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Concerning Amidol Development of Nuclear Emulsions

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A lowering of the development temperature introduces development defects which are characterized by the partial or total destruction of the developed image and by a deep coloration of the emulsion. These defects are eliminated by increasing the sodium sulfite concentration of the developer. Up to now contrast in nuclear emulsions has been evaluated subjectively by the experimenter; this forbids any comparison of the results obtained in different laboratories.

This paper shows that to develop reliably at low temperatures with the usual developers, it becomes necessary to increase the ratio $(Na_2SO_3)/(Amidol)$. For development temperatures of 10–15°C the concentration ratio needs to be 70% greater than the ones commonly used.

A study of track and fog grain properties is carried out. It is shown that at a given temperature the mean gap length is practically independent of the development parameters. The optimum conditions at 14°C are determined with the help of a new quantity termed "track visibility" (K), a track to fog signal, and defined by K=blob length×track grain diameter/fog density×(fog grain diameter)².

INTRODUCTION

IT is generally accepted that a lowering of the development temperature reduces distortion in nuclear emulsions. Some authors[1,2] use monothermal development to reduce distortion. When this technique is used, temperature variations are replaced by pH variations. This procedure has certain drawbacks, and its complexity of operations is by no means the least of them. It has, therefore, been our aim to reduce distortion by developing at a lower than usual temperature (14°C), while at the same time preserving dithermal procedure.[3,4] At the temperature mentioned above, emulsions develop irregularly and very frequently the image does not appear at all, while the fixed emulsions acquire a blue or red coloration. These phenomena were observed at various research centers which use nuclear emulsions. These defects occur relatively infrequently at usual temperatures.

I. EXPERIMENTAL CONDITIONS

Glass-backed Ilford G5 emulsions having a thickness of 100 and 600 μ were irradiated at 30-Mev electrons (supplied by a betatron of Brown & Boveri Company, Baden†) for a track density of 35 000 to 45 000 electrons/cm².

Three developers were used called here A, B, and C (C_1 and C_2); their composition is shown in Table I. Table II shows time and temperature of the various operations.

TABLE I. Developer composition.

	A	B	C_1	C_2	K. Nagahama[a]
g/l H_3BO_3	35	25	25	25	
g/l KBr	0.8	0.8	0.8	0.8	
g/l Na_2SO_3	18	12	19	22	100
g/l Amidol	4.5	2.8	2.8	2.8	15
φ= $(Na_2SO_3)/(Amidol)$	6.2	6.8	10.8	12.5	10.6
pH	6.5	6.4	6.9	7.0	

[a] See reference 13.

II. ELIMINATION OF THE DEVELOPMENT DEFECTS

The emulsions of eight different batches, developed at 14° and 23°C, by developers A and B, were more or less faded and colored. These defects[5–7] are more frequent at 14° than at 23°C, and their intensity is a function of the time spent in the isothermal developing tank. Even in the case of total destruction of the developed image,[6] the latent image is not totally destroyed since it can be brought out by a mercuric chloride treatment. Certain emulsions show zones of good development while the rest of the plate is useless.

The emulsions are colored throughout the entire depth; this is not a surface phenomenon. The fixed emulsions show a blue or red transparency. Numerous experiments have revealed the following.

(1) These defects are not caused by accidental mistakes.

(2) Wet hot-stage development, with developers A and B, does not produce coloration or image destruction.

(3) Dry hot-stage development with the help of a Phenidone developer (1-phenyl-3-pyrazolidone) is accompanied neither by coloration nor by developed-image destruction.

We know furthermore that, when the same photographic

* Now at International Business Machines Corporation, Endicott, New York.

[1] H. Yagoda, Rev. Sci. Instr. **26**, 263 (1955).
[2] G. Marguin, Sci. Ind. Phot. **28**, 321 (1957).
[3] Dilworth, Occhialini, and Vermaeren, Bull. univ. Bruxelles **13a** (February, 1950).
[4] Bonetti, Dilworth, and Occhialini, Bull univ. Bruxelles **13b** (1951).

