Chapter III

Detailed Study of the Emulsion Technique as applied to (ν,γ) interactions.

<table>
<thead>
<tr>
<th>Section</th>
<th>Contents</th>
<th>Page No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1</td>
<td>Processing technique as applied to (ν,γ) reactions.</td>
<td>61-68</td>
</tr>
<tr>
<td>3.2</td>
<td>Study of track structures for distinguishing various particles.</td>
<td>69-83</td>
</tr>
<tr>
<td>3.3</td>
<td>Effect of ammonium salts over the clearing time, shrinkage, and the grain size.</td>
<td>84-94</td>
</tr>
</tbody>
</table>
Thorough discussion of the processing techniques may be found in several review articles. Shapiro (S1) (1941) gave a complete summary of the emulsion techniques and experiments up to 1941. Demers (D1) (1947) gave a detailed account of his experiments in emulsion making, and properties of his emulsions together with a theory for silver grain formation. A comprehensive discussion on the ionization mechanism and the properties of the then commercially available emulsions was given by Webb (W1) (1952). In addition to this the work on processing nuclear emulsions of various thicknesses has been summed up to 1951 and presented by Belser (B1) (1952) in a review article. Among other useful references are the book "Nuclear Physics in Photography by Powell and Occhialini (P1) (1947), an article and a book by Yagoda (Y1) (1948), and also an article by Rotblat (R1) (1950).

The temperature development technique of Dilworth Occhialini and Payne (D2) (1948), which is based on the difference between the temperature coefficients of diffusion and development has received wide usage for thick emulsions.

Nuclear emulsion consists of a concentrated dispersion of silver halide granules of roughly spherical shape in the matrix of gelatine. The size of these grains ranges between 0.1 to 0.6 microns in diameter, and the grains are distributed at random, separated by thin layers of water permeable gelatine. In traversing this heterogeneous medium, the ionizing particles intersect the grains which get reduced to metallic silver during photographic development. Grains which are not activated are removed during fixation, leaving voids of comparable volume in the wet gelatine. On drying,
the gelatine layers contract in thickness owing to the coalescence of the voids left by the extracted grains. The residual row of silver grains outlining the path of the particle is a measure of its range in the original emulsion. Medium being discontinuous the particle may originate and terminate its trajectory in the gelatine, and its recorded range, as defined by the terminating grains of silver, will tend to be somewhat smaller than the actual distance traversed.

The ability to differentiate the tracks of different particles makes the emulsion an important tool in modern nuclear research; and the emulsion processing has to play a prominent role in this. Since all the grains are not of equal sensitivity and require different degrees of energy expenditure for latent image formation, the observation of the mean grain spacing along the developed path track, permits the differentiation of the nuclear particles.

The quantitative aspect of the variation in grain spacing as a function of the ionizing property of the charged particles are fully described in publication by Pignedoli (P2) (1941), Lovera (L1) (1945), Demers (D1) (1945), and Feld (F1) (1948). The more recent investigations of the Bristol group (L2) (1947) show that log n is a linear function of log L, when n is the total number of grains in the track and L corresponds to track length. The experimental data for alpha particles, tritons, protons, and mesons.
have been found to lie on parallel lines. The relationship among
the total number of grains in a track \( n \) is related to the mass
of the particle \( M \) and its recorded range \( L \) by \( n = K Z (2K - 2/3)x \)
\( M (1 - K)^{L/K} \), where \( Z \) is the particle charge, \( K \) is a constant
for a particular particle and the coefficient \( K \) is dependent
on the emulsion characteristics and mode of development. Thus
the mode of development has a prominent role to play in the
particle distinction.

Selective processing has found its application in the
discrimination of alpha tracks from fission fragment and proton
tracks, in the Ilford \( C_2 \) plates with which we are concerned at
the moment. Deuteron and triton tracks are very scarce because
of the large negative \( Q \) values involved in such interaction. Studies
on the action of Oxidants lead Perfilo (F3, P4) (1944, 45) to the
conclusion that the development of all silver bromide grains
receiving only a single impact could be inhibited by oxidation
before development. Perfilo reasoned that a differential oxi-
dation of the emulsion could be arranged for the distinction of
between protons, alpha tracks, and fission fragments. The following
treatment was suggested by him.

<table>
<thead>
<tr>
<th>Percent</th>
<th>Track structure in Ilford ( C_2 ) plates.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>Proton: Paint, Alpha: Distinct, Fission: None</td>
</tr>
<tr>
<td>2.0</td>
<td>Proton: None, Alpha: Light, Fission: Dens</td>
</tr>
<tr>
<td>4.0</td>
<td>Proton: None, Alpha: Very ( \delta ), Fission: Dens</td>
</tr>
</tbody>
</table>
Development in this case is terminated by immersing the plate for 5 minutes in a 1 percent Pot. meta bi sulphite stop bath, and the plates are fixed for 2 hrs. in 30% Sod. thiosulphate solution, washed, and dried.

The investigations of Broda (B2) (1946) showed that the Ilford plates can be desensitized to Χ rays and proton recoils from fast neutrons by bathing the plates for 5 minutes in 1% chromic acid, rinsing with water, and drying prior to neutron exposure. This is one of the devices employed by the author in distinguishing 2 tracks from the proton recoils.

Differentiation between the various tracks can also be secured by using less sensitive emulsions, or by giving the emulsion a weaker development. Stevens (S2)(1948) has shown that by means of low energy para-aminophenol developers fission tracks can be recorded selectively even in emulsions that are electron sensitive. The differentiating power of the emulsion can also be increased by "grain gradation development suggested by Yagoda (Y1) (1948)". The method consists of developing the plate from 10 to 20 minutes at 60° to 70° F in a freshly prepared solution containing 0.5g hydroquinone, 10g. Na₂SO₃, 0.5 g. KBr and 1.0 gms. KOH per lit. of distilled water. This method has been reported by Yagoda to give a very clear discrimination between, alpha, triton and recoil proton tracks. In 50 micron.
plates the development is complete throughout the entire depth, and the fog background is low. By increasing the development time and temperature the method can be successfully employed to secure uniform development in thicker plates.

In view of the diverse formulae presented by various workers in the field for achieving various results, it was found necessary to go into the details regarding the processing of various types of nuclear emulsions; because a study of this type helps in any study in the field of nuclear reactions, using nuclear plates as detectors of the resulting products.

Keeping in view the application of such a study to (γ, δ) cross-section measurements, the processing technique was studied in the following three stages:

1. A general study of the emulsion processing technique was undertaken using the plates of various types and thickness to adopt the formulae and procedures most suited for (γ, δ) study with respect to the climatic conditions of this laboratory. In addition to this a study of the shrinkage factor in C2 Ilford emulsions, and also a study of the fading of tracks was made.

2. A detailed study of track and grain structures in Ilford C2 plates was made to have a better differentiation of the tracks. Elimination of fogging (which is a big handicap in the experiments of the type, and also a study of the background eradication in nuclear emulsions was undertaken.
3. A study of the effect of the addition of various ammonium salts to the usual hypo solution on the clearing time of the various types of emulsions was undertaken, so as to avoid the track distortion which results due to prolonged fixation in (nitrate) study.

**Section 3.1.**

**Processing technique as applied to (nitrate) interactions:**

Before starting with the development, it is necessary to soak the emulsion in water, so that the penetration of the developer may become easier and more rapid. For this, the temperature and time limits suggested by various workers were used, and the following time and temperature limits were found to give the best results:

<table>
<thead>
<tr>
<th>Emulsion thickness</th>
<th>Time</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.5 hrs</td>
<td>2°C</td>
</tr>
<tr>
<td>200</td>
<td>0.75 &quot;</td>
<td>4°C</td>
</tr>
<tr>
<td>400</td>
<td>1.25 &quot;</td>
<td>6°C</td>
</tr>
</tbody>
</table>

To facilitate penetration, presoaking in distilled water with or without the addition of the wetting agent is frequently made use of. This acts to swell the gelatine, permitting more rapid diffusion of the developer. It, however, does not affect the actual development as with the alkaline developments (Dilworth (D4) (1948), Mortier and Vermaessen (M1) (1948), Piccioto (P5-7) (1949). The suitable temperature for the penetration has been found to be 4°C. Below this temperature, the penetration time was found to be too long, and above it, the rate of developer penetration increases less rapidly than its activity.
Two degrees of development were found possible for thin emulsions. Moderate development was found useful when grain densities of comparatively dense tracks (e.g. protons and tracks of several Mev energy) are to be measured. Since in this case, it is essential that the grains be discrete, moderate development is preferred. This has an additional advantage of great reduction in the fog density. Strong development, on the other hand, although accompanied by an increased fog background, permits a full utilization of the emulsion sensitivity, and the heavy ionizing particles appear as solid columns of silver grains. Series of development tests were conducted to determine the development time giving the most preferred combination of background and track densities. The results thus obtained are indicated below:

<table>
<thead>
<tr>
<th>Thickness</th>
<th>Procedure</th>
<th>Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>(moderate development)</td>
<td>10 min.</td>
</tr>
<tr>
<td>100</td>
<td>(strong development)</td>
<td>40 min.</td>
</tr>
</tbody>
</table>

For thick emulsions, the two developer solution method as suggested by Blau and Defilice (B3)(1948) is found to give the best results to secure the even development. The first
contains the developing agent without any alkali, permitting the diffusion of the developer into the emulsion without any appreciable amount of actual development occurring. The second bath containing an excess of alkali permits the development to take place.

