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Investigation of Effects of Ph, Ionic Strength,
and Drying Temp. on Dimensional Stability of Film

by

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Submitted in Partial Fulfillment
of the Requirements for the Degree
Bachelor of Science

School of Photographic Arts and Sciences
College of Graphic Arts and Photography
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SENIOR RESEARCH THESIS

MAY 1, 1964

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An Investigation Into The Effect
of
pH, Ionic Strength, and Drying Temperature
On the Dimensional Stability of a Fine
Grain Printing Ink

Master's Thesis

Harry N. Parsonage

George A. Pittman

Dedicated To:

Our loyal, devoted and understanding

Wives - JoAnn and Judy

ABSTRACT

An investigation into the effects of chemical changes made in a processing system and drying temperature with respect to dimensional displacements of a final image was carried out using Eastman Kodak's # 8430 aerial duplicating film. Image displacements over both long and short distances were measured.

Over approximately 162 mm., drying at a temperature equal to that of processing, 70°F, resulted in no significant spatial displacement of the final image, while drying at higher temperatures produced correspondingly larger displacements. A pH system between baths similar to that found in a conventional process resulted in dimensional displacements, over the short distance or approximately 90 microns, less than processing systems employing gradually increasing or equal pH's throughout the baths. The salt concentrations investigated produced no significant effect on dimensional displacements.

An Investigation Into The Effect Of
pH, Ionic Strength, And Drying Temperature
On The Dimensional Stability Of A Fine
Grain Printing Emulsion

Previous investigations of dimensional displacement of the photographic image have been more concerned with mapping the actual displacements under a given set of processing, drying and/or atmospheric conditions. Conceivably previous workers had hoped to map the distortions to be expected so that the effects due to these factors could be removed from data obtained from films and plates. It was the intention of the authors to investigate the effects of quantitative changes of the process on the dimensional displacements of a photographic image. Displacements of the photographic image appear to be due primarily to the expansion and contraction of the gelatin and the support.

To investigate the effects of such changes three factors were chosen that have a direct bearing on swelling of gelatin and on expansion and contraction of the base. The three factors investigated were:

1. Changes in pH between processing solutions.
2. Changes in salt concentration between processing solutions.
3. Drying temperature.

Changes in pH---¹ As film is put in a solution the gelatin expands. When the film goes from this solution to the acidic stop bath it goes through a region of high pH for the gelatin, the isoelectric point, and then swells again reaching the low pH of that solution. Fixation and washing can change the swelling of the gelatin depending on the hardness and pH of the fixing bath. Hence, changes in pH during the photographic process cause stresses on the film through successive expansions and contractions of the gelatin. (See Diagram # 1) •

Changes in salt concentration---² Swelling of gelatin is usually lowered by increasing the salt concentration of the solution. A developer, of somewhat high salt concentration, is followed by a dilute stop bath, which in turn is followed by a fixer of higher salt concentration than that of the developer. Tap water used for washing has no salts of any consequence. Therefore, as with pH changes within a given system, the salt concentration changes between solutions in the photographic process cause successive expansions and contractions of the gelatin. These may be contributing factors to the dimensional displacements of a photographic image. (See Diagram #2)

Drying temperature---³ An increase in temperature generally causes film to expand. But, if the increase in temperature is not accompanied by an increase in absolute humidity, the relative humidity will decrease. The film will contract with a corresponding decrease in relative humidity (RH). During the drying process the film is subjected to both high temperatures and high RH at the beginning of drying, causing expansion. As drying continues and the film dries the RH decreases causing the film to contract. The drying process imparts

3.

both reversible and irreversible changes on the film. The Reversible, or temporary, changes occur due to the expansion of the film according to the temperature coefficient while the irreversible changes are due to the shrinkage caused by loss of solvents from the film base. Here again there are opposing forces within the film that could lead to dimensional distortions in the final image.

Printing System

The film samples were printed with ultra violet radiation, 3400-3800 A, in contact with a master glass plate. The master was a spectroscopic plate with an image of fine ruled lines (similar to graph paper). The lines were transparent, approximately 0.1 mm wide, on a dense background and formed squares four millimeters on a side. The master glass plate was 9 1/4 X 1 inches. Five inches of the center was transparent. In this transparent area the step tablet was placed. (See Fig. 1) The master and step tablet were placed in the platen of a Pittman Model B Control Sensitometer (xenon flash)⁴ The sensitometer was modified to print the step tablet by light and the reseau grid pattern by ultra violet radiation, as isolated by using the Wratten 18A filter. The ultra violet was used to print the grid in order to print as sharp a line as possible with this printing system. The light exposed step tablet was used to check sensitometric control of the process. Exposures were made in contact under a pressure of three pounds on the platen.

Processing Conditions

pH Systems --- Two systems of gradually changing pH between baths and one of no changes in pH between baths to a system of pH changes between baths similar to that of the normal process, where the changes are very large. (See Appendix for details.)

Solution	Gradual Increase	Equal	Normal*	Gradual Decrease
Developer	4.00	7.00	8.70	10.00
Stop Bath	5.00	7.00	2.82	9.00
Fixer	6.00	7.00	3.88	8.00
Wash	7.00	7.00	7.00	7.00

Table 1

pH systems investigated

*Normal pH values obtained from measurements of freshly prepared solutions of Kodak's formulas: D-76, SB-1, & F-5.

Salt Concentrations Systems --- Salt concentrations were expressed as ionic strength in order to take into account the effects of the charges on the ions. The relationship used for the estimate of ionic strength was:

$$I = 1/2 \sum (m_i \cdot z_i^2)$$

where I represents ionic strength and m_i is the concentration, expressed as molality, of each ion with a charge of z_i . (See appendix for sample calculations) Since the film must start at a high ionic strength, I,

in the developer and go to the wash of I = 0.
 strengths must necessarily start at a ...
 Three systems of ionic strengths were formulated and
 fourth system representing the normal or conventional
 formulated systems consisted of two rates of gradual
 between baths and a third system of ionic strengths equal to ... of
 the developer for all baths except for the wash which was zero. (See
 table 2 for values) (Diagram # 2)

Solution	Normal* Changes	1/2, 1/4 Decreases	3/4, 3/8 Decreases	Equal
Developer	2.26	2.26	2.26	2.26
Stop Bath	0.24	1.12	1.69	2.26
Fixer	3.51	0.56	0.86	2.26
Wash	0.0	0.0	0.0	0.0

Table 2

Ionic strength systems investigated

*Normal ionic strength values calculated from Kodak's formulas:

D-76, SB-1, & F-5.

Drying Temperature --- The temperature equal to that of
 processing and two higher temperatures were compared.

Temperature °F	70	100	130
Time in min.	0-54%	28%	18%
	15 45%	22%	12%

Table #3 . % RH at various drying times and temperatures.

