studied in this way. An improvement was made when Bonetti and Occhialini (BO 51) simply coated the tube with emulsion so as to form a cylinder. After processing, the emulsion was reswelled to its original thickness and no distortion of the tracks was then observed. Unfortunately the optical problems encountered in scanning such cylindrical emulsions were found to be severe, and little practical use has been made of them. Sometimes it is essential that the emulsion-loading material be in the form of a very thin layer—small compared to the range of a fission fragment, for example. For this purpose the emulsion "sandwich" may be suitable. By preparing suspensions of finely divided materials in a gelatin solution, Vigneron and Bogaardt (VB 50) were able to coat the emulsion surface of a plate with a layer containing roughly 50 % by weight of UO_2. The dried thickness of the layer was 0.37 μ. Since it must remain reasonably transparent a greater density of uranium could not be used. This layer is covered by a pellicle which is put in intimate contact with it. Then after exposure and processing all disintegration-product tracks that emerge from a grain of loading material are visible.

"Emulsion chambers" made of several layers of emulsion interleaved with plates of light and heavy elements have had considerable use in cosmic ray investigations, particularly when it is desired to study electromagnetic cascades (E 58).

Experiments to determine how to load emulsion with phosphors have been made (M 58.2). Although the presence of a phosphor in the emulsion matrix could increase the emulsion sensitivity, the most important prospective gain would be an improvement in discrimination.

Layers of solid hydrogen and deuterium have been used in contact with emulsion surfaces (OST S 57). More applications of solidified hydrogen as a target material may be anticipated.

4.11 Sources of Ionizing Particles for Testing Emulsion Sensitivity

One often wishes to test batches of emulsion for sensitivity while perhaps varying the processing technique. If a high energy accelerator is available, one may make an exposure to produce fast meson or proton tracks in the emulsion. Protons of about 3 Bev or pions of about 450 Mev produce tracks at minimum grain density. The grain density rises both above and below this velocity, so one must stay in this vicinity to be sure he is at the minimum. A high energy electron synchrotron is also a possible source of tracks, but if the electron energy is several tens of Mev or higher, the ionization should be assumed to be that corresponding to the plateau—see Table 9.5.1.

A method of testing the emulsion without employing an accelerator is simply to use a beta-ray emitter, preferably one free of gamma rays. A small source of Sr^{90} giving 10^5-10^6 counts per minute has been found by the writer to be simple and safe to use. It should be stored in a lead vessel, however, and kept away from film except when in use. The source is mounted within a few millimeters of the surface of the emulsion under test and left 10 min or more. The processed emulsion will show tracks of electrons radiating out from the source. The fastest, straightest tracks are mostly in the energy interval 1-2 Mev and are, therefore, ionizing at the minimum. Actually these beta rays come from Y^{90} which is a short-lived daughter in equilibrium with Sr^{90}. Sr^{90} has a half-life of 25 years and a maximum beta-ray energy of 0.6 Mev.

A second possible radioactive material which has been tested by Starinin (S 57.1) is P^{32}. When 1.5 Mev electrons from it are focused by a beta-ray spectrometer on the emulsion good tracks were obtained near the surface.

Starinin has reported that possibly the best beta-ray source is Ce^{144}, the daughter of which, Pr^{144}, emits electrons with an energy upper limit of 2.97 Mev. A solution of a cerium salt is placed on a stainless steel plate, and after it is dry it is primed with cement. Electrons of 2.5 Mev or so emerging from this source can be selected by a beta-ray spectrograph and caused to bombard emulsion. Thin layers of emulsion can be tested satisfactorily, but if it is necessary to observe tracks deep in the emulsion, a high energy accelerator is preferred. At the NIKFI Institute a 15 Mev betatron is used for this purpose.

Another method of testing the emulsion is to let it remain exposed to cosmic rays for some weeks. If it is of a type that does not fade, the greatest density of straight, near minimum tracks found in the emulsion when it is processed are those of muons on the plateau of ionization. These tracks may be used to determine the emulsion sensitivity.

If the emulsion is not sensitive enough to record tracks at the minimum of ionization, tracks of any mesons or hydrogen nuclei that terminate in the emulsion can be used to measure the sensitivity. These can be produced by an accelerator, by cosmic rays, or by a radioactive alpha-particle source. The alpha particles may be used to project protons out of a hydrocarbon foil or they can produce neutrons by bombarding beryllium. The neutrons, in turn can be caused to collide elastically with protons in the emulsion or in an external radiator (see Volume II).

The sensitivity of any emulsion can be expressed by assigning to it a value of $g_{\min}$, the density of grains at the minimum of ionization (see Chapter 9). Even if visible tracks are not formed at this particle velocity, proportionality of the grain density to the restricted rate of energy loss should be a good approximation in the region from the minimum of ionization up to the point where very unsaturated tracks first are formed.

Strictly speaking, sensitivity should be separated from halide concentration and grain size. The sensitivity is correctly stated by giving the ratio $g_{\min}/n$ (see Chapter 3). Unfortunately there is no direct correlation between sensitivity to light and to charged particles, although for a given emulsion type such a correlation exists and is used to compare emulsion batches. The correlation with exposure to light is better for conditions of extreme short-exposure reciprocity failure.

4.12 Hypersensitization

If a high grain density is required, it is possible to obtain triethanolamine hypersensitized emulsion from Ilford Ltd. In order to avoid very rapid spoiling of this emulsion it must be kept refrigerated preferably with dry ice. Even keeping the emulsion a single day at room temperature causes the fog to increase to a point where it probably cannot be tolerated. At present there does not seem to be reliable information on the maximum storage time at dry ice temperature.

An alternative is to hypersensitize the emulsion in the laboratory just before it is needed. The use of triethanolamine (TEA) to hypersensitize emulsion that had already been sensitized with gold was first reported by Perfilov *et al.* (PNP 57, P-D 58). TEA had had previous use as a sensitizer for photographic emulsions, and for the emulsions of Demers. Additional Leningrad reports of the effect of triethanolamine were made by Zhdanow *et al.* (Z-C 58, ZKS 60).

A very successful treatment of NIKFI type R emulsions was described by Bogomolov *et al.* (B-D 58). It was subsequently reported by Garin-Bonnet and Moulin (GM 59) that similar results were obtainable with Ilford emulsions. The procedure recommended by Ilford Ltd. (W 60, W 60.1) is essentially that developed at Saclay and described by Garin and Faraggi (GF 60).

In the Lawrence Radiation Laboratory, Dr. H. H. Heckman worked out a method for hypersensitizing thick pellicles which has been employed by the writer's group. The following is an outline of the steps taken to hypersensitize Ilford type L, K, or G pellicles. They are first soaked for 30 sec in a water solution of 60 to 70 % ethyl alcohol. The concentration depends on the emulsion dryness. The pellicle is then rolled on a clean glass plate to which it adheres well enough for the subsequent operations to be carried out. After drying for 2 hr the emulsion is soaked in a solution of 1% triethanolamine and 5 % glycerine for the periods shown in the following tabulation.

Thickness (μ)	Time (min)	Temperature (°F)
100	20	60
200	50	60
400	100	60
600	180	60

The pH of the solution is about 10. (Citric acid to lower the pH to 9.5 has been employed by Garin and Faraggi (GF 60) who report that the acid is a stabilizer and that the fog is reduced by it.)