This method requires that the velocity of the travel of a P - H change should exceed that of the developer itself; a condition which is not actually satisfied. Also, up to 400 micron thickness, the two bath method eliminates any danger of reticulation.

The following modified formula was found to give the best results in the case of thick emulsions so far as two-bath development is concerned:

Sol. A

Elon 1 gm
Sod. sulphite 20 gms.
Hydroquinone 3.5 gms.
Pot. Bromide 2.0 gms.
Distilled water 2 litres.
Sol. B.

Stock Eastman D19  ...  400 c.c.
Distilled water  ...  1600 c.c.
Sod. carbonate  ...  12 gms.

For 200 micron plates the following single solution development formula was found to give the best results. It is the Brussels formula slightly modified by the author:

Sod. sulphite  ...  36 gms.
Pot. Bromide  ...  0.8 gms.
Amidol  ...  2.8 gms.
Boric Acid  ...  12 gms.
Water  ...  1 lit.

Penetration time needed for various developers:

The penetration time in minutes of presoaked and non-presoaked emulsions was investigated at 18°C, and the results obtained are tabulated as under:

<table>
<thead>
<tr>
<th>Developer</th>
<th>Penetration time in min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Azol 100 μm</td>
<td>Presoaked 4.5</td>
</tr>
<tr>
<td>Azol 200 μm</td>
<td>Presoaked 16</td>
</tr>
<tr>
<td>Azol 400 μm</td>
<td>Presoaked 38</td>
</tr>
</tbody>
</table>

D-19

| Presoaked 3.0 | Non-presoaked 4.5 |
| 8.5 | 10.5 |
| 21 | 37 |

Amidol

| Presoaked 1.5 | Non-presoaked 3.0 |
| 5.0 | 9.0 |
| 12 | 20 |

Amidol bisulphite

<table>
<thead>
<tr>
<th>Presoaked</th>
<th>Non-presoaked</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Processing formulae of thick and thin emulsions:
The following processing formulae were found to be most suitable for 100, 200 and 400 micron plates.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Temperature</th>
<th>100 μm</th>
<th>200 μm</th>
<th>400 μm</th>
<th>Time in min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Presoaking</td>
<td>4°C</td>
<td>30</td>
<td>40</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5°C</td>
<td>25</td>
<td>35</td>
<td>90</td>
<td></td>
</tr>
<tr>
<td>Penetration of the cold developer</td>
<td>4°C</td>
<td>30</td>
<td>40</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5°C</td>
<td>26</td>
<td>36</td>
<td>92</td>
<td></td>
</tr>
<tr>
<td>Warm dry development</td>
<td>18°C</td>
<td>25</td>
<td>40</td>
<td>80</td>
<td></td>
</tr>
<tr>
<td></td>
<td>24°C</td>
<td>20</td>
<td>35</td>
<td>70</td>
<td></td>
</tr>
<tr>
<td>Dry cooling</td>
<td>18°C to 5°C</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>Stop bath acetic acid (0.5%)</td>
<td>5°C</td>
<td>30</td>
<td>45</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>&quot; (1.0%)</td>
<td>5°C</td>
<td>30</td>
<td>45</td>
<td>75</td>
<td></td>
</tr>
<tr>
<td>Fixation (clearing time 50% more)</td>
<td>18°C</td>
<td>3 hrs.</td>
<td>8 hrs.</td>
<td>24 hrs.</td>
<td></td>
</tr>
<tr>
<td>Washing</td>
<td>8°C</td>
<td>3 hrs.</td>
<td>8 hrs.</td>
<td>24 hrs.</td>
<td></td>
</tr>
</tbody>
</table>

The following processing formula was found exclusively suitable for thick plates:

Distilled water 1000 c.c.
Sod. sulphite (anhydrous) 12 gms.
Pot. bromide (10% solution) 8 c.c.
Amidol 3.8 gms.

pH of the developer 7.4
Fixing bath (pH 5.3)
- Distilled water 1000 c.c.
- Sodium thiosulphate 400 gms.
- Sod. bisulphite 10 gms.
- NH₄ Cl 7 gms.

Clearing solution (pH 4.2)
- Distilled water 500 c.c.
- Ammonium acetate 15 gms.
- Citric acid 8 gms.
- Thio-urea 8 gms.

Small quantities of sodium bisulphite and ammonium chloride reduce staining and hasten the fixation of the emulsion. But large concentrations of these ingredients lead to distortion. Further, in the fixing solution, one half of the quantity of hypo was replaced at several intervals, thus avoiding salt concentration shocks. For the same reason, washing was also preceded by a gradual dilution of the fixing solution.

Shrinkage:

The considerable reduction in the thickness of nuclear emulsions after fixing is due to the high concentration of the silver bromide in nuclear emulsions. The ratio of the emulsion thickness before and after fixing were found, and the following results were observed:
Emulsion thickness in μm.

<table>
<thead>
<tr>
<th>Before processing</th>
<th>After processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>96</td>
</tr>
<tr>
<td>200</td>
<td>181</td>
</tr>
<tr>
<td>400</td>
<td>359</td>
</tr>
</tbody>
</table>

Increase in the shrinkage factor of Ilford C2 emulsions:

The increase in the shrinkage factor of the emulsions was investigated at 80% relative humidity, and the results plotted in the Figure ( ).

Drying of the emulsion:

Plates of either thickness were soaked in glycerine solution and dried gently. Rapid drying was avoided, because it produces a skin at the surface which traps the water lower down. This in turn produces stresses at the soft emulsion which produce severe distortions in the tracks. Blowing over the surface was avoided, because it introduces severe distortions, although the temperature can be slightly increased to accelerate the process.

Dilworth (D5)(1951). Edges of the plates usually dry first and prevent surface deformations in the emulsion, which was also maintained as far as possible.

These results have been reported in detail by the author (K1) (1961).
Figure: The variation of the fading coefficient with the time of storage at various temperatures.

Time of storage in hours at 80% relative humidity

Shrinkage factor plotted against the time of storage in hours for Model C.
Study of track structures for distinguishing various particles:

In view of the importance of the study of track characteristics in the distinguishing of the ionizing particles, a study of track parameters was taken up. Further, the utility of nuclear emulsions as a tool for the study of particles of transmutation being very much limited by the background and other processing defects, systematic investigation for the removal of these defects was undertaken, and the work done in this direction by other authors was compared with the work undertaken in this laboratory.

Some work has already been reported on the elimination of development defects by various workers e.g. Yagoda (Y₄) (1948), Liebermann and Barschall (L₃) (1943) and others. The methods used here are somewhat different from the methods used by Fatzer (F₂) (1959) in respect of temperature and Na₂SO₃ concentration.

Ilford C₀ nuclear emulsions of 100, 200 and 400 micron thickness were used and the problem studied under the following heads:

1. Elimination of the development defects introduced by amidol developers
2. Dependence of track and grain structures on the hot stage temperature, hot stage time, and sodium sulphite concentration.
3. Removal of microscopic fog from the plates.
4. Background eradication in nuclear emulsions by the accelerated fading of the latent images.

(1) Elimination of the development defects introduced by the use of Amidol developers:

Decrease in the development temperature gives rise to development defects, which are evidenced by a total or partial destruction of the developed image and also a colouration extending to a considerable depth in the emulsion.

By a series of trials it was found necessary to increase the ratio of sodium sulphite/amidol for the development temperatures from 18°C to 24°C.

It is an accepted fact that a decrease in the development temperature minimizes distortion in nuclear emulsions. Monothermal developments suggested by Yagoda (Y5) (1955) and Marquin (M2) (1957) in which temperature variations are replaced by P.H. Variations, was avoided because of its complexity of operation. To avoid distortion, the development was, therefore, carried out at lower than usual temperatures which in turn gave rise to colouration defects, besides an irregularity in development.
To obviate this difficulty, four types of developers indicated below were tried.