Film Handling

Eastman Kodak's 8430 aerial duplicating, 70 mm tri-acetate base film was used in the investigation. The film was removed from the spool and pre-conditioned to 50% RH for twenty-four hours prior to exposure. All of the film for the experiment was exposed at $47 \pm 1\%$ RH at $21 \pm 1^\circ\text{C}$ using the method previously described at 20 second recycle times.

Processing solutions, including the wash and wetting agent, were held at $21 \pm 1^\circ\text{C}$. The relative humidity of the laboratory during processing was $53 \pm 3\%$ RH. The exposed strips were placed on a process rack to enable three different treatment combinations to be run at once.⁵ Each strip was processed in a freshly prepared liter of developer, stop bath, and fixer. The samples remained on the rack for processing and washing. After washing, the samples were bathed in a 2% wetting agent before being squeegeed. The squeegeing process was done using a roller driven film squeegee machine employing two viscose sponges inclined 30° to the film.⁶ Each film sample was dried horizontally in the center of a shelf of the Pittman & Parsonage Control Strip Dryer.⁷ The dryer had capabilities of $\pm 1/2^\circ\text{C}$ temperature control within the ranges used.

Processing Solutions

Developer --- A ferrous ethylenediaminetetraacetic acid (Fe_2EDTA) developer similar to that formulated by James⁸ was chosen because of its low pH dependency with respect to its activity. At the concentration

used the developer produced approximately equivalent characteristics at pH values between 4.00 and 10.00.⁸ Developer activity is primarily pH independent the density is not. As pH increases the pK increases thus increasing the ionic strength of the developer.⁹ Sodium sulfate was used to maintain a constant ionic strength of 2.20 with changes in developer (see appendix for sample calculations) Since sodium ions were present from the chelating salt Na_4EDTA and sulfate ions were present from the FeSO_4 , the use of Na_2SO_4 to balance ionic strength added no new constituents to the developer. The pH of the developer was adjusted to the desired values using glacial acetic acid. (See Table 4 for formulation at various pH values)

Stop Bath --- Since the developer is pH independent with respect to activity the use of an acid stop as in the conventional process could not be done. The technique of a bromide stop bath as used in processing nuclear emulsions was used.¹⁰ A bromide bath of 23 1/2 g/l KB r stopped development as well as a conventional stop bath when using the Kodak formula D-76 developer.¹¹ The pH of the stop baths were maintained with low ionic strength buffers.¹² Again sodium sulfate was used to adjust the total ionic strength of the solution as required.

Fixer --- A fixer that functioned in both acidic and basic states with unusually low ionic strength was formulated using ammonium thiosulfate as the silver halide solvent.¹³ Sodium formaldehyde bisulfite

* See Figure #4

Developer

	PH	Grad.	Equal	Normal	Control
Na ₄ EDTA		60	60	60	60
FeSO ₄		20	20	20	20
Na ₂ SO ₄		27.5	27.5	27.5	27.5
H.C ₂ H ₃ O ₂ *		40 ml.	40 ml.	40 ml.	40 ml.
Water to make		1.0 liter	1.0 liter	1.0 liter	1.0 liter

Stop Bath

KBr	23.5	23.5	23.5	23.5 gms.
Buffers ^{6*}	A	B	A	C
Na ₂ SO ₄				
I - Normal	0.0	0.0	0.0	0.0 gms.
I - Grad. 1	42	42	42	42 gms.
I - Grad. 2	69	69	69	69 gms.
I - Equal	91.5	91.5	91.5	91.5 gms.
Water to make	1.0 liter	1.0 liter	1.0 liter	1.0 liter

Fixer

(NH ₄) ₂ S ₂ O ₃ (60%)	35 ml.	35 ml.	35 ml.	35 ml.
SFB	4.5	4.5	4.5	4.5 gms.
Buffers **	A	B	A	B
Na ₂ SO ₄				
I - Normal	144	144	144	144 gms.
I - Grad. 1	0.0	0.0	0.0	0.0 gms.
I - Grad. 2	20	20	20	20 gms.
I - Equal	87	87	87	87 gms.
Water to make	1.0 liter	1.0 liter	1.0 liter	1.0 liter

in low concentrations provided the necessary format for pH's above and below 7.00. The total ionic strength of the solutions was adjusted as required using sodium sulfate. Low ionic strength solutions similar to those used in the stop bath were used to maintain the pH of the solutions.

Method of Measurement.

The glass master and film samples were stored at 50±1% RH at 19±2°C for eight days prior to transportation to Data Corporation for measurement. Long distance measurements were made on the Mamp-Data Micro-Analyzer* which was used as a micro-comparator. The Whitfield room, in which the microcomparator was located, was held at 50±1% RH and 23.9±.5°C for the three days of measurement. The instrument was calibrated with a glass step tablet (for measurement of the glass master) and with a film step tablet (for measurement of film samples) to account for the different bases of the specimens. Focusing of the instrument was done electronically using the maximum fluctuations of the recording volt meter. This was the best estimate of a sharp focus for the granularity would be at its greatest value.

The samples were placed on the scanning stage and aligned with the X axis of the stage using the edge of the film. Further alignment with the X axis was accomplished by pivoting the stage in the Y direction until the line being measured was perpendicular to the X axis. In order to start at the same position on the line the sample was moved in the Y direction a constant distance (.771 mm) down from the horizontal line that is common to both sides of the master. The stage

*See Figure #10

was moved manually in the X direction until the distance of the vertical line was 1.0 as indicated by the recorder. A second measurement was made on the same line 1.42 mm from the previous measuring point. The screw positions for these two were recorded. Corresponding measurements were made on a similar line at the other end of the film sample. In all cases the measurements were made on the same side of the vertical lines of all the samples. (See fig. 2) Making measurements from the corresponding sides of the lines reduced the error due line width expansion and/or contractions.

Exact alignment of the samples with the direction of measurement was impossible. Therefore, not only would the distance between the two lines be incorrect due to the slight angle of the scan but the chance of measuring to imperfections on the line was increased. In order to compensate for the slight angle of scanning the second line was measured as was the first. The distance in the Y direction that the stage had to be moved to bring the measuring point the same distance below the horizontal reference line (0.771 and 2.313 mm) as for the first line was recorded. The displacement in the Y direction was an estimate of non-alignment and rotation in the sample. (See fig. 3)

Screw position data and displacement in the Y direction due to non-alignment were punched on cards and by the use of the IBM 1620 computer, the actual distances were calculated.¹⁵ The primary mechanism of the program was the use of the Pythagorean Theorem. The actual distance was the hypotenuse of a right triangle in which one leg was the displacement due to rotation and the other leg was the distance measured.

Line widths were measured with the Ansc Microdensitometer. Because the line width was 0.1 mm there was no problem of edge alignment. The line widths were measured at a density of 0.8. The line position for the short measurements was located at a constant distance down from the horizontal line with the long measurements. (This distance was 0.771 mm and was visually placed) Similar to the long measurements the screw was turned manually moving the sample along until the recording volt meter indicated a density of 0.8. The screw position was recorded and the stage backed off. The stage was moved forward after taking up any slack in the screw threads until the recorder indicated that the other side of the line had been reached. The stage was moved until the recorder came down to a density of 0.8; then the screw position was recorded. Duplicate readings were made on line widths.