After drying for 24 hr the pellicles can be detached from the glass and are suitable for assembling into a stack. After exposure the emulsion is developed as quickly as possible. Only when absolutely necessary is the temperature of the emulsion permitted to remain as high as room temperature for more than a few minutes. The development time and temperature may be lowered somewhat in order to reduce the fog and surface blackening. A surface blackening in addition to any effect of light normally occurs during development. This can be wiped off after the stop bath.

It is possible to obtain a grain density of 50 to 100 per 100 μ for a relativistic track in hypersensitized L.4, K.5, and G.5 emulsions.

CHAPTER 5

Processing Procedures

5.1 General Considerations

The conventional photographic processing operations of development, stopping, fixing, washing, and drying must be carried out for nuclear-track emulsion. In addition several new procedures are made necessary by the great thickness and concentration of the emulsion, and by the requirements that there be uniformity of development and low distortion. Even the conventional operations are rather profoundly modified by these conditions.

Reasonably satisfactory solutions to most of the problems of thick-emulsion development have been obtained by the introduction of pre-soaking in distilled water, by the use of amidol developer containing a restrainer and a neutral or acid buffer, and by the invention of low temperature developer penetration (said to have been suggested originally by Odd Dahl). One important procedure that has been used extensively, namely the dry, hot stage of development, is now perhaps being abandoned in favor of immersion development in a warm bath of modified composition or by prolonged immersion development at low temperature.

Currently amidol is the favored developing agent for thick emulsions. Some of the reasons for this choice are the following:

(*a*) It penetrates the emulsion more rapidly than most other agents.

(*b*) It is a strong reducing agent and functions at a low pH. In such a solution the swelling is moderate and serious distortions are not introduced.

Amidol has several faults, however. It is very easily oxidized and its oxidation products seriously reduce the transparency of the gelatin into which they are absorbed. Its oxidation products, also, may sometimes participate in obscure processes that completely destroy the tracks (see below).

G. Meulemans (M54.1) has observed the effect of changing the amidol concentration in the developer formula. When the developer hot-stage temperature is increased, the grain density of minimum tracks in G.5

emulsion seems to reach a plateau at 25°C or so for each concentration. This grain density is about 15/100 μ at 0.5 gm/liter, 22/100 μ at 1 gm/liter, and 31/100 μ at 4.5 gm/liter. The optical opacity of the emulsion rises rapidly with the temperature, however, and it also rises with the concentration of amidol approximately in proportion to the grain density.

In fixing and washing, thick emulsions encounter conditions that cause distortions. Distortions arise from the marked swelling that the emulsion experiences in these stages when it is soft and fragile, and from blistering caused by imperfect bonding of the emulsion to the glass supporting plate.

Fixing by acid hardening fixers or tanning baths is not suitable for thick emulsions because they then have too much tendency to strip off the glass.

Drying, which formerly was a serious source of distortion, is made much more controllable by the use of alcohol-glycerin baths.

The vessels, racks, etc., coming into contact with emulsion and the processing solutions can be of materials of the same sort as those used in optical photography. Glass is good, but of course it usually transmits light. Stainless steel, grade 316, is satisfactory, but welds must not introduce materials which may react with the solutions and produce electrolytic action. Heliarc welding is common. Most other metals should be avoided because of the galvanic and displacement reactions possible. Pure silver plated on copper or brass is not harmful, however, and many plastic materials, such as polyethylene, lucite, teflon, and hard rubber are safe. Paraffin is good, and plastic-lacquer lining of tanks has been found satisfactory. Phenol formaldehyde resins are generally good. Paints or lacquers used in contact with emulsions or processing solutions should be tested before use. Materials that may produce hydrogen peroxide, especially, should be avoided.

Distilled water should be used to make up the processing solutions. Impurity of the chemicals has many times been known to affect the emulsion. For example, solution of the silver grains along with the silver halide crystals may take place in fixing unless the hypo used is of good purity. Prolonged fixing especially at low pH and in the presence of oxygen and the oxidation products of amidol will tend to corrode the silver in any case. If the silver ion concentration in the solution is high, this tendency may be reversed.

Impurities in the wash water accumulate in the gelatin and lower its optical quality. The large volume of wash water required often makes the use of distilled water for this purpose impractical. If city water is used, metallic ions should be removed by a resin column demineralizer obtainable from scientific supply houses. The water should be filtered because it often contains tiny particles which react with the emulsion. For example, the city water in Lund, Sweden, contains finely divided manganese dioxide which produces pits in the gelatin surface.

Microorganisms seldom grow in silver halide emulsion, but the gelatin of the processed plate offers a medium in which several types have been observed. Their colonies are sometimes quite destructive of the emulsion surface. This occurs usually when emulsion remains moist for a prolonged period. A small amount of disinfectant in the wash water prevents their growth, and when alcohol drying is employed this problem has not been encountered.

A process by which severe corrosion of the silver grains may take place is found to occur especially if the emulsion is left in the stop bath too long. Some of the amidol remaining in the emulsion may oxidize, giving a red coloration to the gelatin. Then by an unknown process, it appears that the hypo can attack silver directly, converting it to Ag_2S. Ordinarily the presence of sodium sulfite will prevent the oxidation of amidol, but if it is lost by too long immersion in the stop bath, this destructive process can occur. Hydrogen also blackens emulsion. Because it is sometimes important to make experiments in which hydrogen is in contact with emulsion, the temperature must then be lowered to reduce the chemical activity of the hydrogen.

The design of small details of processing equipment merits careful planning. For example, glass plates are hard to remove from a smooth wet metal surface. Small disks or other bosses that raise most of the area from intimate contact with the surface should be used in place of smooth metal surfaces except when good thermal contact is required.

It is valuable to know of materials that will not adhere to emulsions. Paraffin-coated surfaces adhere only slightly, as does teflon. When these are in the form of thin flexible sheets, they may be readily stripped off wet emulsion if one pulls slowly and continuously. The writer has made extensive use of a commercial paraffin foil material that adheres only weakly to wet emulsion. Throughout processing, but especially when the emulsion is weak and swollen, it must be kept in a horizontal plane to avoid gravitational distortion (see section 6.7).

When one contemplates any processing operation on emulsion that has received a valuable exposure, it can hardly be too strongly recommended that all operations and baths be tried with actual test emulsion to be sure that no mistake has been made, and that each bath will perform as expected. There is hardly an acceptable reason that can be given for a processing failure, because there is usually adequate time to test thoroughly each phase of the processing before the immersion of irreplaceable material.

5.2 Processing Thin Emulsion Layers

Thin emulsion plates are now little used in high energy physics as the primary research instrument. It is chiefly a monitoring device which is employed for rapid assessment of the intensity, distribution, and character of radiations. Such plates are not expensive, and one should have no inhibitions about employing them freely. The processing should be simple, quick, and foolproof.

A common developer for thin emulsion layers is standard factory-mixed elon-hydroquinone formula D-19. In order to slow the development so that the bottom layers of emulsion experience virtually the same amount of developing action as the surface, the developer is diluted, usually 1 part to 6 of water. This gives reasonably uniform development to a maximum thickness of about 200 μ, when development is permitted to proceed at 20°C for 30 min.