† I want to express my thanks to Dr. Wideroe and his collaborators.
[5] Fatzer, Weill, Gaillond, Haenny, Colloq. Phot. Corpusculaire, Strasbourg (1957).
[6] Vanderhaege, Colloq. Phot. Corpusculaire, Strasbourg (1957).
[7] Birge, Litkerth, Richman, Stork, Whetstone, Bull Univ. Cal. Radiation Lab. 2690.

TABLE II. Processing conditions.

	Water presoak	Developer presoak	Development time	Stop bath[a] 1% CH_3-COOH	Rinse
	T°C Hours ±0.1	T°C Hours ±0.1	T°C Hours ±0.1	T°C Hours ±0.1	T°C Hours ±0.1
100-μ emulsions	2° 0.5	2° 0.5	variable	2° 0.5–1	2° 1–2
600-μ emulsions	2° 4	2° 4	variable	2° 4	2° 4

	Fixation 45% $Na_2S_2O_3$ T°C Hours ±0.5 ±1	Washing T°C Hours ±1	Glycerinizing 4% glycerine T°C Hours ±0.1 ±1	Drying T°C Hours ±1 ±24
100-μ emulsions	6° 8	5–15° 24	5° 24	20° 168
600-μ emulsions	6° 168	5–15° 24	5° 24	20° 168

[a] Once the stop bath is finished, the fog is taken off by rubbing.

material was used, similar phenomena were observed in Belgrade, in Brussels, and at Berkeley[7] with an Amidol developer. This image destruction must not be confused with usual corrosion, which sets in during prolonged fixation and which begins at the surface of the emulsion. In our case, the disappearance of the image begins deep in the emulsion and is linked to the coloration. References 8 and 9 lead one to believe that these phenomena are due to the displacement of the adsorption equilibria of the organic compounds and Na_2SO_3 on silver halide grains. These defects may be explained by the equations

$$2AgNaSO_3 \rightarrow 2\underline{Ag} + Na_2S_2O_6$$

$$AgS_2O_3^- + Ag^+ + H_2O \rightarrow Ag_2S + H_2SO_4$$

proposed by H. Chateau *et al.*[10,11]

According to this interpretation, one can eliminate these defects by increasing the Na_2SO_3 concentration.[12] While keeping the Amidol concentration constant, that of sodium sulfite is increased; various values between 12 and 22 g/l are chosen.

It must be noted that these defects are equally frequent for emulsion thicknesses of 100 and 600 μ, however, the 600-μ emulsions are less colored than the 100 μ.

Emulsions developed with these different solutions reveal that the development defects disappear when the sodium sulfite concentration reaches 19 g/l ($\varphi = 10.8$), but remain quite serious at concentrations of 15 and 17 g/l of sodium sulfite. It is interesting to note that K. Nagahama[13] gives the formula of an Amidol developer for development of ordinary photographic emulsions at temperatures ranging from 0° to 10°C and which has a φ approximately equal to that of developer C_1. This very concentrated developer is useless for nuclear emulsions.

In conclusion, the emulsions develop reliably if immersed in the developer. In case the dithermal procedure is used with the usual developers, it may be stated that in order to avoid coloration of the emulsion and destruction of the developed image, phenomena which increase in frequency with decreasing temperature, it is necessary to choose a $(Na_2SO_3)/(\text{Amidol})$ ratio which is 70% greater than the ratios adopted in the Brussels developer (A) and the Chicago developer (B).

III. INFLUENCE OF THE TEMPERATURE OF THE HOT STAGE AND OF THE SODIUM SULFITE CONCENTRATION ON TRACK AND FOG STRUCTURES

Having found the means of eliminating the development defects, we tried to determine optimum development conditions. To this end we measured, for various development conditions, the following parameters, diameters of track (ϕ_t) and fog (ϕ_v) grains, volumetric fog density (D_v), mean gap length ($\bar{G}$) and blob density (D_b). The experiments were carried out on Ilford G5 emulsions of 100 μ thickness; these came from two different batches numbered $Z = 9998$ and $Z = 125$ by Ilford Ltd.