The glass master was measured on both instruments. Using the micro-comparator the distance between the two lines chosen was measured (long measurements) at an edge density of 1.0. The Ansc Microdensitometer was used to measure the line width using the 18A filter over the photocell. Focusing was done with maximum fluctuations of the recording volt meter. Several traces were made using different degrees of knife edge focus to find the best estimate of the original line width. Thus, the line was measured in a similar manner as this line printed the samples, both with the same UV radiation and both with diffuse illumination on the master.

Sensitometric Adjustments Of Line Widths

The processing systems were not exactly equivalent in sensitometric characteristics that they produced. Any difference that would be found in the line width when one of the processing constituents were altered could be caused from either:

1. the chemical change made to the processing system.
2. the slight sensitometric differences that were a result of these chemical modifications.

An analysis of the line width measurements indicated that, with an alpha risk of 0.001, there was a significant difference in line widths due to the pH systems used. However, associated with each of these pH systems there was a slight sensitometric difference (See fig. 4).

The sensitometric difference could have caused a larger line width to occur than if the pH alone was changed (with no accompanying sensitometric shift). There may have been no line width change at all. The solution to this problem lies in an analysis of the components that contributed to the line width difference and then factoring out that which can be attributed to the sensitometry.

All of the samples were measured at a constant density of 0.8. Since the shape of the characteristic curve is different for each sample, each of these 0.8 densities was printed by a different Log E. (See fig. 5) Because the edges of the line on the master had some finite slope, the edges acted as a continuous wedge modulating the exposure to the edge of the sample. The Log E produced from the edges of the master's line was also finite. The Log E difference required to produce a constant density on the samples with different characteristic curve shapes is

equal to the density difference of the samples. Therefore, the line width of one sample was not necessarily the same distance on the master as another sample. The solution to the problem, then, is to go to the original sensitometry associated with each sample and find what the distance was on the master that printed each sample. Subtracting the samples distance from the distance that printed that particular sample would then give the deviation in line width due only to the pH changes made, having eliminated the effects due to sensitometry entirely.¹⁶

With a 99.9% confidence there was a significant difference in line width due to the drying temperatures used.¹⁷ Just as with the pH systems used, there was a significant difference in speed and slope with respect to the different drying temperatures. Here again, there is the problem of which caused the change in line width:

1. the different drying temperatures, or
2. the different density levels caused by the changing of drying temperature.

When the corrections were made for the sensitometric differences from the pH systems, the corrections for sensitometric differences due to the drying temperatures was also done. It would be impossible to factor out just the effects of pH since each sample had some drying temperature. Fig. 8 indicates the relation of drying temperature to line width before (with falsified distances due to sensitometric differences), and after with the sensitometric differences with each temperature factored out.

RESULTS

Treatments	Deviations from the master over		S ₁	Average Maximum Slope
	162 mm.	0.1 mm		
Drying Temperature	in microns			
70°F	- 0.7	- 4.3	57.4	1.97
100°F	-27.9	- 4.6	53.2	1.98
130°F	-29.1	- 4.1	54.9	2.03
pH Systems				
Normal	-22.0	- 5.0	63.4	2.02
Decrease	-19.9	- 5.1	63.4	2.05
Equal	- 9.9	- 3.6	52.2	1.98
Increase	-16.7	- 3.7	50.7	1.92
Ionic Strengths				
Normal	-13.5	- 4.6	54.6	1.98
1/2, 1/4	-15.9	- 4.1	55.2	2.01
3/4, 3/8	-14.5	- 4.2	55.9	1.98
Equal	-13.9	- 4.3	54.7	1.97
Confidence Limits	± 4.34 μ	± 0.35 μ	± 1.58	± .027

Discussion of Results

Long measurements, 162 mm --- Since there was a change in temperature of 3.9°C from the time of exposure to measurement, the film and glass master both expanded. The glass master expanded 17 while the film samples expanded 29, according to their respective temperature coefficients. 24 was subtracted from the film measurements for the final averages of the long measurements.

Line widths, 0.1 mm --- Because of mechanical and optical error in the microdensitometer the measured edge of a line would be at a lower slope than that of the actual edge. An error of only one micron would be multiplied by four because of the system being used to obtain a final measurement of the difference in sample line width and master line width. A factor of two from the two samples being compared, and a factor of two because there are two sides to each sample. (See fig. 9)

Therefore, the values given for the deviation from the master over the line widths are incorrect. But, the relation between the numbers are the same. In this case, the largest negative number (-5.1) is the 'best' because it is the smallest deviation.

Arithmetic speed --- The arithmetic speed system was based on the log exposure required to produce a density that is 0.3 above base plus fog.¹⁸ This corresponds to a density just into the straight line of the characteristic curve.

Average Gradient --- The maximum gradient obtained over a 0.6 log exposure range as found by the IBM 1620 computer with H.N. Parsonage program #3.¹⁹

Conclusions

Long measurements, 162 mm --- With a beta risk of 0.05, no significant difference was found over the long measurements of the pH and ionic strength systems investigated.

With an alpha risk of 0.01, there was found a significant difference in the long dimensions with respect to drying temperature. The 70°F temperature produced an image with dimensions closest to the original dimensions of the master. The two other temperatures resulted in film with dimensions significantly less than that of the master.

Line widths, 0.1 mm --- With a beta risk of 0.05, no significant difference was found in line widths with respect to any of the ionic strength or drying temperatures used. Although there was a significant difference in line width with a change in the drying temperature, it was not the heat itself that caused the change, but the sensitometric differences that resulted from those temperature differences. With no sensitometric differences, there would be no significant difference in line width at any of the temperatures tested.

With an alpha of 0.05, there was found a significant difference in line width with respect to the pH systems used. The Normal and Gradually decreasing pH systems produced lines significantly smaller than the other two systems.

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FOOTNOTES:

1. Pages 64 - 67 Reference 4B
2. Pages 18 - 19 Reference 2B
3. Page 10 Reference 3P
4. Reference 4R
5. Page 2 Reference 1R
6. Page 3 Reference 2R
7. Reference 5R
8. Page 19 Reference 1M
9. Page 35 Reference 5B
10. Page 3 Reference 2P
11. Page 4 Reference 3R
12. Page 951 Reference 3B
13. Page 3 Reference 3R
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18. Program #3 Ibid
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Diagram # 1