Slight variations of formula D-19 are sold as ID-19 or D-19b. Formulas D-76, D-82, and D-8 are also used for developing thin layers of emulsion. If used diluted, the time of development is extended but the uniformity is generally improved in each case.

At high pH, necessary for the metol-hydroquinone developer efficiency, the gelatin does not permit deep penetration by the developing agent. For thicker emulsions, therefore, a developer such as amidol that is effective at low pH values is required, but amidol oxidizes rapidly and must be made up fresh for each development.

A stop bath of 0.2-0.5 % acetic acid is generally employed after development, but mere washing for a few minutes in water at about 15°C is not unsatisfactory for 50μ emulsion.

Fixing in sodium thiosulfate solution prepared by adding 300 gm of $Na_2S_2O_3 \cdot 5H_2O$ and 22.5 gm of sodium metabisulfite to 1 liter of distilled water is satisfactory. At 15°C the time for fixing with agitation is roughly 80 z_0^2 min where z_0 is the emulsion thickness in units of 100 μ. A slow flow of fixer across the emulsion surface is best, but other methods of agitation, including that of bubbling nitrogen through it so as to produce a slow mixing of the fixer, may be employed.

After fixing for a period 50 % longer than that required for the plates to clear, they may be transferred to flowing wash water. The washing time should be perhaps 40 z_0^2 min or more depending on the agitation and temperature. The washing complete, the plates should be soaked in a solution of glycerin for 10 z_0^2 min, and then dried in warm circulating air or in alcohol. The alcohol drying is carried out by immersion for a time 10 z_0^2 min in 190-proof alcohol to which 5 % glycerin has been added. If the fixing and washing are carried out in solutions above about 20°-22°C, the gelatin begins to suffer serious distortion; eventually reticulation and shearing-tears occur in it. Such plates are virtually useless.

Sometimes it is important to speed up the processing of plates, such as test plates used for monitoring the exposure of a large stack, while the exposure is continuing. Then one uses emulsion of 50 μ or less in thickness; he develops for only 15 min; he immerses in a stop bath for only 2 min; he fixes with agitation in ammonium thiosulfate (rather than sodium thiosulfate) only until the plates clear; he washes for 3 min in rapidly running water and dries in concentrated alcohol, or he examines the plate while wet either with a water-immersion microscope objective or under low power without oil.

Slightly better unformity of development in emulsion of any thickness is obtainable on employing the low temperature developer soak recommended for thick emulsions (see below).

It is difficult to make a serious error of processing when employing the procedure described above and using ready-mixed D-19 developer. It can be carried out by anyone with ordinary photographic darkroom equipment.

For comparison, the processing recommended by the NIKFI Institute for their type A-2, Я-1, Я-2, K, and R plates of 50 μ will be given. The procedure doubtless is capable of yielding good results, but seems quite troublesome and seems to offer numerous chances for error.

The Russian procedure follows: (1) Soak in distilled water at 20°-22°C for 10 min. (2) Develop in amidol developer of the following composition: amidol, 2.5 gm; sodium sulfite (anhydrous), 10.0 gm; water, (approximately) 1 liter; citric acid to bring the solution to pH 6.8 using potentiometer control. Length of development—10 min. An alternative method of development omits the presoak in distilled water. The development is carried out at temperatures of 24° ± 0.5°C for plates of type R, and 22° ± 1°C for plates of type A-2, Я-1, Я-2, and K. (It is noted that if a black sediment is precipitated when the amidol is dissolved, it is then necessary to use 3 gm of amidol.) The developer must be made up immediately before using. (3) The stop bath is a 0.5 % solution of acetic acid (pH 3.4) at a temperature of 5°-7°C—for 10 min. (4) It is then recommended to employ 1 to 2 min in removing the black deposit from the emulsion. (5) Fixing is carried out in a solution of the following composition: crystalline sodium thiosulfate, 400 gm; potassium metabisulfite (5 ml of a 5 % solution to attain a pH of the fixing solution of 5.8); water, (approximately) 1 liter. The plates are fixed at a temperature of 5°-7°C for 90-120 min, and the fixer is changed every 30 min. (6) Washing is by a running-water method of decantation at 9°-10°C for 120 min. (7) The washing is finished in a 2 % solution of glycerin at a temperature of 9°-10°C for 5-10 min. (8) Drying is in circulating air with a gradual decrease of humidity. Processing is carried out under 25-watt yellow-green lighting with a light filter that is designated No. 117 or 118.

The use of (*a*) citric acid buffer in place of hard-to-dissolve boric acid, (*b*) the accurate control of solution hydrogen ion concentrations, (*c*) the use of potassium metabisulfite in the fixer, (*d*) the employment of no restrainer in the developer, and (*e*) the great variety of temperatures for which close control is demanded, are notable features of the processing. This procedure requires attention by a skilled technician. It seems preferable to the writer to streamline the processing and make it less of a custom operation; to use factory-mixed and standardized chemicals; to carry out the whole operation at room temperature; and to depend on accurately proportioned pure chemicals for correct pH values. It also appears probable that if the common ion effect were utilized to restrain the development by adding KBr to the developer, and if it were used diluted for a longer development time, that uniform development of much greater thicknesses of emulsion than 50 μ could be carried out by the procedure recommended.

For 100 μ emulsions Ilford Ltd. recommended soaking in distilled water at 20°C for 20 min and developing in ID-19 diluted 1 part of developer to 9 of water at 20°C with agitation. For G.5, the development is 60-70 min; for C.2 and E.1, 15-35 min. The plates are transferred to 1 % acetic acid as a stop bath at 20°C for 10 min. The surface fog is removed at this stage by gentle rubbing with cotton wool. Fixing is in 30 % plain hypo (2 hr with agitation). Washing is for 4 hr with agitation. For layers thicker than 100 μ they recommend the "Brussels" amidol development (see below).

Development in D-19 as described above is not suitable for obtaining the maximum discrimination in processing, because physical development, by which silver from the solution is plated out on the developing grains, occurs when D-19 developer is used. Thin emulsion plates may be employed usefully in some studies requiring that particles of different masses, charges, and rates of energy loss be separated by observations on the track. Then special processing is valuable. For example, when the tracks of protons or alpha particles must be seen in the presence of a heavy electron background, or when it is required that particles of nearly the same masses or charges be separated, special procedures, as described in Chapter 2, may be introduced. Discriminating development, combined with fine grain emulsions and controlled latent image weakening, can give improved ionization resolution.

The Eastman Kodak Company recommends no dilution of D-19 developer for emulsions of 50 μ or less (B 52). For moderate development of 25-50 μ emulsion, only 4 min in the developer is suggested. Such a procedure seems, however, certain to lead to nonuniformity of development.

5.3 Mounting Pellicles on Glass

It is usual to carry out the development and fixing of the pellicles of an emulsion stack after they are mounted on the glass plates from which they receive support. Unmounted pellicles can also be processed as described in Section 5.9. Mounting must be carried out in such a manner that the strength of the pellicle is maintained until it is firmly bonded to the glass, and prolonged soaking of the pellicle and/or high soaking temperatures are therefore proscribed.