**TABLE 1**

Developers Compositions: (in gms/lit. of the solution)

<table>
<thead>
<tr>
<th></th>
<th>1.2</th>
<th>1.2</th>
<th>1.2</th>
<th>1.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>gms/lit. potassium bromide (K Br)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>gms/lit. sodium sulphite (Na₂SO₃)</td>
<td>11</td>
<td>10</td>
<td>17</td>
<td>20</td>
</tr>
<tr>
<td>gms/lit. amidol</td>
<td>4.0</td>
<td>2.8</td>
<td>2.8</td>
<td>2.8</td>
</tr>
<tr>
<td>gms/lit. boric acid (H₃BO₃)</td>
<td>30</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
</tbody>
</table>

Ratio of sodium sulphite: amidol: 2.75 3.57 6.07 7.14

By a comparative study of these formulae, it was found that to develop reliably at low temperatures, it was necessary to increase the ratio sodium sulphite/amidol to minimize the colouration. In certain cases of low sodium sulphite concentration, it was observed that the emulsion showed zones of good development, whereas the rest of the plate became useless. Further, in such cases the images were found to show a blue or red transparency. The development was carried out at temperatures higher and lower than 18°C, and the colouration was found to be more predominant at lower temperatures, and was found to be a function of development time.
Results, almost resembling those indicate above, were reported by Birge (B4) (1954) and others. In some cases the images were completely destroyed, but one thing is clear that this image destruction has nothing to do with the corrosion which sets in during prolonged fixation, and is more or less a surface phenomenon. In the present case, the disappearance of the image begins deep down in the emulsion.

These phenomena as already reported by James and vanesslow (J1) (1953) are due to the displacement of the adsorption equilibria of organic compounds and sodium sulphites, on silver halide grains. The following equation as suggested by Chateau (C1)(1956) can explain the phenomenon

\[ 2Ag\text{Na}_2\text{SO}_3 \rightarrow 2Ag + Na_2S_2O_6 \]
\[ AgS_2O_6 + Ag + H_2O \rightarrow Ag_2S + H_2SO_4 \]

All the three sets of plates of 100, 200 and 400 micron thickness were tried, and it was found that all the three sets showed the colouration defect; but the thicker ones are less coloured than their thinner counterparts.

In conclusion, it was found necessary to choose a sodium sulphite to amidol ratio which is 75% greater than the ratio adopted in Brussels or Chicago developers. Further the
Accelerating column of the Neutron generator
High tension unit of the neutron generator
following processing conditions were found to give the best results:

<table>
<thead>
<tr>
<th>Emulsion thickness (microns)</th>
<th>Water pre-soak Temp</th>
<th>Developer pre-soak Temp</th>
<th>Development time</th>
<th>Stop bath 1% CH₃COOH Temp</th>
<th>Stop bath Time Hrs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>4 0.4</td>
<td>4 0.4</td>
<td>variable</td>
<td>4 0.4</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>4 1.0</td>
<td>4 1.0</td>
<td>variable</td>
<td>4 1.0</td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>4 2.6</td>
<td>4 2.6</td>
<td>variable</td>
<td>4 2.6</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>(Microns) Fixation (40% hypo) Temp</th>
<th>Washing Temp</th>
<th>Glycerinization one percent Temp</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>100 18 3.5</td>
<td>8 4.0</td>
<td>6 0.5</td>
<td></td>
</tr>
<tr>
<td>200 18 8.0</td>
<td>8 10.0</td>
<td>6 1.0</td>
<td></td>
</tr>
<tr>
<td>400 18 24.0</td>
<td>8 28.0</td>
<td>6 2.0</td>
<td></td>
</tr>
</tbody>
</table>

Fog was removed by rubbing immediately after the stop bath treatment.

(11) Dependence of track and grain structures on the hot stage temperature, hot stage time and sodium sulphite concentration:

In this connection various parameters, viz., diameter
of the track grains, volumetric grain density, total gap length, mean gap length and blob density were studied by the author, in the case of Ilford C2 200 micron plates. Some work has already been reported by Fatzer (F2) (1959) in this connection. Sodium sulphite:amidol ratios and temperatures different from those used by Fatzer have been used. Results, although identical with those reported by Fatzer, differ in the peak values of the curves as reported:

a) Diameter of the track grains:

Mean grain diameters were plotted against the development time, for two temperatures and two sodium sulphite concentrations. The results obtained are plotted in Figs. ( ).

Mean diameter of the track grains was found to increase rapidly with the time, in the region of under-development. It, however, attained a constancy in value for longer development periods.

Further, it was observed that an increase in the sodium sulphite concentration and development temperatures causes a decrease in the grain diameters.

b) Volumetric grain density was found to increase with an increase in the hot stage time, and the sodium sulphite concentration. The observations are plotted in Figs. ( ).
1. **Total gap length**: 

In the region of under-development total gap length was found to decrease with an increase in the development time. At long development periods, the total gap length remains constant, and at normal developments, the total gap length decreases with increasing temperature, and is independent of sodium sulphite concentration. (Fig.)

2. **Mean gap length**: 

The following points were observed in connection with the mean gap length, which was calculated by dividing the total gap length by the number of blobs.

i) M.G.L. is independent of the duration of hot stage time, excepting in the case of underdevelopment.

ii) M.G.L. decreases with an increase in temperature.

iii) It does not vary with a change in sodium sulphite concentration.

5) **Variations of blob density**: 

Blob density was calculated by dividing the number of blobs by the total length of the tracks. It was observed that the blob density increases in the region of underdevelopment. Rise in temperature was found to cause a considerable rise in the blob
Fig. 1. Grain diameters plotted against development time for two different temperatures.

Fig. 2. Volumetric grain density plotted against hot stage time at two NaSO₄ concentrations.
**Fig. 1.** Total gap length plotted against development time in hours.

**Fig. 2.** Mean gap length plotted against the development time at two different temperatures.

**Fig. 3.** Blob density plotted against development time in hrs.
density and after a certain value of development time, the blob density attained constancy in value. These observations are incorporated in Fig. ( ).

3) Removal of microscopic fog from the plates:

Background of developable but unexposed grains of silver bromide, manifesting itself in the form of microscopic fog, is a great handicap in the emulsion work. The idea underlying removal of fog is to develop an unexposed plate to reduce all the developable silver bromide grains to free silver, which is then removed by reduction. Photographic reducers consist of oxidizing agents which oxidize free silver to a suitable salt, which in turn causes a reduction in the opacity of the image.

Well known formulae using potassium permanganate and potassium bichromate were used in this laboratory. Potassium permanganate formula was found to give better results, and also it was observed that potassium bichromate formula does not cause complete reduction, and the grains thus reduced partially are again developable.

Out of the formulae suggested by various workers, the
following formula, modified by the author was found to give the best results, without affecting silver bromide in the emulsion.

Sol. 1. — Potassium permanganate — 3 gms; water to make 1000 c.c.

Sol. 2. — Sulphuric acid — 8 c.c.; water to make 1000 c.c.

Procedure:

The plate was first developed in the Eastman developer D - 11 for a time greater than the usual, so as to be sure about the development of undesirable grains. The plate was then washed and immersed for about 40 minutes in a freshly prepared mixture of equal volumes of solutions 1 and 2. This procedure was tried for plates of various thickness. Following timings were found necessary:

<table>
<thead>
<tr>
<th>Plate thickness (micron)</th>
<th>100</th>
<th>200</th>
<th>400</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time in minutes</td>
<td>30</td>
<td>40</td>
<td>55</td>
</tr>
</tbody>
</table>

Thus longer time was needed for the clearance of thick plates, as compared to their thinner counterparts. In order to remove the brown stain produced by potassium permanganate, the plate was again washed and placed for 15 minutes in a 10% solution of sodium bisulphite. This procedure was found best for the removal of the background fog.
4. Background eradication in nuclear emulsion:

The background is partly due to the accumulated latent images of $\alpha$-tracks and stars produced by traces of Ra, and Th, normally present as impurities in the emulsion and partly due to the cosmic ray background.

It was found that storage for several hours in humid atmosphere goes a long way in accelerating the fading of latent images due to the background. The emulsions thus treated were then dessicated for 1 to 2 hrs. over calcium chloride.

Further, the effect of both the temperature and humidity on fading was investigated so as to find out their effect on the eradication of the background tracks.

Effect of temperature on fading:

The variation of the fading rate with the storage temperature may be determined from a consideration of the effect of temperature on the velocity of a gas solid chemical reaction, such as is assumed to occur during fading between certain constituents of the atmosphere and the silver development centres of the exposed emulsion. This gives a form of the Arrhenius relation with $-\frac{dN}{dt}$, the rate of disappearance of the development centres as the equation

$$-\frac{dN}{dt} = Ce^{-K/T}$$

where $C$ and $K$ are constants and $T$ the absolute storage temperature.
The equation shows that the fading produced under otherwise fixed condition will be an exponential function of the reciprocal of the absolute temperature, a result confirmed by Farragi (1949). This shows the effect of storage temperature on the background eradication, and it was found that the experimental findings were in accord with the theory.