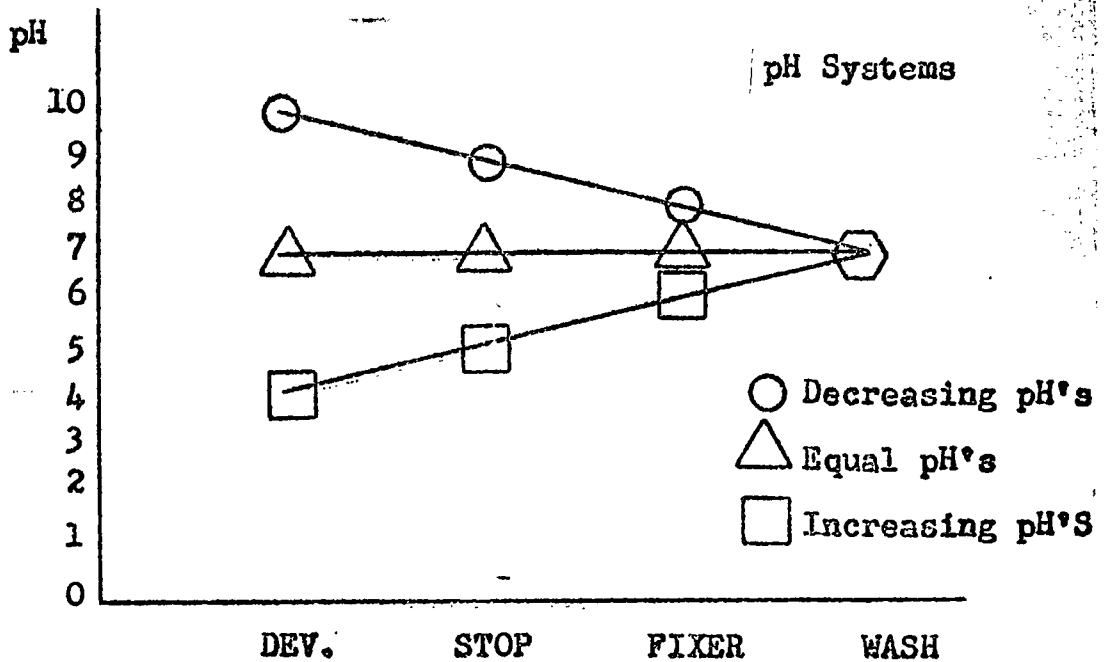
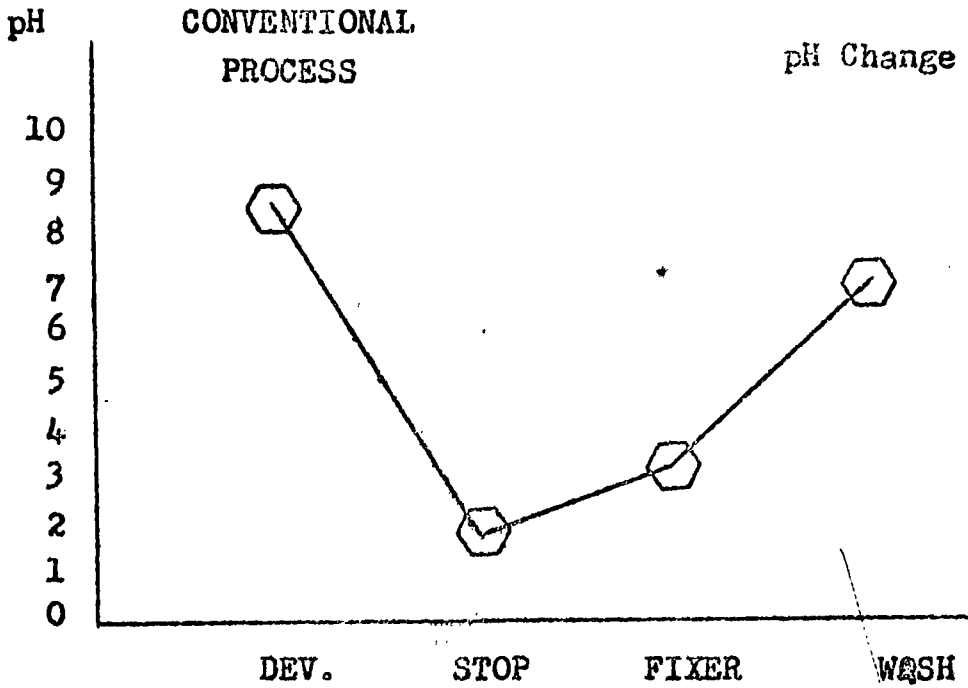
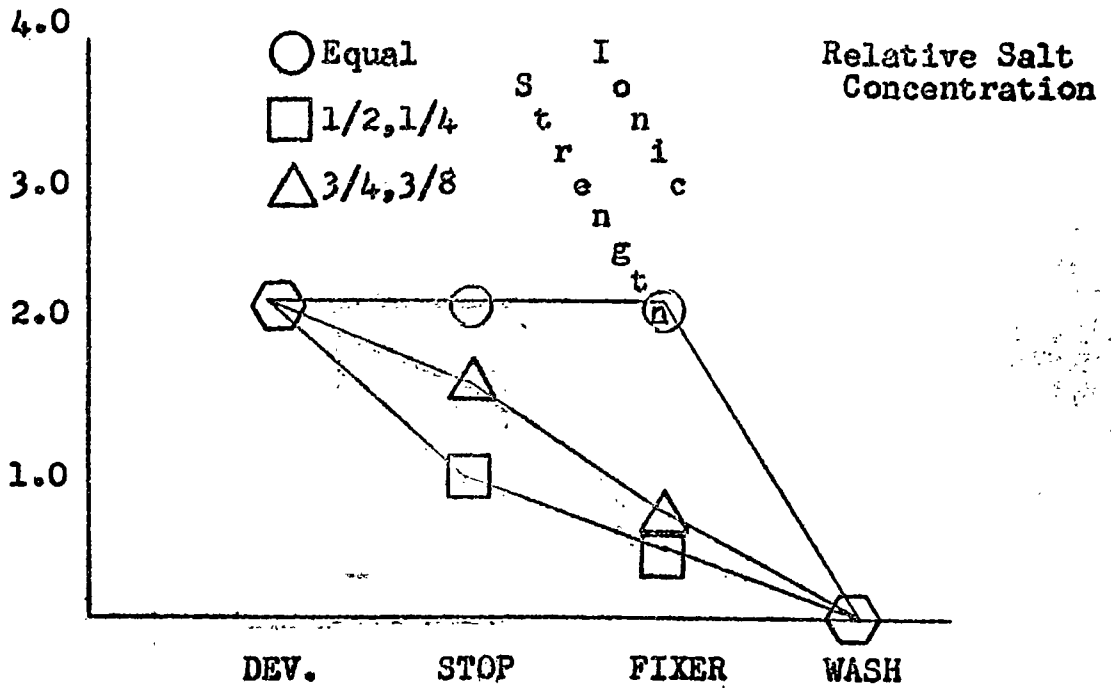
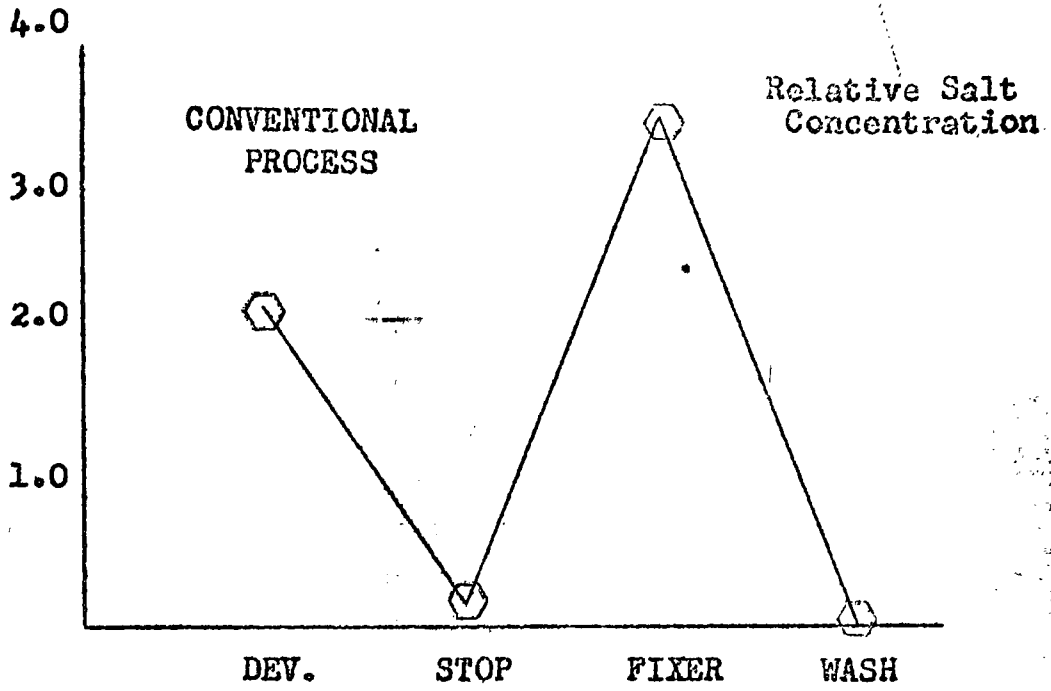
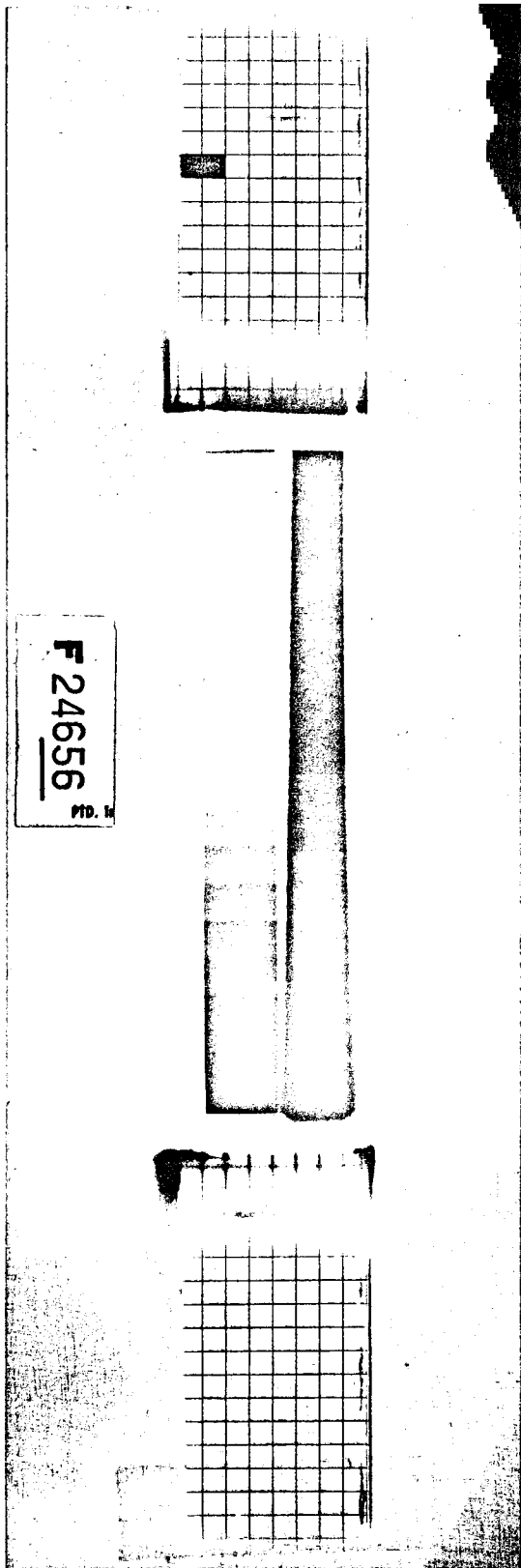