The glass recommended for mounting is obtainable at present from Ilford Ltd. The Ilford glass is coated on both sides with a very thin, hard gelatinlike material. This makes a good bond to the glass, and the pellicle usually adheres strongly to this coating.

The pellicles will not stick well to an ordinary glass surface. It is said, but it has not been proven to the author's satisfaction, that the glass has merely to be thoroughly *clean* for good adhesion. A slight etching with hydrofluoric acid might help. It is simplest in any case to obtain from Ilford Ltd., glass plates specially prepared for mounting. To these the pellicles usually can be made to adhere so strongly that if the emulsion is dried and contracts strongly, the emulsion-glass bond does not rupture, but instead the surface layer of glass is pulled away.

Several methods of mounting stripped pellicles are described below that have been extensively employed by the writer. Other methods have also been used. For various reasons no method yet devised can be recommended without considerable reservation.

Method (*a*). The glass plate is immersed in water at 0°-5°C containing 1.5 % gelatin, 0.5 % glycerin, and 0.05 % Kodak wetting agent. A few pieces of ice floating in the water suffice to keep its temperature low.

Plates are kept clean and are handled by the edges to keep them free of such surface contamination as greasy fingerprints. The glass is wiped with a chamois skin while submerged so that it is uniformly wetted and free of adhering bubbles. After its glossy surface has been inspected to eliminate foreign matter and imperfections, the pellicle is immersed in the cold water for 30-60 sec, depending on its dryness. This surface is then brought against the glass while both remain immersed. While viewing the operation through the glass, the pellicle is brought into the desired position (using the spread fingers of both hands) and then pulled snugly against the glass. If carried out while the pellicle and glass are immersed, there should be no bubbles of air trapped between pellicle and glass, but in any case, such bubbles can be squeezed out by finger pressure. The pellicle and glass are then lifted together out of the water and rolled through a large clothes wringer with smooth soft rubber rollers. The wringer pressure and the time of immersion are adjusted for each batch of pellicles. The time of immersion for a given stack can be determined using the first few pellicles, and should remain unchanged thereafter. For average emulsion, Birge *et al.* (B-W 54) recommend 7 lb of roller force per linear inch of contact with the pellicle. The pressure must be sufficient, and the pellicle pliable enough so that it adheres uniformly all over its area. Then no bubbles will be visible through the glass. At the same time, the pellicle must retain sufficient strength so that it is not distorted in the mounting operation. These conflicting requirements can usually be satisfactorily compromised if just the surface of the pellicle is wetted and if the temperature is kept low. When the pellicle appears firmly attached, excess water should be taken up from the surface with a chamois skin or photographic blotting paper, and it should then be placed in the rack in which it is to be developed (if the developing procedure outlined below is followed). The racks containing the mounted pellicles should be placed in a refrigerator adjusted to ≈ 5°C for a period of at least 2 and preferably 4 days for the bond between the pellicle and glass to become firmly established. The time must be sufficient for water trapped between glass and pellicle to diffuse away. The low temperature is necessary to prevent eradication of the latent image during this setting period. For emulsion with a less stable latent image than G.5 even 2 days may be too long.

In a large stack of 240 G.5 pellicles, 9 × 12 inches in size, which the writer mounted by this method, no eradication was observed and the adherence was perfect on most plates. About one plate out of five in the dilution stage developed a blister. The areas of the blisters averaged less than 1 cm^2.

The reduction of blisters has been stated by the Bristol group (BDVP 57) to be directly correlated with the setting time.

Method (*b*). The method described above is capable of producing good adherence of the pellicles to the glass, but serious discomfort and inconvenience may be entailed in the low water temperature required to keep the pellicle strong when it is wet.

Another method, developed by the Naval Research Laboratory (S 58.1, SSO 54), has been employed by the writer's research group, and was more popular, partly because immersion of the pellicles in cold water is avoided. The mounting procedure is as follows: the glass is prepared by washing it in water containing 0.1 % of aerosol (a dioctyl ester of sodium sulfosuccinic acid) at room temperature. The surface of the glass is carefully wiped to produce a uniformly clean wet surface. A second solution is prepared by adding 20 gm of gelatin to a liter of the first solution, and dissolving it by heating. It is then filtered and chilled so that it is about as thick as house paint, and can be applied with a paint brush in the same manner. While the dull surface of the pellicle is kept dry, the shiny surface is "painted" with the gelatin solution at room temperature. The painted surface is then applied to the wet, clean glass surface being careful to avoid trapping of air.

The initial contact of the pellicle with the glass is brought about along a line which moves across the glass as the pellicle is pressed against it. The other surface of the pellicle may be kept dry by covering it with a thin foil of such a material as Parafilm, perhaps held down at the periphery of the pellicle by a small amount of the gelatin solution. After the pellicle is in contact with the glass it may be either immediately rolled on as in method (*a*) with a thin foil of paraffin sheet protecting the surface, or it may be placed under a weight for a few minutes before rolling. The latter procedure sometimes causes trouble as the foils may become difficult to remove from the surface.

Method (*b*) wets the pellicle less than method (*a*) and also requires less time to set. After 15 hr or more of drying at room temperature the pellicles are bonded to the glass and may be developed. To preserve the latent image, the setting should be carried out under refrigeration.

Method (*c*) (*Heckman method*). Good pellicle mounting requires wetting it only enough to cause it to adhere to the glass. Additional moisture trapped between the pellicle and the glass must diffuse away, and a long setting time may be required to produce a good bond. In the Lawrence Radiation Laboratory it has been attempted recently to minimize the wetting by using an alcohol-water mixture. Current practice is as follows: the glass is first cleaned by washing in distilled water containing aerosol. If not checked earlier, the glass is then inspected with light at grazing incidence for evidence of surface nonuniformity, perhaps caused by colonies of organisms, and any imperfect pieces are discarded.

The following stock solution has previously been prepared: 20 gm of Keystone gelatin No. 2191 (American Agricultural Chemical Co., P.O. Drawer 2458, Detroit 31, Michigan) is added to about seven times its volume of water at 22°C and allowed to soak for 1 hr. Then it is heated in a water bath to 50°C. To it is added a solution at room temperature prepared as follows: 1 liter of water, 10 ml of glycerin, 5 ml of 1 % aerosol solution (too much aerosol causes frothing), and 430 ml of alcohol. The gelatin specified does not precipitate with this quantity of alcohol.

This solution is diluted by a factor of four and the glass plates are immersed in it prior to mounting the pellicles.

The pellicle is then placed for 1 min in a water solution of 60-75 % ethyl alcohol, depending on the pellicle dryness. The time of immersion, however, does not appear to be critical.

When alcohol is put into water in a large concentration, dissolved air in the water precipitates out, causing numerous small bubbles. These can be harmful if present in the somewhat viscous solution used in bonding pellicles to glass. Boiled water may be used or the bubbles allowed to rise and dissipate before dipping emulsion into the solution.