**Effect of humidity on fading:**

According to Albouy and Farragi (A1) (1949), fading is a result of the oxidation of the development specks by the atmospheric oxygen in the presence of water, the reaction as proposed by Albouy and Farragi proceeds as

\[ Ag^+ + O + H_2O \rightarrow AgOH \]

It is evident that the presence of an excess of OH ions i.e. PH value above 7, will act to inhibit the reaction, while the more acid concentrations accelerate it. Thus an increase in the humidity causes a decrease in the fading rate.

The following figures showing the effect of both temperature and humidity on the background eradication is in accord with the theory.

Theoretical investigations due to Beiser (B5) (1951) also lead to the following equation for the fading coefficient
\[
\frac{D_0 - D}{D_0} = 1 - \exp(-ct)
\]

where \(D_0\) = grain density produced upon immediate development
\(D\) = grain density after a time \(t\).
\(C\) = constant depending upon the size of Ag specks.

The fading coefficient plotted as a function of \(t\), for various values of \(C\), is in agreement with the experimental curves.

These results have been reported in detail by the author (Kg)(1961).
Effect of ammonium salts over the clearing time, grain size and shrinkage:

Thick plates of 200 µm and above used here for (v, <£) study sometimes do not get cleared up in the desired time. This prolonged fixation may be due to many causes. It was found that if the plate of 200 µm is kept in the fixer for more than 5 hrs., severe distortion take place in the tracks, and therefore the plate becomes useless for scanning. This, therefore, necessitates the use of some device by which fixation could be speeded up to avoid all these undesirable effects. Although some authors have worked on this problem, but it was found necessary to take up the work in this laboratory in view of some contradictory results reported by these authors.

The addition of such ammonium salts can be allowed only if such salts do not effect the grain size or the shrinkage factor of the emulsions, and therefore a study was undertaken in this direction also, to determine the feasibility of the use of ammonium salts for speeding up the fixation for (v, <£) study, and to choose a suitable ammonium salt. It has been found that the addition of ammonium salts to hypo decrease the fixation time considerably; because the ammonium salts of hypo are better
complex forming salts with silver halides than the sodium salts of hypo.

\[ 2\text{NH}_4\text{Cl} + \text{Na}_2\text{S}_2\text{O}_3 \rightarrow (\text{NH}_4)_2\text{S}_2\text{O}_3 + 2\text{NaCl} \]

Ammonium Salt of Hypo

Piper (P$_9$) worked on this problem, and investigated the change in the clearing time of thin plates with a change in the concentration of ammonium salts. He reported a fall in the clearing time with an increase in the concentration of ammonium salts, till an optimum concentration is attained, after which, the clearing time has been reported to increase again. Also according to him the Cl$^-$ ion in ammonium chloride is responsible for eating up the developed grains near the surface of the emulsion.

Y. Prakash and others (P$_9$) (1958) reported a contradiction to the observations made by Piper. According to them, the clearing time decreases up to an optimum concentration of the ammonium salt, after which it remains more or less constant, and does not have an appreciable rise as reported by Piper.

In view of the above contradictions the author found it useful to take up some work in this direction. The observations and the results arrived at are reported as under.
Experimental procedure and observations :-

The ammonium salts selected by the author were ammonium chloride, ammonium acetate, and ammonium sulphate. The first salt was selected, because in addition to ammonium thiosulphate, the chlorine ion concentration affects the diffusion rate of the fixer, and therefore may suppress the reaction; and also because it offers a direct verification of the observations made by Piper and those due to Y. Prakash and others; ammonium sulphate, and acetate were selected so as to study the effect due of any other two suitable ammonium salts, not tried so far.

For this work Ilford K2 plates of 100, 200 and 400 microns, and also K0 and G5 plates of 200 micron thickness were used so as to study the effect on the clearing time by a change in the thickness and the type of the emulsion. Further, the experiments were conducted in the following stages :-

1) Effect of the change in the concentration of ammonium chloride on the clearing time of Ilford 100 micron plates—K0 and G5 was studied, using different concentrations of hypo. Plates were directly put in the fixing solution maintained at 17°C. Care was taken to see that the level of the fixing solution is
the same in all the three cases, and the results are plotted as shown in Fig. ( )

The curves show a fall upto an optimum value of concentration, and then a rise, although the rise is not as pronounced as reported by Piper. Moreover a shift in the position of optimum concentration was observed with a change in the hypo concentration, as shown in Fig. ( ) Change in the type of emulsion however has been found to have no effect on the clearing time of the emulsion.

2) Secondly, the change in the clearing time of $K_2$ and $K_0$ 200 micron plates with a change in the concentration of the three ammonium salts was studied. A fall, and then a rise has been observed in the case of all the three curves; for ammonium chloride, ammonium sulphate and ammonium acetate. However, the rise is more pronounced in the case of ammonium chloride, than in the case of other two ammonium salts. Results, which have further been found to be independent of the type of emulsion used, are represented in Fig. ( )

3) Thirdly, the effect of the emulsion thickness over the percentage reduction in the clearing time was investigated in the case of $K_2$ plates for all the three ammonium salts. The curves were found to have a fall upto 200 microns in all the cases, excepting for ammonium chloride where the fall persists beyond 200 microns. These results are plotted as shown in Fig. ( )
CLEARING TIME IN MINUTES

CONCENTRATION OF THE AMMONIUM SALT

CONCENTRATION OF NH\textsubscript{4}CL

THICKNESS OF THE COMPOSITION PM.
Discussion of the results:

The results may be discussed in the light of the three stages of observations mentioned above.

1. The rise and fall of curves shown in Fig. ( ) can be accounted for as follows:

Ammonium salts of hypo being better complex forming salts with silver halides, as compared to sodium salts of hypo, any increase in the concentration of the ammonium salts should decrease the clearing time.

The optimum concentration can be explained on the basis of the reversible nature of the reaction, and so also the shift in the position of the optimum with respect to different hypo concentrations. The rise in the curves may be explained as being due to the fact that higher complexes of ammonium thiosulphate and silver halides are unstable as compared to the higher complexes of sodium thiosulphate, and silver halides. The clearing time, however, is independent of the type of the emulsion as is expected.

2) A fall, and then a rise in the curves is obtained in case of all the three ammonium salts, as is explained on the
basis of the explanations given above. The more pronounced rise observed in the case of ammonium chloride can be explained as being due to the presence of Cl ion which affects the rate of diffusion. Chlorine hardens the gelatine of the emulsion, thereby increasing the clearing time by effecting the diffusion rate. As this factor is absent in the case of the other two ammonium salts, rise is not as pronounced in their case.

3) The percentage fall goes on decreasing in the case of higher thicknesses for NH₄Cl, whereas for other salts it remains constant after 200 microns. The reason for this is that upto 200 microns the diffusion velocity of the fixing solution decreases with an increase in the thickness, and hence a fall in the curve. In case of ammonium chloride this effect persists beyond 200 microns, because of the contribution of the Cl ion, which however, is not the case with other ammonium salts. Also, the diffusion velocity becomes independent of the emulsion thickness beyond 200 microns. Effect on the shrinkage factor and the grain size:

Effect on the grain size was investigated so as to find out whether or not the chlorine eats away the developed grains at the surface as reported by Piper. For this, the grain size at the top and the bottom of the plates was found the case...
of pure hypo, and hypo with each of the other ammonium salts. (The optimum concentration being used in each case.) The plates of the same type were simultaneously put in the four different baths and the fixing carried out under the same experimental conditions. The observations regarding the shrinkage factor and the grain size are tabulated as under:

### Observations regarding the shrinkage factor

<table>
<thead>
<tr>
<th>Plate thickness in microns</th>
<th>Shrinkage factor</th>
<th>Pure hypo</th>
<th>Hypo with optimum concentration of ammonium salts</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Ammonium chloride</td>
</tr>
<tr>
<td>100</td>
<td>2.00</td>
<td>2.16</td>
<td>2.05</td>
</tr>
<tr>
<td>200</td>
<td>2.50</td>
<td>2.60</td>
<td>2.4</td>
</tr>
<tr>
<td>400</td>
<td>2.75</td>
<td>2.50</td>
<td>2.50</td>
</tr>
</tbody>
</table>