Diameter of Track Grains ($\bar{\phi}_t$)

The curves in Fig. 1 show that the mean diameter of track grains increases rapidly in the region of underdevelopment and reaches a constant value for longer development periods. Increase in sodium sulfite concentration as well as increase in development temperature cause a decrease in grain diameter. These two variations can be explained by the solvent effect of sodium sulfite on silver halides. The statistical error, which corresponds to a 95% confidence interval is given by: $\epsilon = 2\sigma/N^{\frac{1}{2}}$ where σ is the standard deviation, and N is the number of measurements taken.

Diameter of Fog Grains ($\bar{\phi}_v$)

The silver grains which compose the fog are grouped in two classes. One of them is composed of large grains and

[8] T. H. James and W. Vanselow, Sci. Ind. Phot. **23**, 144 (1952); **26**, 233 (1955); **25**, 243 (1954); **23**, 36 (12952).
[9] T. H. James and W. Vanselow, Colloq. Emul. Phot. 122, Revue Optique, Paris (1953).
[10] H. Chateau and J. Pouradier, Sci. Ind. Phot. **27**, 81 (1956); **25**, 305 (1954); **27**, 465 (1956).
[11] M. Durante and J. Pouradier, Sci. Ind. Phot. **28**, 194 (1957).
[12] This prescription was given by Birge *et al.*, reference 7.
[13] K. Nagahama and M. Egaschira, Sci. Ind. Phot. **22**, 16 (1951).

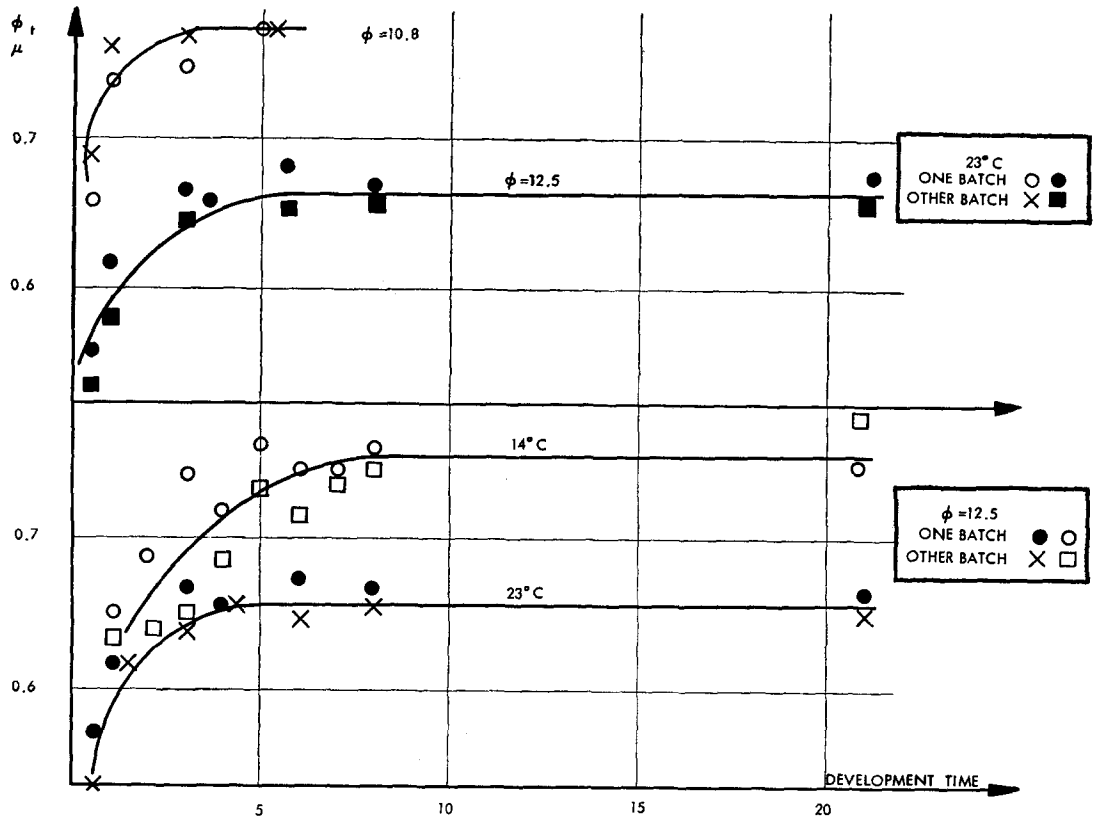

FIG. 1. Track grain diameter in μ for two batches, two Na_2SO_3 concentrations and two temperatures. Error $\pm 0.04\ \mu$.