In preparation for mounting the pellicle on the glass, a smooth foam-rubber sheet (≈ 2 inches thick) is first covered with a layer of absorbent paper and then a layer of paraffin foil that is slightly larger than the pellicle. One lays the moistened pellicle on the paraffin foil with the mounting surface up. The stock gel solution is diluted with an equal volume of water. With a syringe, about 0.1 ml/cm of this gel solution is put as a bead along one edge of the pellicle. The glass plate is then put on it slightly inclined so that there is a line of contact with the liquid. The line extends at first only along the beaded edge. The plate is then pressed down firmly, causing the line of contact and the excess liquid to move across the pellicle and to extrude along the opposite edge. The mounter can depress the emulsion at a point on the exit edge to establish the last point of contact which prevents the trapping of any excess liquid and air bubbles. The operation is viewed through the glass. A weight can be placed on the glass for 1 or 2 min before the glass and emulsion is placed on a firm surface and hand rolled to complete the bonding. The absorbent paper is removed, and the finished plates with their paraffin layers still in place are stacked under a weight of 1/2 lb per square inch for about 1 hr. The plates are then unstacked, the paraffin foils stripped off, and placed in racks for further drying and then processing. The total drying time after stripping can be as little as 2 hr because of the sparing use of water in the mounting operation. By this method, the estimated mounting rate is thirty large pellicles per hour by a crew of four people.

Of course, many variations of the mounting procedures yield more or less satisfactory results, and every part of the operation is subject to change. The highly qualified group in Brussels, for example, follow this procedure: the sticking-down solution temperature is maintained at 23°C so that the viscosity remains constant. It consists of 1.5 % gelatin, 0.5 % glycerin, and 0.5 % disinfectant in distilled water. The drying period is 60 hr at 58 % R.H. and at a temperature of 13°C.

Rolling at Milan is by a motor-driven rubber roller to obtain a constant and controllable pressure (IO 58). A sheet of plastic is placed on the emulsion during the process.

Englehardt *et al.* (EHK 60) have described the mounting procedure for NIKFI 400 μ emulsion when no special glass for mounting is available. After thorough cleaning, the glass plates are prepared by immersing them three times in the following solution: 4 gm of gelatin, 10 gm of sodium silicate, 0.75 gm of chrome alum, 0.5 gm of thymol, 60 ml of ethyl alcohol and 1 liter of distilled water. It was stated, however, that henceforth NIKFI would supply special glass for mounting.

Before mounting, the surface of the pellicle must be rubbed down with an alcohol-water solution of 25-30 % concentration. A surface layer of 3 or 4 μ is removed. This was stated to be necessary to avoid the formation of a black sediment on the surface. The pellicle is mounted by immersing it and the glass in the solution given above. The pellicle is moved back and forth on the glass until it sticks. This takes 3-5 sec. The plate is then immediately taken out of the solution, the surface cleaned, and then rolled by a specially designed machine roller. The emulsion is dried for 2 hr while chilling from room temperature to 3°C. This type of mounting gave few blisters. Only 0.038 % of the plate area was damaged by blistering.

A frequent trouble with the mounting occurs during fixing and dilution (see below). Then if any points of poor bonding between pellicle and the coating on the glass exist, a blister may be formed between the glass and the pellicle. These blisters may sometimes become very serious. No one cause of blisters can be blamed in all cases, but the following are suspected causes: (*a*) a pocket of water, air or other foreign matter trapped between pellicle and glass; (*b*) greasy fingerprints on pellicle or glass; (*c*) bacterial action on the glass coating (this is suspected from the appearance of the nonadhering areas and because old glass seems to be worse for blisters); (*d*) imperfect glass or pellicle surfaces; (*e*) insufficient time allowed for a good bond to form. If a blister of water is left after mounting this will diffuse into the pellicle, and given sufficient time, the area will dry out enough to form a good bond.

It has been noticed that in mounting pellicles at least a narrow margin of glass surface should be left around the pellicle. If the pellicle is flush with the edge, the glass is easily ruptured, and with moderate drying the pellicle will pull up at the edge tearing up a glass layer as shown in Fig. (3.3.1).

In criticism of the mounting procedure, the treatment received by the pellicle is still much too rough. Serious distortions are introduced by a wringer and it should be eliminated. In the writer's experience with uniform weighting of the emulsion, difficulty was experienced with fading of the latent image because too much water was trapped with the pellicle. The question has not been attacked systematically, however, and it seems likely that the methods of mounting can be greatly improved.

Mounting of pellicles on polystyrene has been described by Dahl-Jensen (D 60.3). The polystyrene is dipped in fuming sulfuric acid. Then the polystyrene sheet is washed in more and more diluted acid and finally in distilled water. The wet polystyrene after washing will stick permanently to dry emulsion.

Another possibility is to cement down the dry pellicle to glass with a bonding material that is unaffected by the solutions. Araldite has been suggested (D 60.3) for this purpose.

5.4 Equipment for Developing Large Emulsion Stacks

There are many possible good processing systems, all of which cannot be discussed in detail; a fairly complete description will be given only of the equipment needed to carry out the procedure recommended here. Other processing methods, however, require many of the same pieces of apparatus. The recommended equipment has been found by experience to meet the requirements of simplicity and practicality, and at the same time to yield results of high quality.

It is necessary to adopt a scale factor determined by the maximum volume of emulsion to be processed in one batch; to be specific let us assume that this is 12 liters (perhaps 300 sheets of 9 × 12 inch emulsion 600 μ thick). The dimensions can be scaled according to one's needs; in the writer's experience, progressively larger facilities seem always to be required. Actually a great deal of flexibility is available if sufficiently large tanks are provided initially.

The plates are assumed to be held in racks somewhat like the one shown in Fig. 5.4.1. If the spacing of the plates is 1/4 inch (which is about as close as it is feasible to pack them), then the racks can be contained in a tank of about 50-gal capacity.

In order to reduce the handling of the plates a system is adopted whereby they are not transferred from tank to tank; the various solutions are introduced, one after the other, into a single main tank containing the plates.

The main tank should have the following features: (*a*) Its temperature should be controlled by a heat-exchanger unit or by coils in the walls through which liquid controlling the tank temperature can be pumped. (*b*) The tank should be heat-insulated. (*c*) It should have a light-tight cover. (*d*) It should have a large bottom drain located in a depression so one can quickly and completely empty the tank. (*e*) Provision for flowing liquid through the tank with a uniform horizontal velocity everywhere in the tank. This can be accomplished by providing multiple inlets and outlets and a circulating pump.