Diagram # 2

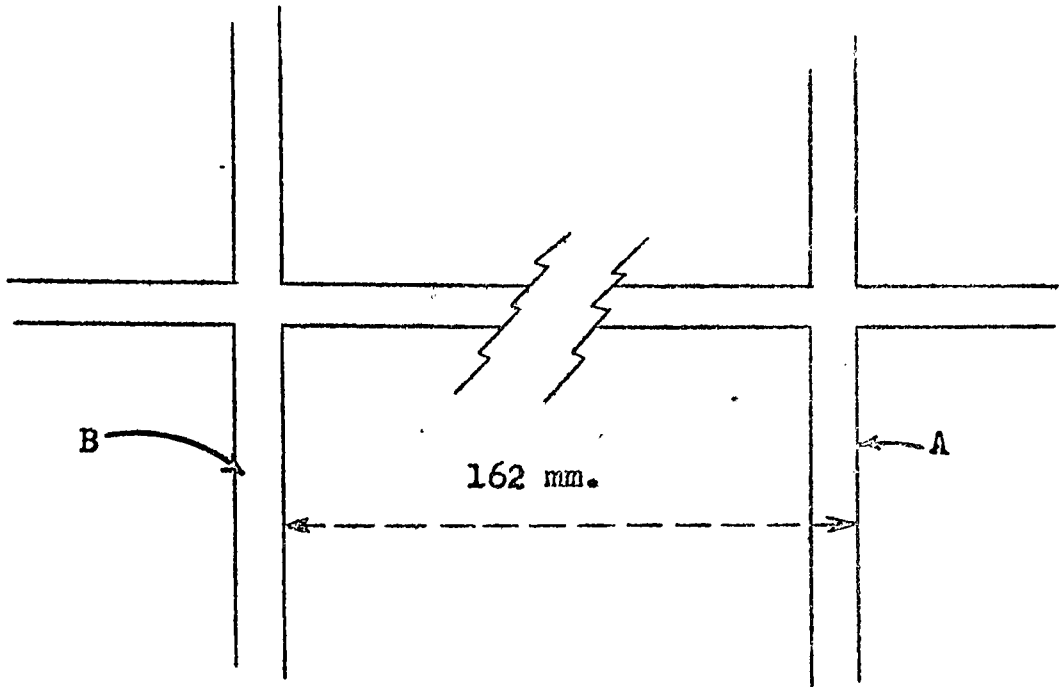




Sample print of
master and step
tablet.