### Observations regarding the grain size

<table>
<thead>
<tr>
<th>Thickness of plate in microns</th>
<th>Surface Emulsion of Free hypo Hypo having optimum concentration of ammonium salts</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>Bottom</td>
</tr>
<tr>
<td></td>
<td>top</td>
</tr>
</tbody>
</table>
The study of shrinkage factor and the grain size reveals that within the allowed statistics, the shrinkage and the grain size is in no way affected by the addition of the ammonium salts. It also contradicts the hypothesis that chlorine is responsible for eating away the emulsion at the surface. The optimum concentration of the various ammonium salts used by the author was found to be 1.5% for ammonium chloride, 2.5% for ammonium sulphate and 2.5% for ammonium acetate. Relative study of the suitability of these salts as regards the clearing of thick plates:

For thick plates the clearing time depends predominantly upon the diffusion velocity of the fixing solution, as against the thin plates. Consequently the pH value of the fixing solution becomes an important factor. To make a relative study of the usefulness of these salts as regards the clearing of thick plates, pH values of the various concentrations of these solutions were calculated by the usual formulae, Vogel (V₁) (1958). The results
are tabulated as follows:

<table>
<thead>
<tr>
<th>Conc. of the Amm. Salt %</th>
<th>0.5</th>
<th>1.0</th>
<th>1.5</th>
<th>2.0</th>
<th>2.5</th>
<th>3.0</th>
<th>3.5</th>
<th>4.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>PH of (NH₄)₂SO₄</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>+ Hypo</td>
<td>5.3</td>
<td>5.6</td>
<td>5.8</td>
<td>6.0</td>
<td>5.7</td>
<td>5.6</td>
<td>5.3</td>
<td>5.0</td>
</tr>
<tr>
<td>PH of NH₄C₂O₄H₃</td>
<td>6.3</td>
<td>6.8</td>
<td>6.9</td>
<td>7.0</td>
<td>7.1</td>
<td>7.3</td>
<td>7.9</td>
<td>8.1</td>
</tr>
<tr>
<td>+ Hypo</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The table clearly shows that the pH value of Hypo + Amm. sulphate and also Hypo + Amm. chloride solution is well within the acid region, thus ensuring a high diffusion velocity, and the pH values of amm. acetate being in the basic region, it does not have a high diffusion velocity. Therefore, amm. acetate + hypo is not as suitable for clearing as the other two salts. Ammonium sulphate, however, has been found to be most suitable for this purpose.

These results have already been reported by the author (Kg) (1961).
Processing tank used during the course of this work.

a) $4^2$ bath used during presoaking and cold development.
b) $10^2$ baths used during fixing.
c) chamber used during warm development stage.
d) cold storage chambers.
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ON THE PROCESSING OF NUCLEAR EMULSIONS

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(Received March 28, 1961)

ABSTRACT. The paper gives an account of the comparative study of the various process formulae used by the author during the course of the emulsion work, using both thick and thin plates. Modified formulae for various processing stages found to give the best results have also been suggested.

The paper also gives the penetration time needed for various developers both in the case of presoaked and non-presoaked emulsions of various thicknesses, and also an account of the shrinkage factor in nuclear emulsions.

The author has made a detailed study of the processing technique using 9C nuclear emulsions of 100, 200 and 400 micron thickness, and also K, G, plates. The details of the investigation are given below under the various stages.

e-soaking stage

Before starting with the development, it is necessary to soak the emulsion in water, so that the penetration of the developer may become easier and more rapid. For this, the temperature and time limits suggested by various workers are used, and the following time and temperature limits were found to give best results:

<table>
<thead>
<tr>
<th>Emulsion thickness</th>
<th>Time</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.5 hrs.</td>
<td>2°C</td>
</tr>
<tr>
<td>200</td>
<td>0.75 hrs.</td>
<td>4°C</td>
</tr>
<tr>
<td>400</td>
<td>1.25 hrs.</td>
<td>6°C</td>
</tr>
</tbody>
</table>

To facilitate penetration, presoaking in distilled water with or without the addition of the wetting agent is frequently made use of. This acts to swell the emulsion, permitting more rapid diffusion of the developer. It, however, does not take the role of the alkali developments (Dilworth et al, 1947; Mortier and Vermaesen, 1948; Picciotto, 1949). The suitable temperature at which the penetration is to occur has been found to be 4°C. Below this temper-
perature the penetration time was found to be too long, and above it, the rate of developer penetration increases less rapidly than its activity.

Development

Thin emulsions of 100 μm order:

Two degrees of development were found possible for thin emulsions. Moderate development was found useful when grain densities of comparatively dense tracks (e.g., protons and α tracks of several MeV energy) are to be measured. Since in this case it is essential that the grains be discrete, moderate development is preferred. This has an additional advantage of great reduction in the fog density. Strong development, on the other hand, although accompanied by an increased fog background, permits a full utilization of the emulsion sensitivity, and the heavy ionizing particles appear as solid columns of silver grains. Series of development tests were conducted to determine the development time giving the most preferred combination of background and track densities. The results thus obtained are indicated below:

<table>
<thead>
<tr>
<th>Thickness</th>
<th>Procedure</th>
<th>Time</th>
<th>Agitation</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>(moderate development)</td>
<td>10 min,</td>
<td>no agitation</td>
</tr>
<tr>
<td>100</td>
<td>(strong development)</td>
<td>40 min,</td>
<td>no agitation</td>
</tr>
</tbody>
</table>

Thick emulsions of 400 micron order and above

For thick emulsions, the two developer solution method as suggested by Blau and Defilice (1948) is found to give the best results to secure the even development. The first contains the developing agent without any alkali, permitting the diffusion of the developer into the emulsion without any appreciable amount of actual development occurring. The second bath containing an excess of alkali permits the development to take place.

This method requires that the velocity of the travel of a pH change should exceed that of the developer itself—a condition which is not actually satisfied. Also, up to 400 μm thickness the two bath method eliminates any danger of reticulation.

It has been found by the author that the following modified formulae give the best results in the case of thick emulsions so far as two-bath development is concerned:

Sol. A.

Elon 1 gm
Sod. sulphite 20 gms
Hydroquinone 3.5 gms
Pot. bromide 2.0 gms
Distilled water 2 litres
Processing of Nuclear Emulsions

Sol. B.

Stock Eastman D_{19} ... 400 c.c.
Distilled water ... 1600 c.c.
Sod. carbonate ... 12 gms

For 200 micron plates the following single solution development formula found to give the best results. It is the Brussels formula slightly modified by the author:

- Sod. sulphite ... 38 gms.
- Pot bromide ... 0.8 gms.
- Amidol ... 2.8 gms.
- Boric acid ... 12 gms.
- Water ... 1 lit.


treating time needed for various developers

The penetration time in minutes of presoaked and non-presoaked emulsions was investigated by the author at 18°C.

<table>
<thead>
<tr>
<th>Developer</th>
<th>Penetration time in min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Azol 100/μm</td>
<td>200/μm</td>
</tr>
<tr>
<td>Presoaked</td>
<td>4.5</td>
</tr>
<tr>
<td>Non-presoaked</td>
<td>6</td>
</tr>
<tr>
<td>D-19</td>
<td></td>
</tr>
<tr>
<td>Presoaked</td>
<td>3.0</td>
</tr>
<tr>
<td>Non-presoaked</td>
<td>4.5</td>
</tr>
<tr>
<td>Amidol</td>
<td></td>
</tr>
<tr>
<td>Presoaked</td>
<td>1.5</td>
</tr>
<tr>
<td>Non-presoaked</td>
<td>3.0</td>
</tr>
<tr>
<td>Amidol bisulphite</td>
<td></td>
</tr>
<tr>
<td>Presoaked</td>
<td>2.5</td>
</tr>
<tr>
<td>Non-presoaked</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Processing formulae of thick and thin emulsions: The following processing formulae were found to be most suitable for 100, 200 and 400 μm plates.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Temperature</th>
<th>Time in min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Presoaking</td>
<td>4°C</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>5°C</td>
<td>25</td>
</tr>
<tr>
<td>Penetration of the cold developer</td>
<td>4°C</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>5°C</td>
<td>26</td>
</tr>
<tr>
<td>Warm dry development</td>
<td>18°C</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>24°C</td>
<td>20</td>
</tr>
<tr>
<td>Dry cooling</td>
<td>18°C to 5°C</td>
<td>5</td>
</tr>
<tr>
<td>Stop bath acetic acid (0.5%) 5°C</td>
<td>30</td>
<td>45</td>
</tr>
<tr>
<td></td>
<td>1.0% 5°C</td>
<td>20</td>
</tr>
<tr>
<td>Fixation</td>
<td>18°C</td>
<td>3 hrs.</td>
</tr>
<tr>
<td>(clearing time +5% more)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Washing</td>
<td>8°C</td>
<td>3 hrs.</td>
</tr>
</tbody>
</table>
The following processing formula was found exclusively suitable for thick plates:

Distilled water 1000 c.c.
Sod. sulphite (anhydrous) 12 gms.
Pot. bromide (10% solution) 8 cc
Amidol 3.8 gms.
pH of the developer 7.4

Fixing bath (pH 5.3)
Distilled water 1000 c.c.
Sod. thiosulphate 400 gms.
Sod. bisulphite 10 gms.
NH₄Cl 7 gms.