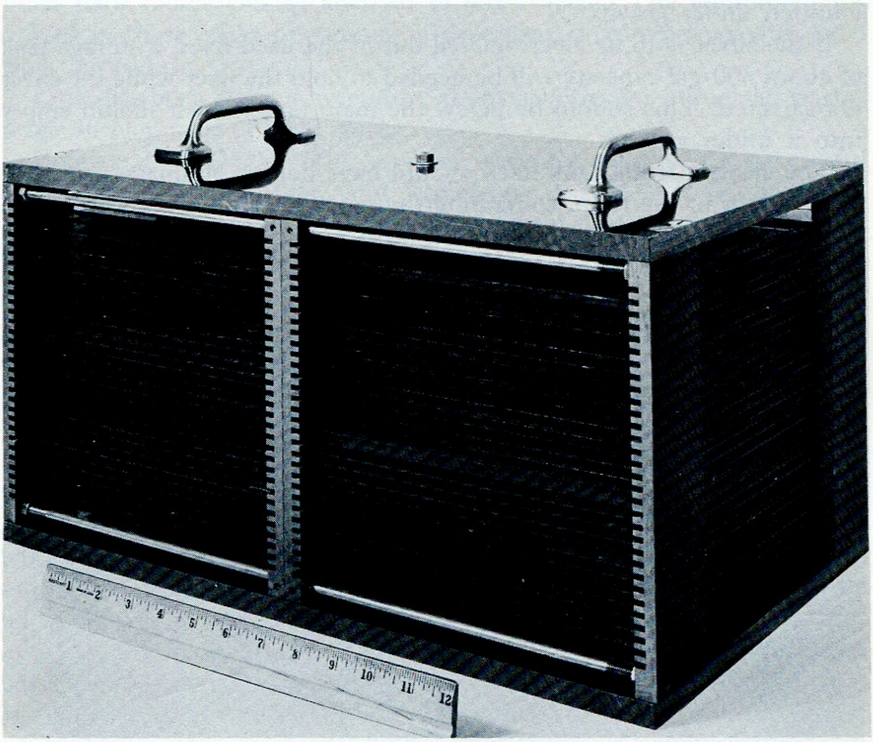

FIG. 5.4.1. Rack in which pellicles mounted on glass are placed for processing (IDLRL).

Solution preparation and storage tanks are also required. Each needs all the features of the main tank except (*c*) and (*e*). They are preferably mounted above the main tank so the main tank can be filled by gravity.

The main tank and the solution tank must be provided with temperature-sensing elements for measurement and control of the temperature as well as external liquid-level gauges and overflow-prevention devices.

A supply of distilled water for making up solutions should be on hand. One way to insure a supply when it is needed is to keep a few hundred gallons in a storage tank which need not be temperature-controlled, but should be provided with a liquid level gauge, and overflow protection. The storage tank should be at a convenient height to be emptied under gravity.

If the silver is to be electroplated out of the used fixer, a storage tank of about 300-gal capacity will be needed to hold the fixer while the silver is recovered. This should be below the main tank, which should empty into it under gravity.

The electroplating tank itself should have bottom drainage provided.

The floor of the room containing the tanks should be waterproof and slipproof, and should be provided with a drain at its lowest point. Spilling on the floor where large quantities of liquid are handled is inevitable.

In addition a 50-gal power mixer is rather essential. It is important that the mixer not only be able to mix the solution, but that it be provided with a fast pump which will deliver the solution to the elevated storage tanks or to the main tank.

Circulation of the fluid in the main tank is accomplished by a stainless steel or plastic pump that withdraws liquid at several points on one side of the tank and introduces it again at many points on the other side.

A pump is required to transport solutions from the main tank to the storage tanks or between other tanks. One delivering at least 10 gal per minute is best for this purpose. Pumps that have been proven to be corrosion-resistant and generally trouble-free are essential.

A hot water supply and refrigeration capacity sufficient to change the temperature of 50 gal of solution by 20°C in about 1 hr is desirable. It is necessary that it be possible to maintain a temperature below 5°C, but if a supply of distilled-water ice is kept on hand to lower solution temperatures quickly, the peak refrigeration capacity need not be very high. To speed the heating of solutions, an electrical immersion heater encased in stainless steel can be employed.

One does not attempt to produce enough distilled water for the dilution and washing of the emulsion unless the local water cannot be processed to make it satisfactory. Ordinarily filtration and ion-exchange resin purification is sufficient. For the washing and dilution phase of the processing, the solution tank can be used to chill the wash water.

The processing room should, of course, provide complete exclusion of outside light, and access to the room preferably should be through double doors or by some other means so that people can enter and leave freely without letting light into the room.

Lights, using recommended or tested safelight filters, should be installed at points where they are needed. If the developing room is large enough, it can also be used for the other operations, initial assembly of the stack, density measurement, gridding, and mounting. If not, other dark rooms can be adapted to one's needs. The equipment involved in these other operations is described in connection with each such procedure.

Little has been written heretofore about facilities for immersed warm-stage development of large stacks.

Hooper *et al.* (HDN 60), however, recently described a successful installation at Copenhagen, and Armstrong *et al.* (AFR 58) described another at Los Alamos. An ordinary household refrigerator was adapted by them, as shown in Fig. 5.4.2, to the development of emulsion stacks.

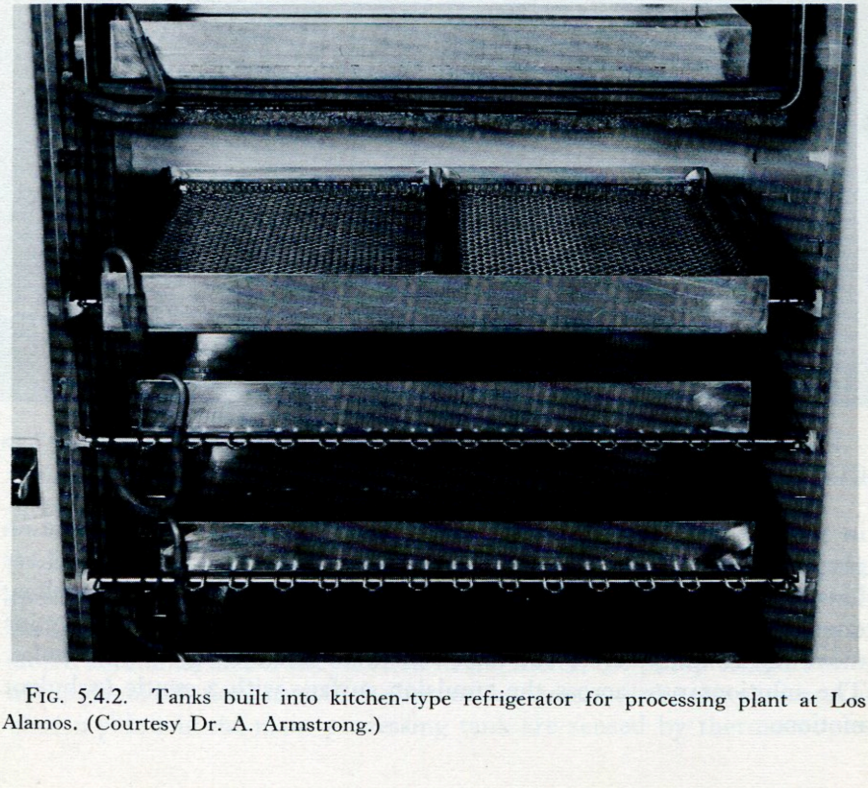

FIG. 5.4.2. Tanks built into kitchen-type refrigerator for processing plant at Los Alamos. (Courtesy Dr. A. Armstrong.)

In the following are given details of immersed warm-stage processing facilities at Berkeley (B 60).

In this plant it is convenient to process at once 12 liters of emulsion. This capacity can, if necessary, be raised somewhat by making space economies.

The facility is housed in a room with a floor area of 90 square ft. The main tank is shown in Fig. 5.4.3. All the processing solutions pass in and out of it—the plates are not moved. The emulsion, mounted on glass, is in racks, such as the one shown in Fig. 5.4.1. The plates are level and spaced 1/4 inch apart. Numerous small holes in pipes along one wall admit circulating fixer and wash water to the tank. The liquids move horizontally across the tank and drain out through similar holes. The solutions pass across the emulsion surface with a gentle turbulent motion.