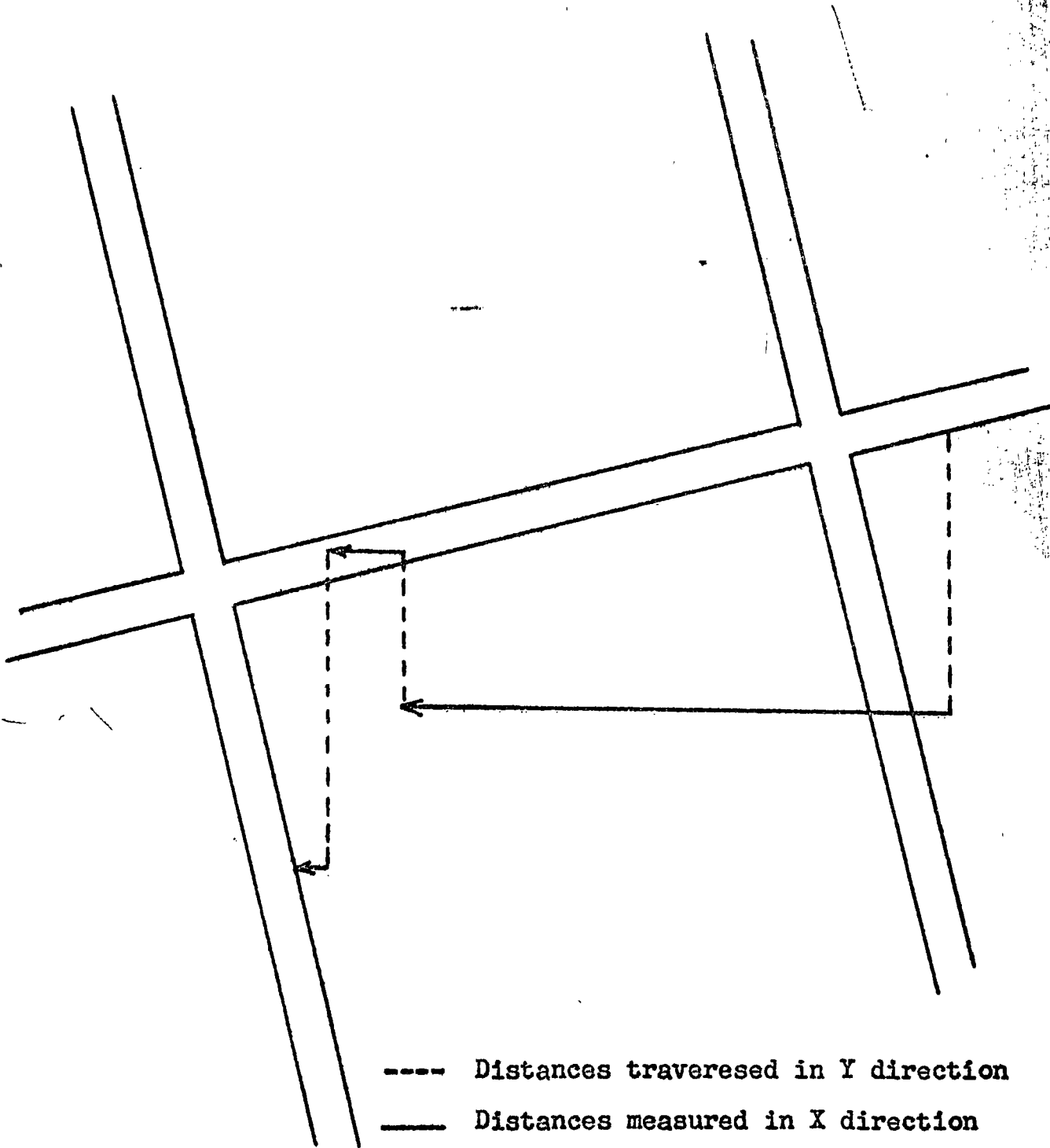
fig. 1

Figure # 2



The long distances on the samples were measured from the right side of line A to the right side of line B. Thus, if the lines expand errors in the long measurements would be at a minimum because the sides would be expanding in the same direction --- at the same time.

Figure # 3



----- Distances traversed in Y direction

———— Distances measured in X direction

Figure # 5&4

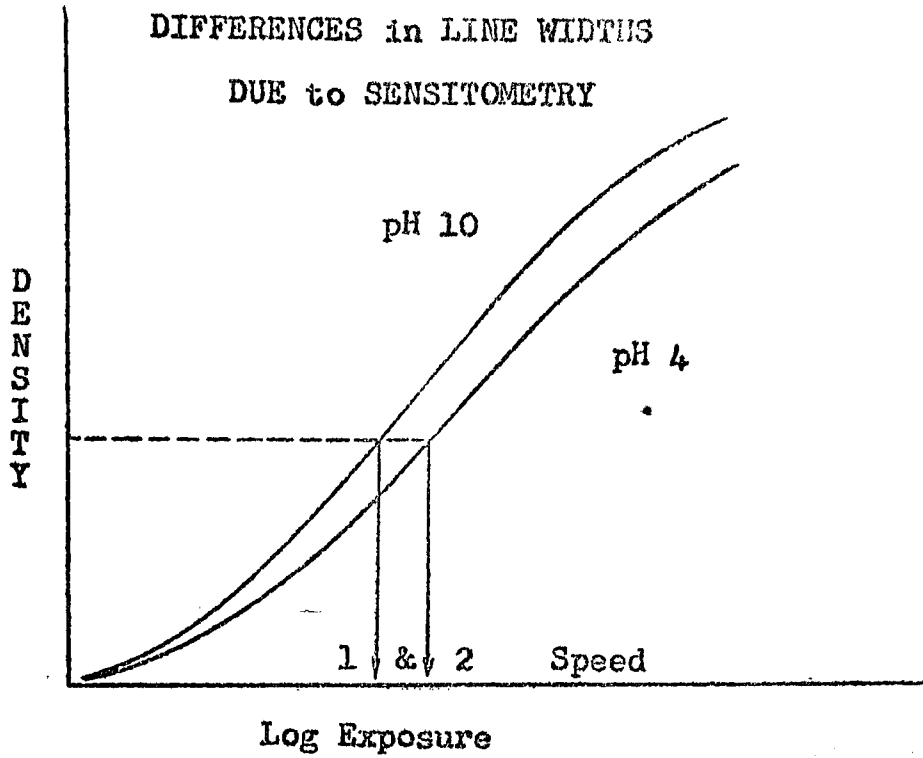
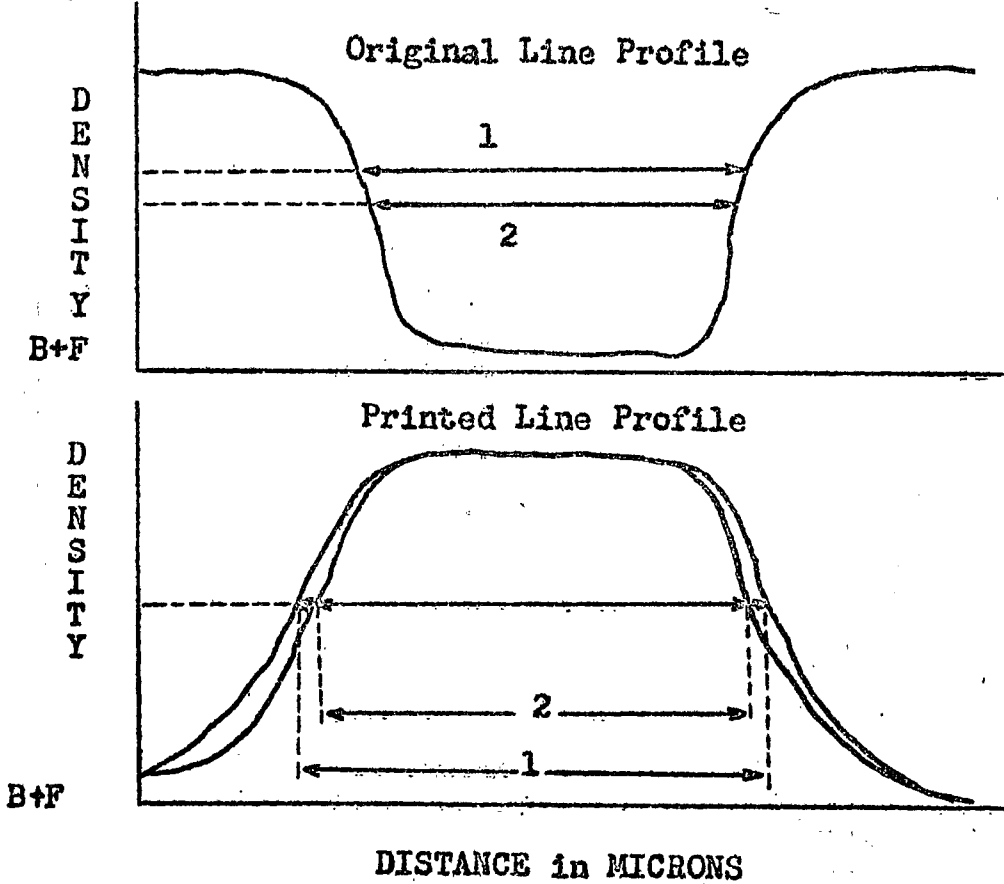