Clearing solution (pH 4.2)
Distilled water 500 cc
Ammonium acetate 15 gms.
Citric acid 8 gms.
Thio-urea 8 gms.

Small quantities of sodium bisulphite and ammonium chloride reduce staining and hasten the fixation of the emulsion. But large concentrations of these ingredients lead to distortion. Further, in the fixing solution, one half of the quantity of hypo was replaced at several intervals, thus avoiding salt concentration shock. For the same reason, washing was also preceded by a gradual dilution of the fixing solution.

**Shrinkage**

The considerable reduction in the thickness of nuclear emulsions after fixing is due to the high concentration of the silver bromide in nuclear emulsions. The ratio of the emulsion thickness before and after fixing were found, and the following results were observed:

<table>
<thead>
<tr>
<th>Emulsion thickness in μm.</th>
<th>Before processing</th>
<th>After processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>96</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>181</td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>359</td>
<td></td>
</tr>
</tbody>
</table>
The increase in the shrinkage factor of the emulsions was investigated at relative humidity.

Fig. 1. Shrinkage factor plotted against the time of storage in hours for Ilford C2 nuclear emulsions.

Plates of either thicknesses were soaked in glycerine solution and dried gently. Any drying was avoided, because it produces a skin at the surface which traps water lower down. This in turn produces stresses at the soft emulsion which cause severe distortions in the tracks. Blowing over the surface was avoided, as it introduces severe distortions, although the temperature was slightly raised to accelerate the process (Dilworth, 1961). Edges of the plates usually first cause surface deformations in the emulsion, which was also minimized as much as possible.

ACKNOWLEDGMENTS

The author is grateful to Prof. B. D. Nagchandhuri, Director of the Institute, for encouragement and guidance.

Thanks are also due to Prof. D. N. Kundu, Head of the Accelerator Division, providing the laboratory facilities and to Mr. Bhupesh Purkayastha, Head of Nuclear Chemistry Division, for many helpful discussions.
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STUDY OF DEVELOPMENT DEFECTS AND TRACK STRUCTURES IN NUCLEAR EMULSIONS USING AMIDOL DEVELOPERS

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(Received, December 30, 1960)

ABSTRACT. Development defects in nuclear emulsions, and also the dependence of track and grain structures on the various development parameters have been investigated. Modified formulae have been suggested for the removal of microscopic fog, coloration, and also for the background eradication.

The procedures of investigation followed during the course of this work, differ from those adopted by others like Fatzer, Yagoda, Barschall, and Liebermann; and the results, though identical in most of the cases, differ slightly in certain respects as reported.

Some work has been reported on the elimination of development defects by us workers, e.g., Yagoda, (1948), Liebermann and Barschall (1943) and others. The present aim of the author has been to make a comprehensive study of development defects introduced by Amidol developers to compare the modes suggested by various workers for their elimination and also to suggest modified formulae suitable for work in this laboratory.

Besides this, a critical study of the track and grain structures pertaining to their dependence on various development parameters (temperature, time, sodium sulphite concentration) has been made by methods somewhat different from those adopted by Fatzer (1959).

Ilford C4 nuclear emulsions of 100, 200 and 400 micron thickness were used in this study under the following heads:

1) Elimination of the development defects introduced by Amidol developers.

2) Dependence of track and grain structures on the hot stage temperature, time, and sodium sulphite concentration.

3) Removal of microscopic fog from the plates.

4) Background eradication in nuclear emulsion by the accelerated fading of latent images.
Elimination of the development defects introduced by the use of Amidol developers:

Decrease in the development temperature gives rise to development defects, which are evidenced by a total or partial destruction of the developed image and also a coloration, extending to a considerable depth in the emulsion.

By a series of trials carried out by the author, it was found necessary to increase the ratio sodium sulphite/Amidol for the development temperatures from 18°C to 24°C.

It is an accepted fact that a decrease in the development temperature minimizes distortion in nuclear emulsions. Monothermal development suggested by Yagoda (1955) and Marguin (1957) in which temperature variations are replaced by P. H. variations, was avoided because of its complexity of operation. To avoid distortion, the development was, therefore, carried out at lower than usual temperatures which in turn gave rise to coloration defect, besides an irregularity in development.

To obviate this difficulty, four types of developers as indicated below were tried by the author.

**TABLE I**

Developer compositions: (in gms/lit. of the solution)

<table>
<thead>
<tr>
<th>Developer compositions</th>
<th>1.2</th>
<th>1.2</th>
<th>1.2</th>
<th>1.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>gms/lit. potassium bromide (KBr)</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>gms/lit. sodium sulphite (Na₂SO₃)</td>
<td>11</td>
<td>10</td>
<td>17</td>
<td>20</td>
</tr>
<tr>
<td>gms/lit. Amidol</td>
<td>4.0</td>
<td>2.8</td>
<td>2.8</td>
<td>2.8</td>
</tr>
<tr>
<td>gms/lit. boric acid (H₃BO₃)</td>
<td>30</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Ratio of sodium sulphite : Amidol</td>
<td>2.75</td>
<td>3.57</td>
<td>6.07</td>
<td>7.14</td>
</tr>
</tbody>
</table>

By a comparative study of these formulae it was found that to develop reliably at low temperatures, it was necessary to increase the ratio sodium sulphite/Amidol to minimize the coloration. In certain cases of low sodium sulphite concentration, it was observed that the emulsion showed zones of good development, whereas the rest of the plate became useless. Further, in such cases the images were found to show a blue or red transparency. The development was carried out at temperatures higher and lower than 18°C and the coloration was found to be more predominant at lower temperatures, as well as to be a function of development time.

Results, almost resembling those indicated above, were reported by Birge (1954) and others. In some cases the images were completely destroyed,
one thing is clear that this image destruction has nothing to do with the erosion which sets in during prolonged fixation, and is more or less a surface phenomenon. In the present case, the disappearance of the image begins deep within the emulsion.

These phenomena as already reported by James and Vonselov (1953) are due to the displacement of the adsorption equilibria of organic compounds and thiourea, on silver halide grains. The following equation as suggested by Steau (1956) can explain the phenomenon:

\[
\begin{align*}
2\text{Ag} \cdot \text{NaSO}_3 & \rightarrow 2\text{Ag} + \text{Na}_2\text{S}_2\text{O}_3 \\
\text{AgS}_2\text{O}_3 + \text{Ag}^+ + \text{H}_2\text{O} & \rightarrow \text{Ag}_2\text{S} + \text{H}_2\text{SO}_4
\end{align*}
\]

All the three sets of plates of 100, 200 and 400 micron thickness were tried, and it was found that all the three sets showed the coloration defect; but the thicker ones are less coloured than their thinner counterparts.

In conclusion, it was found necessary to choose a sodium sulphite to Amidol which is 75% greater than the ratio adopted in Brussels or Chicago developers. Further, the following processing conditions used by the author were found to give best results:

<table>
<thead>
<tr>
<th>TABLE II</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
<tr>
<td>Plate thickness (microns)</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Pre-soak developer</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Stop bath developer</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Development time</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Fixation (40% hypo)</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Washing</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
<tr>
<td>Glycerization</td>
</tr>
<tr>
<td>Temp.</td>
</tr>
<tr>
<td>Time</td>
</tr>
</tbody>
</table>

Fog was removed by rubbing immediately after the stop bath treatment.

2) Dependence of track and grain structures on the hot stage temperature, hot plate time and sodium sulphite concentration:

In this connection various parameters, viz., diameter of the track grains, areometric grain density, total gap length, mean gap length and blob density.
were studied by the author, in the case of Ilford Cz 200 micron plates, some work has already been reported by Fatzer (1959) in this connection. Sodium sulphite/Amidol ratios and temperatures different from those used by Fatzer have been used. Results, although identical with those reported by Fatzer, differ in the peak values of the curves as reported:

a) Diameter of the track grains:

Mean grain diameters were plotted against the development time, for two temperatures and two sodium sulphite concentrations. The results obtained are plotted in Figs. 1 and 2.

Mean diameter of the track grains was found to increase rapidly with the time, in the region of under-development. It, however, attained a constancy in value for longer development periods (Figs. 1 and 2).