the other of small grains. The former appear rapidly during development and increase in number in the same way as track grains, whereas the latter, quite infrequent for underdevelopment, appear suddenly and in great number for an average hot-stage time.

From the curves giving the mean diameter of fog grains as a function of development time, conclusions can be drawn.

(a) The mean diameter remains constant during the time interval here considered, namely, $\frac{1}{2}$ to 21 hr.

(b) The mean diameter of fog grains is practically independent of development temperature.

(c) For a given emulsion, the mean fog grain diameter is smaller than that of track grains.

(d) The increase in sodium sulfite concentration causes a small increase of the mean diameter, for the oxidation-reduction potential decreases and the developer becomes more energetic.

The calculation of the mean diameter ϕ_v was based on some 50 grains, ranging in diameter from 0, 2 to 1, 3 μ. The lower limit corresponds to the resolving power of the microscope; the upper limit is the end point of the distribution in diameters. The error is given by $\epsilon = 2\sigma/N^{\frac{1}{2}}$.

Summarizing the results of the grain diameter measurements, we can state that the mean diameter of track grains decreases when development temperature and sodium sulfite concentration are increased. While under the same conditions the mean fog grain diameter remains constant or increases, respectively.

The above study of track and fog properties as a function of the usual development variables reveals a difference in behavior between fog and track grains.

Volumetric Fog Density D_v (See Fig. 2)

The volumetric fog density increases when the hot-stage time, the temperature, or the sodium sulfite concentration are increased.

The error shown is the root of the squares of the following contributions: the statistical error in the number of grains per emulsion column, the error in the thickness of the developed emulsion (4%), the error in the area of a basic grid square (2%), and the error due to the observer.

Blob Density D_b

The blob density is given by the ratio of the number of blobs n to the length of the measured track L_t. A correction (Δn) must be introduced for the presence of the fog grains on the track, and blob density is given by: $D_b = 100(n - \Delta n)/L_t$.

Δn was calculated with the help of the fog density and total gap length.

Figure 3 shows that blob density increases in the region of underdevelopment and then reaches a constant value when the hot-stage time is increased. A temperature increase, on the other hand, causes a significant increase in blob density whatever the duration of the hot-stage time.

The error shown is the root of the squares of the following contributions: the statistical error in the number of blobs, the error due to the observer (~1.5%), and the error due to the fog correction (~1.5%).

Total Gap Length L_1

As can be seen from Fig. 4, total gap length L_1 decreases in the region of underdevelopment, with increasing hot-stage time; indeed, as previously seen, track grain diameter increases under these conditions. At longer development periods, including those resulting in overdevelopment, L_1 tends to become constant. At any development time total gap length decreases with increasing temperature and is independent of the sodium sulfite concentration

Mean Gap Length $\bar{G}$

Mean gap length is found by dividing total gap length L_1, by the number of blobs n. Corrections Δn and Δl must