Figure # 6



Sample Calculations of Ionic Strength

Ionic Strength represented by the symbol I follows the equation:

$$I = 1/2 \sum M_i Z_i^2$$

where M_i represents the concentration in Molality and Z_i represents the charge on the ion.

For $I = 1.120$

$$\text{Buffer } I = 0.04$$

$$\text{KBr } - \quad I = 1/M_w / 23.5\text{g/l} = 23.5/114.02 = 0.196 \text{ M}$$

$$\text{KBr } I \quad 0.196$$

$$\text{Desired } I \quad 1.120$$

$$\text{Buffer } \underline{0.040}$$

$$- \underline{0.236}$$

$$\text{Required } I = 0.884$$

$$\text{Std } I = 0.236$$

$$\begin{aligned} 0.884 &= 1/2((2 \cdot X \cdot 1^2) + (1 \cdot X \cdot 2^2)) \\ &= 1/2(6X) \\ &= 3X \end{aligned}$$

$$X = 0.295 \text{ M} \cdot 142.05 = 42 \text{ grams } \text{Na}_2\text{SO}_4$$

Mw of
Na₂SO₄

Hence the addition of 42 grams of sodium sulfate to the standard amount of bromide and buffer should give the desired ionic strength.

Sample Calculation of Ionization of EDTA Ion

pH	Degree pK	The approximate degrees of ionization of the EDTA ion are given on the left.
4.0	2.5	
7.0	3.0	
8.68	3.5	
10.0	4.0	

$$\text{FeSO}_4 \quad 20 \text{ g/l} / 152 \text{ g/mole} = 0.131 \text{ Molar}$$

$$\text{Na}_2\text{EDTA} \quad 60 \text{ g/l} / 380.2 \text{ g/mole} = 0.157 \text{ Molar}$$

$$\begin{aligned} I_{\text{pH } 4} &= 1/2((2.5 \cdot 0.157 \cdot 1^2) + (1 \cdot 0.157 \cdot 2.5^2) \\ &\quad + (1.0 \cdot 0.131 \cdot 2^2) + (1.0 \cdot 0.131 \cdot 2^2)) \\ &= 1/2(\underset{\text{Na}^+}{0.39} \quad \underset{\text{EDTA}^{-2.5}}{0.98} \quad \underset{\text{Fe}^{++}}{0.524} \quad \underset{\text{SO}_4^-}{0.524}) \\ &= 2.418 / 2.0 = 1.209 \end{aligned}$$

40 ml. of $\text{H}^+\text{C}_2\text{H}_3\text{O}_2$ required to obtain pH of 4.0 $I = 0.680$

$$1.209 + 0.680 = 1.889$$

$$\begin{array}{r} \text{Desired } I = 2.257 \\ - 1.889 \\ \hline 0.368 \end{array}$$

$$0.368 / 3.0^* = 0.123$$

$$0.123 \times 148 \text{ g/mole} = 27.5 \text{ grams of Na}_2\text{SO}_4 \text{ to be added}$$

* See sample calculations of ionic strength for details.