Further, it was observed that an increase in the sodium sulphite concentration and development temperatures causes a decrease in the grain diameters.

b) Volumetric grain density:

Volumetric grain density was found to increase with an increase in the hot stage time and the sodium sulphite concentration. The observations are plotted in Fig. 3.
1. Grain diameters plotted against the development time at two different temperatures.

\[ \text{Scd. sulphite Amidol} = 15 \]

2. Volumetric grain density plotted against hot stage time at two \( \text{Na}_2\text{SO}_3 \) concentrations.
1) **Total gap length:**

In the region of under-development total gap length was found to decrease with an increase in the development time. At longer development periods, the total gap length becomes constant. At normal developments, however, the total gap length decreases with increasing temperature and is independent of sodium sulphite concentration. (Fig. 4).

![Graph showing total gap length plotted against development time in hours.](image)

**Fig. 4.** Total gap length plotted against development time in hours.

2) **Mean gap length:**

The following points were observed in connection with the mean gap length, which was calculated by dividing the total gap length by the number of blobs.

1) M.G.L. is independent of the duration of hot stage time, excepting in the case of under-development.

2) M.G.L. decrease with an increase in temperature.

3) It does not vary with a change in sodium sulphite concentration.

5) **Variation of blob density.**

Blob density was calculated by dividing the number of blobs by the total length of the tracks. It was observed that the blob density increases in the region of under-development. Rise in temperature was found to cause a considerable rise in the blob density and after a certain value of development time, the blob density attained constancy in value. These observations are incorporated in Fig. 6.
Study of Development Defects and Track Structures, etc. 189

Mean gap length plotted against the development time at two different temperatures.

Fig. 6. Blob density plotted against development time in hrs.

(3) Removal of microscopic fog from the plates:
Background of developable but unexposed grains of silver bromide, manifesting itself in the form of microscopic fog, is a great handicap in the emulsion. The idea underlying removal of fog is to develop an unexposed plate to reveal all the developable silver bromide grains, to free silver, which is then reduced by reduction. Photographic reducers consist of oxidising agents which...
oxidise free silver to a suitable salt, which in turn causes a reduction in the opacity of the image.

Well known formulae using potassium permanganate and potassium bichromate were used in this laboratory. Potassium permanganate formula was found to give better results, and also it was observed that potassium bichromate formula does not cause complete reduction and the grains thus reduced partially are again developable.

Out of the formulae suggested by various workers, the following formula modified by the author was found to give best results, without affecting silver bromide in the emulsion.

**Sol. 1**—Potassium permanganate—3 gms water to make 1000 cc

**Sol. 2**—Sulphuric acid—8 cc water to make 1000 cc.

**Procedure**:

The plate was first developed in the Eastman developer D-11 for a time greater than the usual, so as to be sure about the development of undesirable grains. The plate was then washed and immersed for about 40 minutes in a freshly prepared mixture of equal volumes of solutions 1 and 2. This procedure was tried for plates of various thicknesses. Following timings were found necessary:

<table>
<thead>
<tr>
<th>Plate thickness (microns)</th>
<th>100</th>
<th>200</th>
<th>400</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time in minutes</td>
<td>30</td>
<td>40</td>
<td>55</td>
</tr>
</tbody>
</table>

Thus longer time was needed for the clearance of thick plates, as compared to their thinner counterparts. In order to remove the brown stain produced by potassium permanganate, the plate was again washed and placed for 15 minutes in a 10% solution of sodium bisulphite. This procedure was found best for the removal of the background fog.

4) **Background eradication in nuclear emulsion**

The background is partly due to the accumulated latent images of tracks and stars produced by traces of Ra and Th, normally present as impurities in the emulsion and partly due to the cosmic ray background.

It was found that storage for several hours in humid atmosphere goes a long way in accelerating the fading of latent images due to the background. The emulsions thus treated were then dessicated for 1 to 2 hrs. over calcium chloride.

Further, the effect of both the temperature and humidity on fading was investigated so as to find out their effect on the eradication of the background tracks.
The variation of the fading rate with the storage temperature may be determined from a consideration of the effect of temperature on the velocity of a gas chemical reaction, such as is assumed to occur during fading between certain elements of the atmosphere and the silver development centres of the exposed ion. This gives a form of the Arrhenius relation with \(- \frac{dN}{dt}\), the rate of appearance of the development centres, as the equation

\[
- \frac{dN}{dt} = C e^{-K/T}
\]

where \(C\) and \(K\) are constants and \(T\) the absolute storage temperature. The ion shows that the fading produced under otherwise fixed conditions will be an exponential function of the reciprocal of the absolute temperature, a result found by Farragi (1949). This shows the effect of storage temperature on background eradication and it is found that the experimental findings accord with the theory.

### Effect of humidity on fading:

According to Albouy and Farragi (1949) fading is a result of the oxidation of development specks by the atmospheric oxygen in the presence of water, reaction as proposed by Albouy and Farragi proceeds as

\[
Ag + O + H_2O \rightarrow Ag + 2OH^-
\]

It is evident that the presence of an excess of OH\(^-\) ions i.e. pH value above 1 act to inhibit the reaction, while the more acid conditions accelerate it. An increase in the humidity causes a decrease in the fading rate.

The following graph showing the effect of both temperature and humidity on background eradication is in accord with the theory.

Theoretical investigations due to Beiser (1951) also lead to the following ion for the fading coefficient:

\[
\frac{D_0 - D}{D_0} = 1 - e^{Ct}
\]

where

- \(D_0\) = grain density produced upon immediate development
- \(D\) = grain density after a time \(t\)
- \(C\) = constant depending upon the size of Ag specks

The fading coefficient plotted as a function of \(t\), for various values of \(C\), is in agreement with the experimental curves.
Fig. 7. The variation of the fading coefficient with the time of storage at various temperatures.

ACKNOWLEDGMENTS

The author expresses his deep sense of gratitude to Prof. B. D. Nag Chaudhuri, under whose direction and guidance this work has been carried out. The author is also thankful to Mr. S. K. Mukherjee for providing the laboratory facilities; and to Messrs. Naresh Sen and Barin Chatterjee for their valuable assistance.

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ROLE PLAYED BY AMMONIUM SALTS IN THE CLEARING OF NUCLEAR EMULSIONS

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ABSTRACT. The effect of the addition of various ammonium salts (ammonium chloride, ammonium sulphate, and ammonium acetate) to the usual hypo solution on the clearing time of nuclear emulsions and also their influence on the grain size and the shrinkage factor, have been investigated in view of some contradictory results reported by various authors in recent years. The addition of the ammonium salts has been found to cause a decrease in the clearing time, and then a slight increase after attainment of the optimum concentration. The addition of ammonium salts, however, has been found to have no effect on the grain size or the shrinkage factor of nuclear emulsions. Results obtained have also been explained on the basis of the current theory.

Thick emulsions necessitate longer clearing times, and this prolonged fixation leads to severe distortions in nuclear emulsions. As such, the methods for decreasing the clearing time have been studied by various authors, and it has been found that the addition of ammonium salts to hypo decreases the fixation time considerably, because the ammonium salts of hypo are better complex forming salts with silver halides than the sodium salts of hypo.

\[
2\text{NH}_4\text{Cl} + \text{Na}_2\text{S}_2\text{O}_3 \rightarrow \frac{(\text{NH}_4)_2\text{S}_2\text{O}_3}{\text{2NaCl}} \text{ Ammonium salt of hypo}
\]

Piper worked on this problem, and investigated the change in the clearing time of thin plates with a change in the concentration of ammonium salts. He reported a fall in the clearing time with an increase in the concentration of the ammonium salts, till an optimum concentration is attained, after which, the clearing time has been reported to increase again. Also, according to him the \(<\text{Cl}>\) ion in ammonium chloride is responsible for eating up the developed grains, near the surface of the emulsion.

Prakash et al. (1958) reported a contradiction to the observations made by Piper. According to them, the clearing time decreases up to an optimum concentration of the ammonium salt, after which it remains more or less constant, and does not have an appreciable rise as reported by Piper.

In view of the above contradictions the author found it useful to take up some work in this direction. The observations and the results thus arrived at are reported in this paper.
EXPERIMENTAL PROCEDURE

The ammonium salts selected were ammonium chloride, ammonium acetate, and ammonium sulphate. The first salt was selected, because in addition to ammonium thiosulphate the chlorine ion concentration affects the diffusion rate of the fixer, and therefore, may suppress the reaction and also because it offers a root verification of the observations made by Piper and those due to Prakash et al. Ammonium sulphate and acetate were selected so as to study the effect due of any other two suitable ammonium salts, not tried so far.

For this work Ilford K2 plates of 100, 200, and 400 microns, and also K4 and K5 plates of 200 micron thickness were used, so as to study the effect on the clearing rate by a change in the thickness and type of the emulsion. Further, the experiments were conducted in the following stages:

1) Effect of the change in the concentration of ammonium chloride on the clearing time of K2 Ilford 100 micron plates K4 and K5 was studied, using different concentrations of hypo. Plates were directly put in the fixing solution maintained at 17°C. Care was taken to see that the level of the fixing solution is same in the three cases. The results are plotted as shown in Fig. 1.