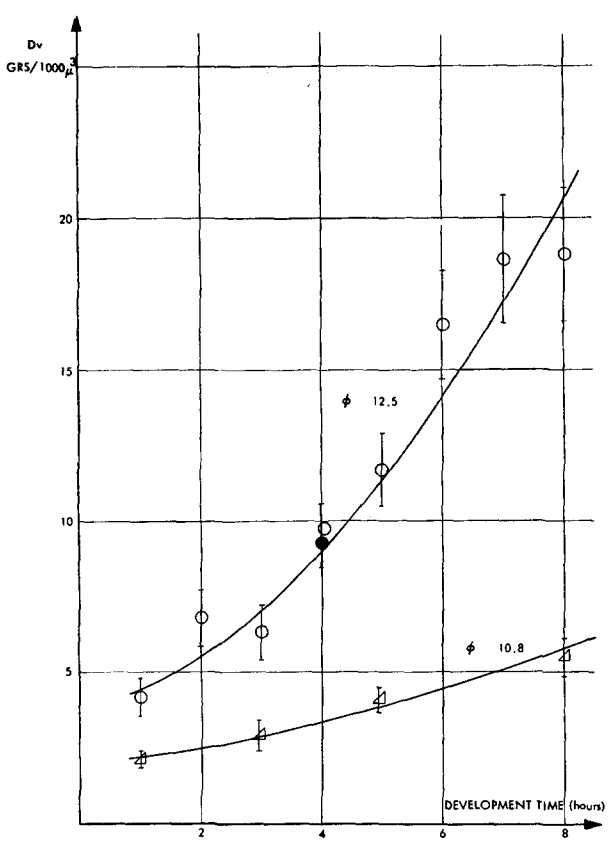

FIG. 2. Volumetric fog density D in grains/1000 μ^3 for one temperature and two Na_2SO_3 concentrations.

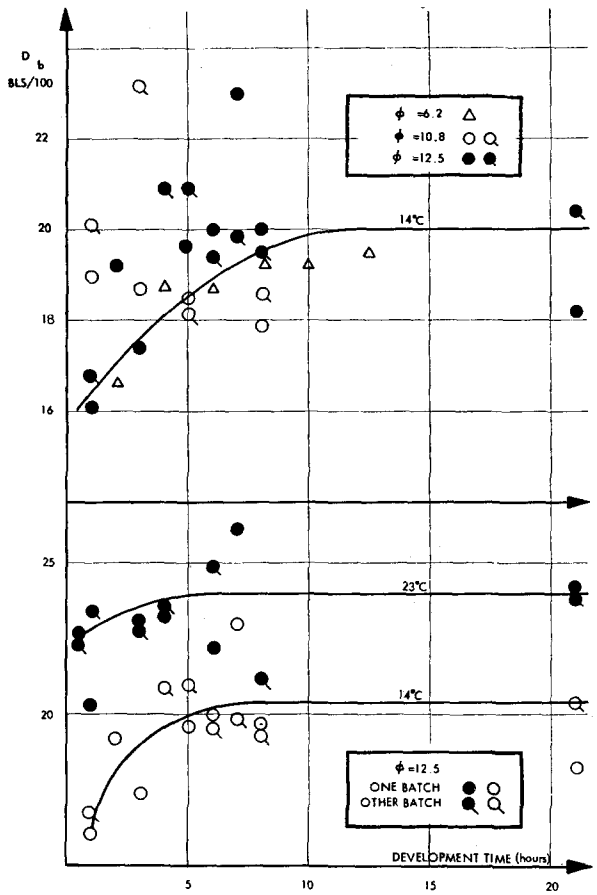

FIG. 3. Blob density in blobs/100 μ for two batches, three Na_2SO_3 concentrations, and two temperatures. Error ±0.9 blob/100 μ.

be applied for the presence of fog grains. The corrected mean gap length is: $\bar{G}=(L_1+\Delta l)/(n-\Delta n)$, where $\Delta l=\Delta n$, $\bar{\phi}_v$ with Δn=number of fog grains on the track; Δl=fog grain length on track; $\bar{\phi}_v$=mean fog grain diameter.

From the plots in Fig. 5 it can be seen that the mean gap length $\bar{G}$:

(1) is independent of the duration of the hot stage, except in case of underdevelopment;

(2) is practically independent of the sodium sulfite concentration, the three developers considered being A, C_1, and C_2;

(3) decreases with increasing temperature.

The error shown is the root of the squares of the following contributions: the statistical error in the mean gap length $\bar{G}/n^{\frac{1}{2}}$, the error due to the observer (2–3%), and the error due to fog correction (2–3%).

Summarizing the results of mean gap-length measurements, it can be stated that at a given temperature the mean gap length is, to a considerable extent, independent of the development parameters, development time, and sodium sulfite concentration.