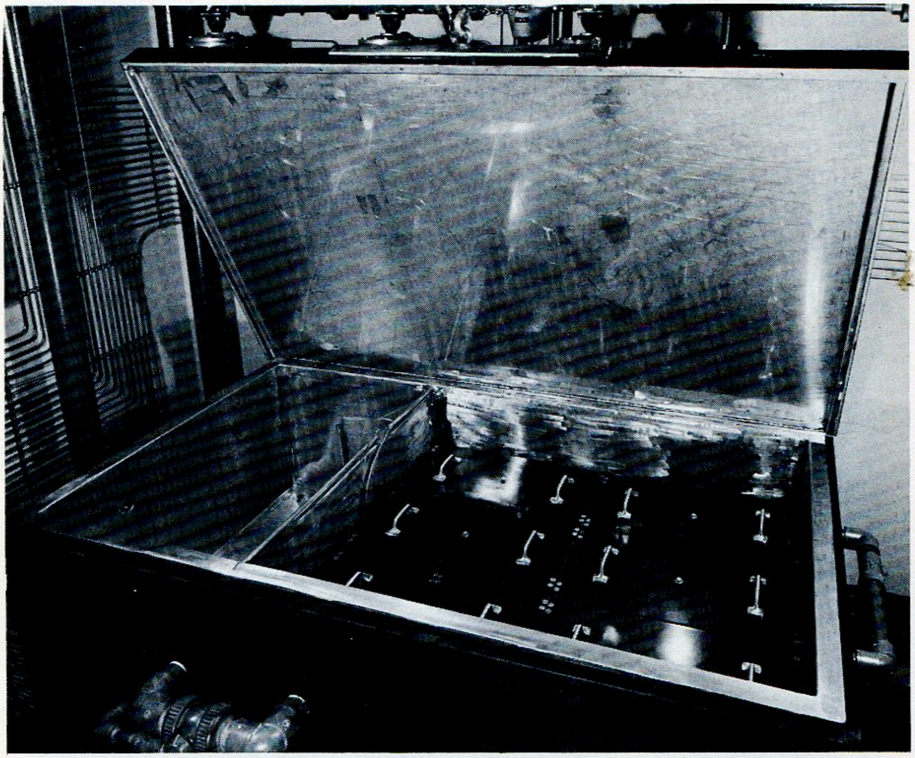

FIG. 5.4.3. View of main processing tank which is used for all the solutions (IDLRL).

The bottom of the tank is heated or chilled. Hot or cold 50 % ethylene glycol solution passes through a "thermal panel" (Dean Products, Inc.) which maintains intimate contact with the bottom by means of a material known as thermomastic. The whole tank is heat-insulated with Styrofoam, and is covered with black Formica. Indicating temperature controllers (Minneapolis Honeywell) operating pneumatic valves maintain the solution temperatures at the desired values. The temperatures of stored solutions, that of the solution in the mixer, the pump temperatures of the glycol solutions used in the heat exchangers, and the temperatures at chosen points in the main processing tank are sensed by thermocouples

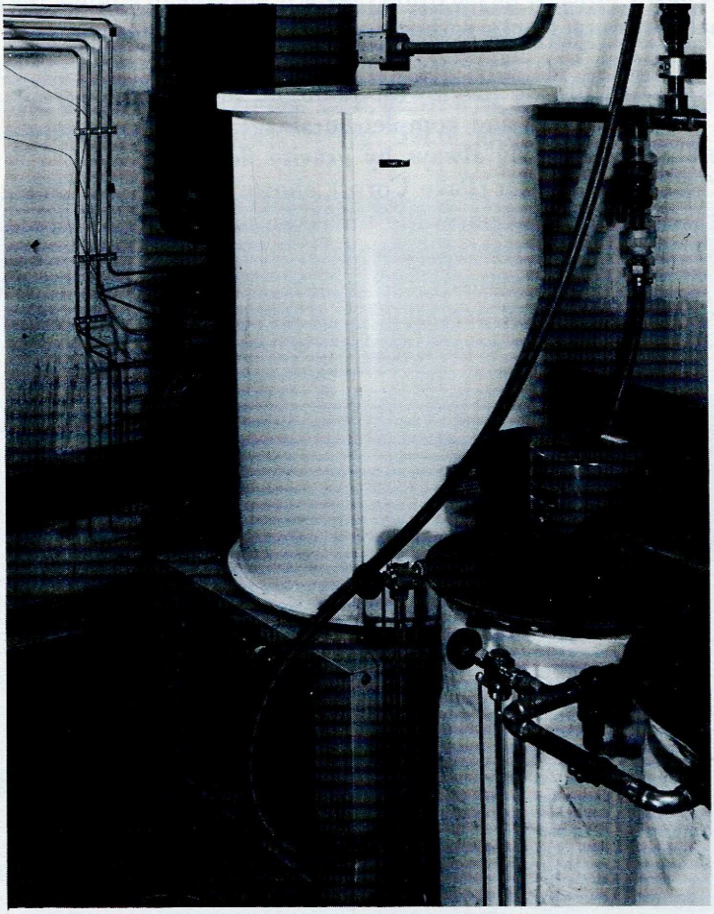

FIG. 5.4.4. Power mixer and pump for preparing solutions and delivering them to storage tanks (IDLRL).

and recorded by a multichannel tape instrument (Leeds and Northrup, Speedomax).

The main processing tank is of heliarc-welded No. 316 stainless steel. It contains a partition dividing it into two parts, one having twice the volume of the other. The partition makes it possible to employ the system for small emulsion stacks without preparing and handling large volumes of the solutions.

A large aperture at the lowest point of the bottom permits quick filling, and also rapid and complete drainage of the main tank. Filling and draining are nearly always by gravity flow.

A 55-gal power mixer (Pako Corp.), shown in Fig. 5.4.4, is equipped with a pump that transports the mixed solution to one of the storage tanks or to the main processing tank.

A 150-gal tank contains a reserve of distilled water prepared at a maximum rate of 9 gal per hour by a still (Precision Scientific Company). This equipment is on the roof of the building.