![Fig. 1. The percentage concentration of NH₄Cl against the clearing time in minutes for different concentrations of hypo for 100 plates K2 & K5 (temperature 17°C).](image)

The curves show a fall up to an optimum value of concentration, and then a rise, although the rise is not as pronounced as reported by Piper. Moreover, a shift in the position of optimum concentration was observed with a change in the hypo concentration, as shown in Fig. 1. Change in the type of emulsion, however, has been found to have no effect on the clearing time of the emulsion.

2) Secondly, the change in the clearing time of K2 and K4 200 micron plates with a change in the concentration of the three ammonium salts was studied. A
fall, and then a rise has been observed in the case of all the three curves, for ammonium chloride, ammonium sulphate and ammonium acetate. The rise is more pronounced in the case of ammonium chloride than in the case of other two ammonium salts. Results, which have further been found to be independent of the type of emulsion used, are represented in Fig. 2.

![Graph showing percentage concentration of various ammonium salts cleared time in minutes for 200 Ilford K2 & K0 plates. Temperature during clearing is 17°C. Hypo concentration in each solution is 40%.

3) Thirdly, the effect of the emulsion thickness over the percentage reduction in the clearing time was investigated in the case of K2 plates for all the three ammonium salts. The curves were found to have a fall up to 200 microns in all the cases excepting for ammonium chloride where the fall persists beyond 200 microns. These results are plotted as shown in Fig. 3.

RESULTS AND DISCUSSION

The results may be discussed in light of the three stages of observations mentioned above.

1. The rise and fall of curves shown in Fig. 1 can be accounted for as follows:

Ammonium salts of hypo being better complex forming salts with silver halides, as compared to sodium salts of hypo, any increase in the concentration of the ammonium salts should decrease the clearing time.

$$2\text{NH}_4\text{Cl} + \text{Na}_2\text{S}_2\text{O}_3 \rightarrow (\text{NH}_4)_2\text{S}_2\text{O}_3 + 2\text{NaCl}$$

Complex formation $3(\text{NH}_4)_2\text{S}_2\text{O}_3 + \text{AgBr} \rightarrow \text{Ag}[(\text{NH}_4)_2\text{S}_2\text{O}_3\text{Br}] + \text{NH}_4\text{Br}$

Ag complex
Role Played by Ammonium Salts in the Clearing, etc. 565

The optimum concentration can be explained on the basis of the reversible nature of the reaction, and so also the shift in the position of the optimum with respect to different hypo concentrations. The rise in the curves may be explained being due to the fact that higher complexes of ammonium thiosulphate and silver halides are unstable as compared to the higher complexes of sodium thiosulphate, and silver halides. The clearing time, however, is independent of the type of the emulsion as is expected.

![Graph](image)

**Fig. 3.** Emulsion thickness Vs % reduction in the clearing time.

2) A fall, and then a rise in the curves is obtained in case of all the three ammonical salts, as is explained on the basis of the explanations given above. The more pronounced rise observed in the case of ammonium chloride can be explained as being due to the presence of $<\text{Cl}>$ ion which affects the rate of fusion, as chlorine harden the gelatine of the emulsion, thereby increasing the clearing time by effecting the diffusion rate. As this factor is absent in the case of other two ammonium salts, the rise is not as pronounced in their case.

3) The percentage fall goes on decreasing in the case of higher thicknesses of NH$_4$Cl, whereas for other salts it remains constant after 200 microns. The reason for this is that upto 200 microns the diffusion velocity of the fixing solution increases with an increase in the thickness, and hence a fall in the curve. In case of ammonium chloride this effect persists beyond 200 microns, because of the contribution of the $<\text{Cl}>$ ion, which, however, is not the case with other ammonium salts. Also, the diffusion velocity becomes independent of emulsion thickness beyond 200$\mu$m, which explains the flatness of the curves beyond 200$\mu$m.

**Effect on the shrinkage factor and the grain size:**

Effect on the grain size was investigated so as to find out whether or not chlorine eats away the developed grains at the surface as reported by Piper.
For this, the grain size at the top and the bottom of the plate was found in case of pure hypo, and hypo with each of the other ammonium salts. (The optimum concentration being used in each case). The plates of the same type were simultaneously put in the four different baths and the fixing carried out under the same experimental conditions. The observations regarding the shrinkage factor and the grain size are shown in Tables I and II.

**TABLE I**

Observations regarding the shrinkage factor

<table>
<thead>
<tr>
<th>Plate thickness in μm</th>
<th>Pure hypo</th>
<th>Hypo with optimum concentration of NH₄Cl</th>
<th>(NH₄)₂SO₄</th>
<th>NH₄C₂O₄-H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>2.00</td>
<td>2.16</td>
<td>2.05</td>
<td>2.94</td>
</tr>
<tr>
<td>200</td>
<td>2.50</td>
<td>2.60</td>
<td>2.4</td>
<td>2.72</td>
</tr>
<tr>
<td>400</td>
<td>2.75</td>
<td>2.50</td>
<td>2.50</td>
<td>2.75</td>
</tr>
</tbody>
</table>

**TABLE II**

Observations regarding the grain size

<table>
<thead>
<tr>
<th>Thickness of the plate in μm</th>
<th>Surface of emulsion</th>
<th>Free hypo</th>
<th>Hypo having optimum concentration of NH₄Cl</th>
<th>(NH₄)₂SO₄</th>
<th>NH₄C₂O₄-H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>Bottom</td>
<td>0.5</td>
<td>0.75</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>Top</td>
<td>0.5</td>
<td>0.50</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>200</td>
<td>Bottom</td>
<td>0.75</td>
<td>0.75</td>
<td>0.5</td>
<td>0.75</td>
</tr>
<tr>
<td></td>
<td>Top</td>
<td>0.75</td>
<td>0.75</td>
<td>0.75</td>
<td>0.75</td>
</tr>
<tr>
<td>400</td>
<td>Bottom</td>
<td>0.5</td>
<td>0.75</td>
<td>0.5</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>Top</td>
<td>0.75</td>
<td>0.75</td>
<td>0.75</td>
<td>0.50</td>
</tr>
</tbody>
</table>

The study of the shrinkage factor, and the grain size reveals that within the allowed statistics, the shrinkage and the grain size is in no way affected by the addition of the ammonium salts. It also contradicts the hypothesis that chlorine is responsible for eating away the emulsion at the surface. The optimum concentration of the various ammonium salts used by the author are 1.5% for ammonium chloride, 2.5% for ammonium sulphate and 2.5% for ammonium acetate. Relative study of the suitability of these salts as regards the clearing of thick plates:

For thick plates the clearing time depends predominantly upon the diffusion velocity of the fixing solution, as against the thin plates. Consequently then
Role Played by Ammonium Salts in the Clearing, etc. 567

pH value of the fixing solution becomes an important factor. To make a relative study of the usefulness of these salts as regards the clearing of thick plates, pH values of the various concentrations of these solutions were calculated by the usual formulae Vogel (1958). The results are shown in Table III:

**TABLE III**

Observations regarding clearing time

<table>
<thead>
<tr>
<th>Cone. of the amm. salt %</th>
<th>0.5</th>
<th>1.0</th>
<th>1.5</th>
<th>2.0</th>
<th>2.5</th>
<th>3.0</th>
<th>3.5</th>
<th>4.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH of (NH₄)₂SO₄ (opt. conc.) + hypo (40%)</td>
<td>5.3</td>
<td>5.6</td>
<td>5.8</td>
<td>6.2</td>
<td>5.7</td>
<td>5.6</td>
<td>5.3</td>
<td>5.0</td>
</tr>
<tr>
<td>pH of NH₄C₂O₄H₂ (opt. conc.) + hypo (40%)</td>
<td>6.3</td>
<td>6.8</td>
<td>6.9</td>
<td>7.0</td>
<td>7.4</td>
<td>7.8</td>
<td>7.9</td>
<td>8.1</td>
</tr>
<tr>
<td>pH of NH₄Cl (opt. conc.) + hypo (40%)</td>
<td>6.1</td>
<td>6.5</td>
<td>6.7</td>
<td>6.8</td>
<td>7.2</td>
<td>7.7</td>
<td>5.4</td>
<td>5.2</td>
</tr>
</tbody>
</table>

The table clearly shows that the pH value of hypo + amm. sulphate and also hypo + amm. chloride solution is well within the acid region, thus ensuring a high diffusion velocity, and the pH value of amm. acetate being in the basic region, it does not have a high diffusion velocity. Therefore, amm. acetate + hypo is not as suitable for clearing as the other two salts. Amm. sulphate, however, has been found to be most suitable for this purpose.

**ACKNOWLEDGMENTS**

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