Blob density, total gap length, and mean gap length

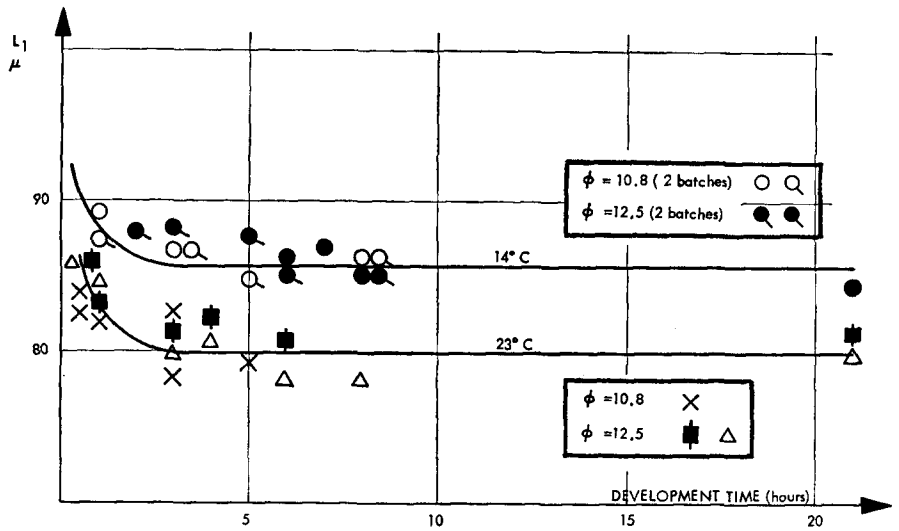

FIG. 4. Total gap length $L_1/100\ \mu$ track cell for two batches, two Na_2SO_3 concentrations, and two temperatures.

were measured with the help of an automatic device called a gap meter.[14]

Table III shows that the variations of $\bar{G}$ agree with those found for ϕ_t, L_1, and D_b. Thus our results appear to be internally consistent.

IV. TRACK VISIBILITY K

The precision of nuclear measurements is obviously linked to track and fog properties. Authors working with nuclear emulsions mention contrast but experimenters evaluate it quite subjectively, and this excludes *a priori* any valid comparison of the results obtained in different laboratories. We have tried to close this gap and suggest the following definition of a quantity K called "*track visibility.*"