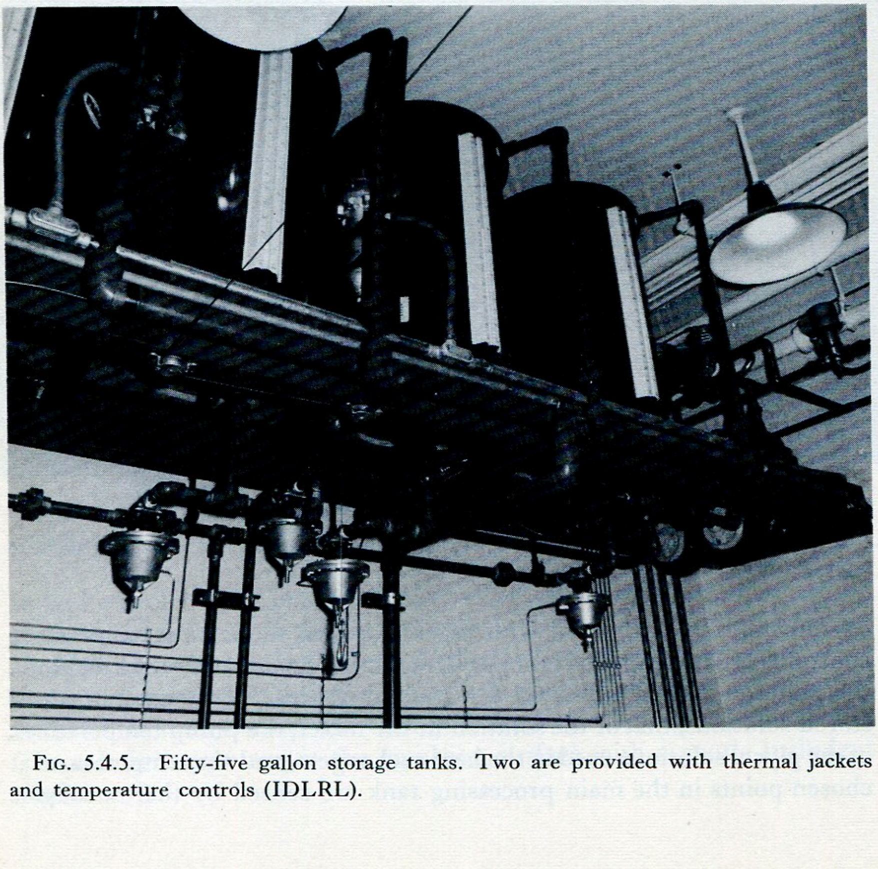

FIG. 5.4.5. Fifty-five gallon storage tanks. Two are provided with thermal jackets and temperature controls (IDLRL).

Above the main tank there are four 55-gal storage tanks (Pako Corp.) shown in Fig. 5.4.5. All have liquid level indicators, and can be filled and emptied by remote control. Two can be chilled.

One also can be heated. The two tanks with thermal control are heat-insulated. All have hopper bottoms for complete drainage. The tubing used to carry the solutions, and the valves (Chemtrol) are of polyvinyl chloride with teflon sealing rings. The main tank, the pumps, and some pneumatic valves are of stainless steel, grade 316, with teflon diaphragms.

On a control panel shown in Fig. 5.4.6 pilot lights indicate which of the electrically controlled valves are open, and the adjacent switch operates each indicated valve. While electrically controlled, many of the valves are pneumatically operated. Others are solenoid valves (Automatic Switch Company).

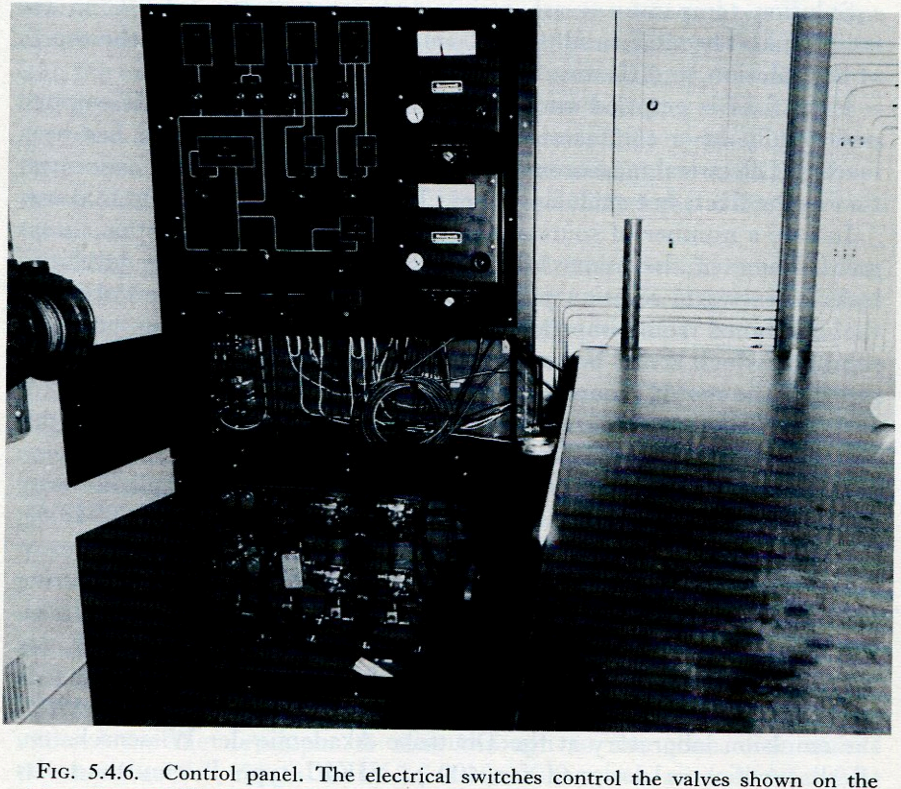

FIG. 5.4.6. Control panel. The electrical switches control the valves shown on the piping diagram. The meters indicate solution temperatures and maintain them at predetermined values through control mechanisms (IDLRL).

For washing, tap water is used but it is demineralized. A commercial unit (Barnstead) of 8 gal per hour capacity is installed for this purpose. A refrigerating unit (Brunner 1 hp) is installed on the roof with a heat exchanger. A 50 % ethylene glycol solution is circulated through heat-insulated pipes from this heat exchanger to other heat exchangers in the processing room. These (American Radiator and Standard Sanitary Corporation) are of stainless steel. Local heating of solutions is by means of electricity. Both turbine pumps (Pacific Pumping Company) and combined rotor-impeller pumps (Corley Magnaflow) of stainless steel are used.

Solution temperatures are maintained constant to ± 1/2°C in the main tank. The time to fill it is 10 min or less depending on the source of the solution, and the time to drain it is 1.5 min.

Used fixer is emptied into an external 300-gal tank which is emptied periodically by a commercial silver-recovery firm. Provision has been made for electrical measurement and control of the silver ion concentration in the fixer, but such equipment has not actually been put into use.

In use, a number of sources of trouble have developed in this equipment. Some of the joints in the polyvinyl chloride piping developed leaks when warm solution passed through them. It was found also that the combined rotor-impeller pumps are not suitable for pumping solutions which are to be temperature-controlled because all the energy put into the motor pump is carried away in the liquid as heat.

A detailed, step-by-step manual of procedure for operating this system of valves, pumps, etc., of course is necessary.

A number of descriptions of representative processing plants using a dry, warm stage have been given (DOV 51, BDO 51, IO 58, BMS 58, SSO 54, SSR 60, N 61, S 58.1).

Dahl-Jensen (D 60.3) has collected the summarized current processing procedures used in the European laboratories of Copenhagen, Bristol, Durham, Lausanne, CERN, and Milan. Most of these use the dry warm stage, their facilities having been built several years ago.

Recently, too, Engelhardt, Hauser, and Krecker (EHK 60) described the emulsion laboratory at the Deutsche Akademie der Wissenschaften (Berlin) which is designed for 400 μ NIKFI type R emulsions. Its capacity is 3.5 liters of emulsion in 5 days using essentially the dry, warm stage procedure of Dilworth *et al.* (DOV 51). The development is carried out with batches of 0.7 liters.

Further information on the processing of NIKFI emulsions is contained in a report by Uvarova *et al.* (UKMR 58) and by Samoilovich and Barkov (SB 60) who have given rather complete details of the processing equipment and procedures.