$$K = L_g \cdot \phi_t / D_v \cdot \phi_V^2,$$

ϕ_t = track grain diameter,

ϕ_v = fog grain diameter,

D_v = fog density,

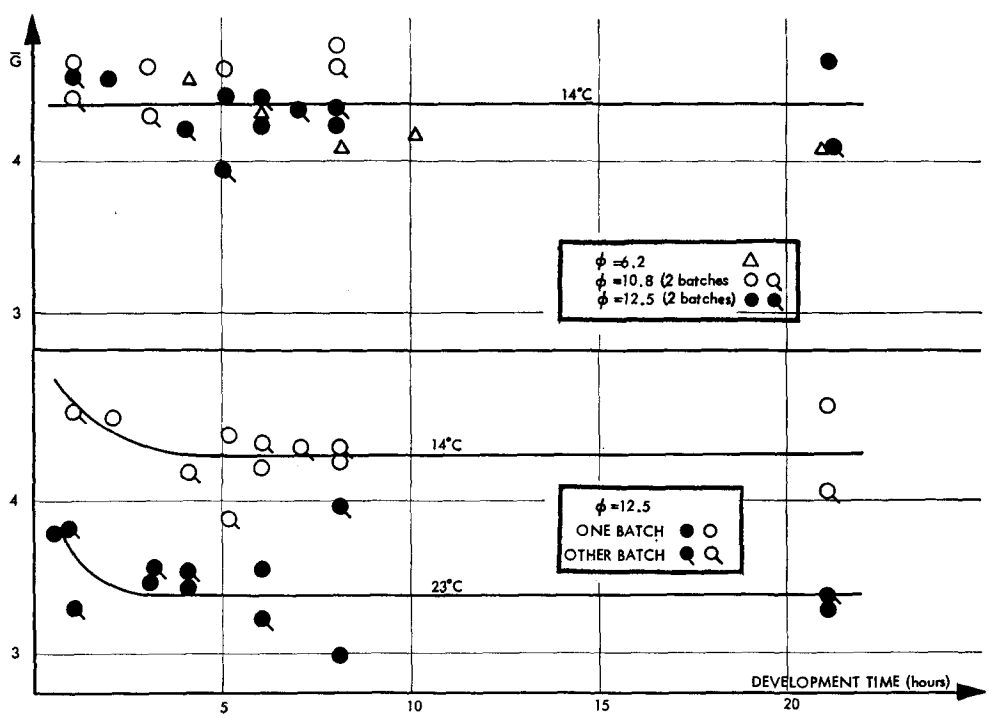

FIG. 5. Mean gap length $\bar{G}$ in μ for two different batches, three Na_2SO_3 concentrations, and two temperatures. Error $\pm 0.2\ \mu$.

[4] Weill, Joseph, Gailloud, and Rosselet, presented at the Swiss Physical Society meeting, September, 1957.

TABLE III. Summary of the measured variations.

	Time	Temperature	Na_2SO_3 concentration
(a) Case of normal development			
	$t+$	$T+$	$C+$
Grain diameter (ϕ_t)	0	−	−
Total gap length (L_1)	0	−	0
Blob density (D_b) or n	0	+	0
Mean gap length ($\bar{G}=L_1/n$)	0	−	0
(b) Case of underdevelopment			
	$t+$	$T+$	$C+$
ϕ_t	+	−	−
L_1	−	−	0
D_b or n	+	+	0
$G=L_1/n$	−	−	0

+, 0, − represent increasing, constant, and decreasing values.

where L_g is given by the expression

$$L_g=100[L_t-(L_1+\Delta l)]/L_t,$$

L_t = track length,

L_1 = total gap length,

Δl = fog grain length on track.

L_g is the sum of the lengths of all the grains in a 100-μ track segment. The numerator of the expression for track visibility is the area of all track grains projected on the observation plane; the denominator is proportional to the projection, on the same plane, of the area of all fog grains. Figure 6 shows track visibility plotted as a function of the hot-stage time for a few values of development temperature and sodium sulfite concentration. The maximum of the (14°C) track visibility curve gives a blob density of 20 blobs/100 μ. It can be seen that hot-stage temperature has little influence on track visibility, while an increase in sodium sulfite causes it to decrease strongly. The optimum development conditions are: a hot-stage temperature of 14°C, with a duration of three hours and a sodium sulfite concentration of 19 g/l, which at this temperature is the least concentration sufficient to eliminate development defects. It may be noted that one is not certain to obtain normal development after a three hour hot-stage time; see the plateau in Fig. 5. In order to assure better reproducibility, it might be useful to choose a somewhat longer hot-stage time, for example, 3 to 5 hr. The quantity K objectively expresses contrast variations in terms of track and fog properties.

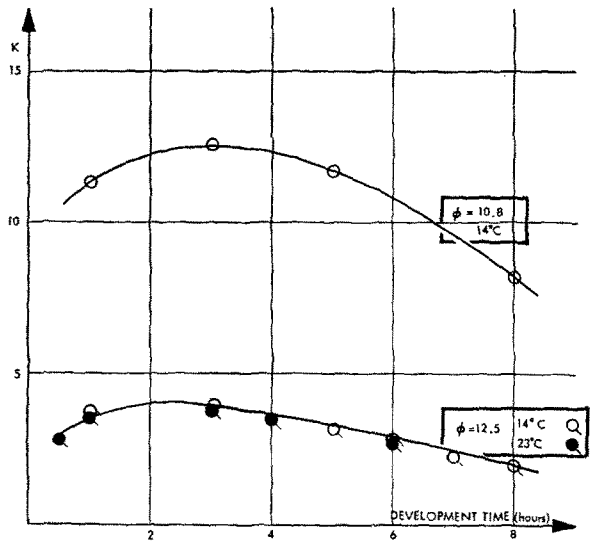

FIG. 6. Track visibility K for two temperatures and two Na_2SO_3 concentrations.

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