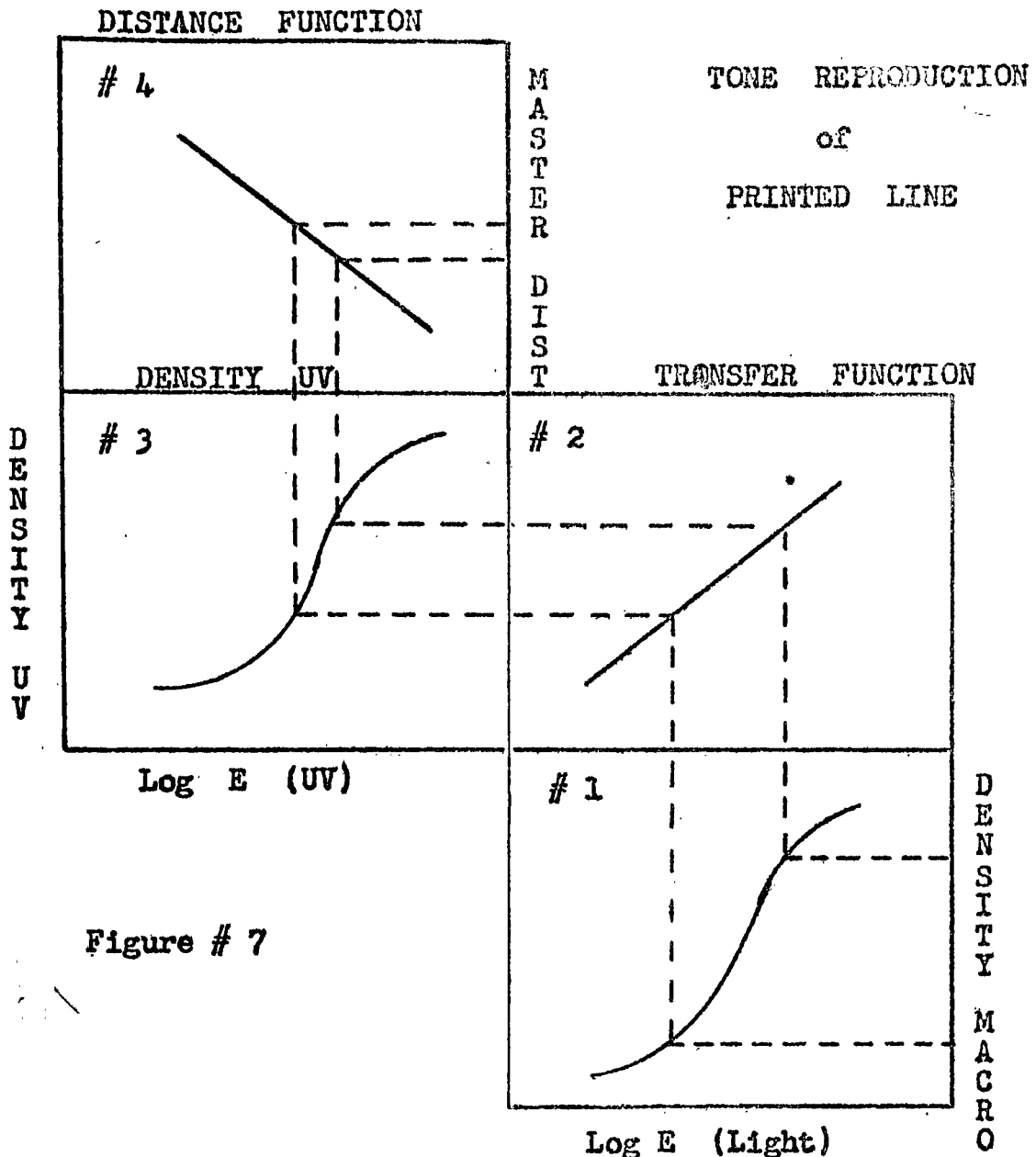
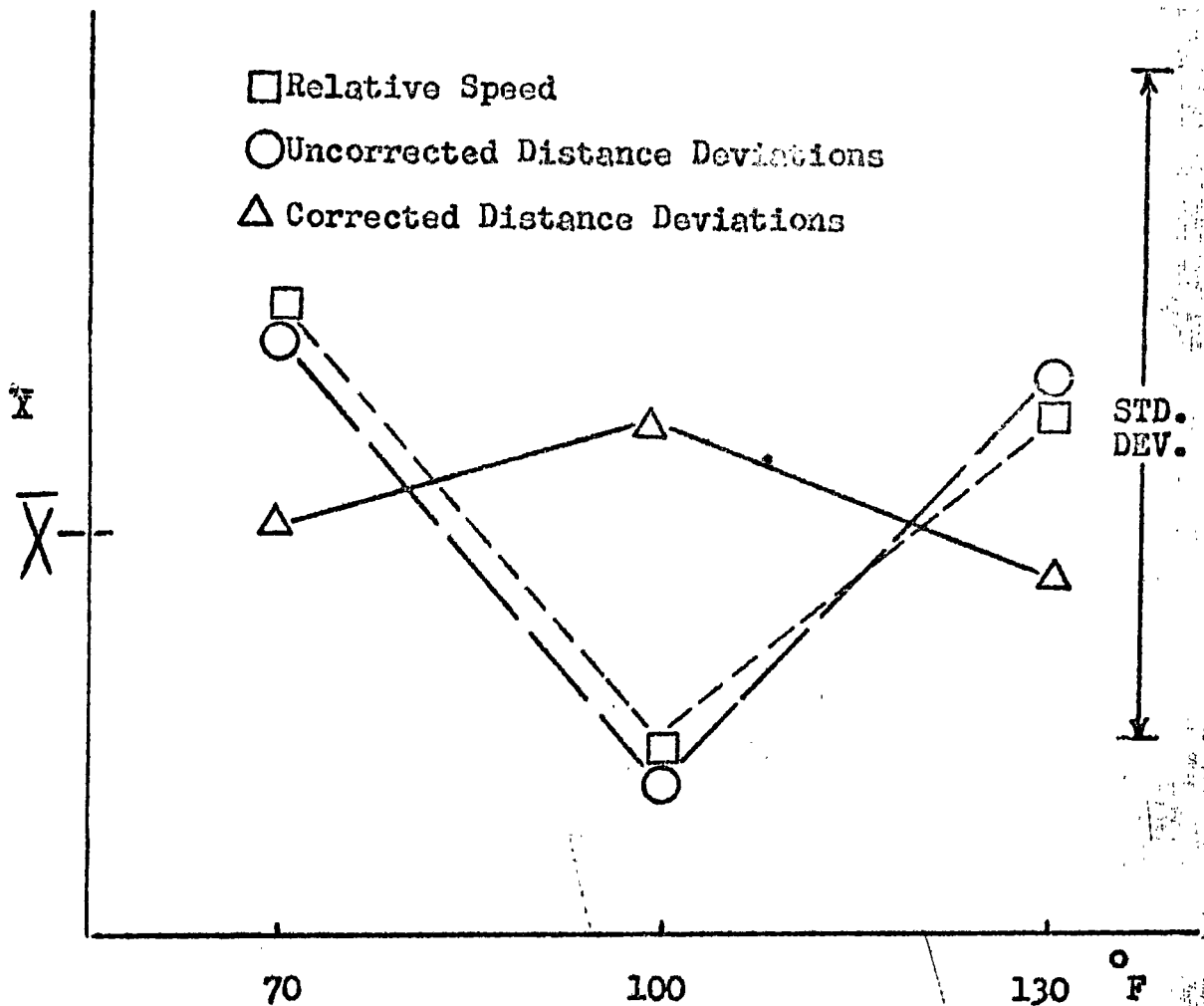


Figure # 7

Since the samples were exposed with ultra violet radiation and the step tablet was exposed by light a relation between these two methods had to be determined. The transfer function, in quadrant # 2 was used to determine effective log UV that the samples recieved, using the whitel light control strip from each sample. From this effective exposure differences, the different distances on the master were calculated for each sample.

Figure # 8



In order to have a direct comparison between response variables of different and indirectly related units the above graph was prepared. Each of the response variables, speed, and dimensional differences, are plotted with their averages normalized to \bar{X} . The scales of each of these variables are plotted with the standard deviations for each at unit length. Therefore any changes in the value of unrelated variables can be compared through changes of eachs relative change from the one standard deviation.

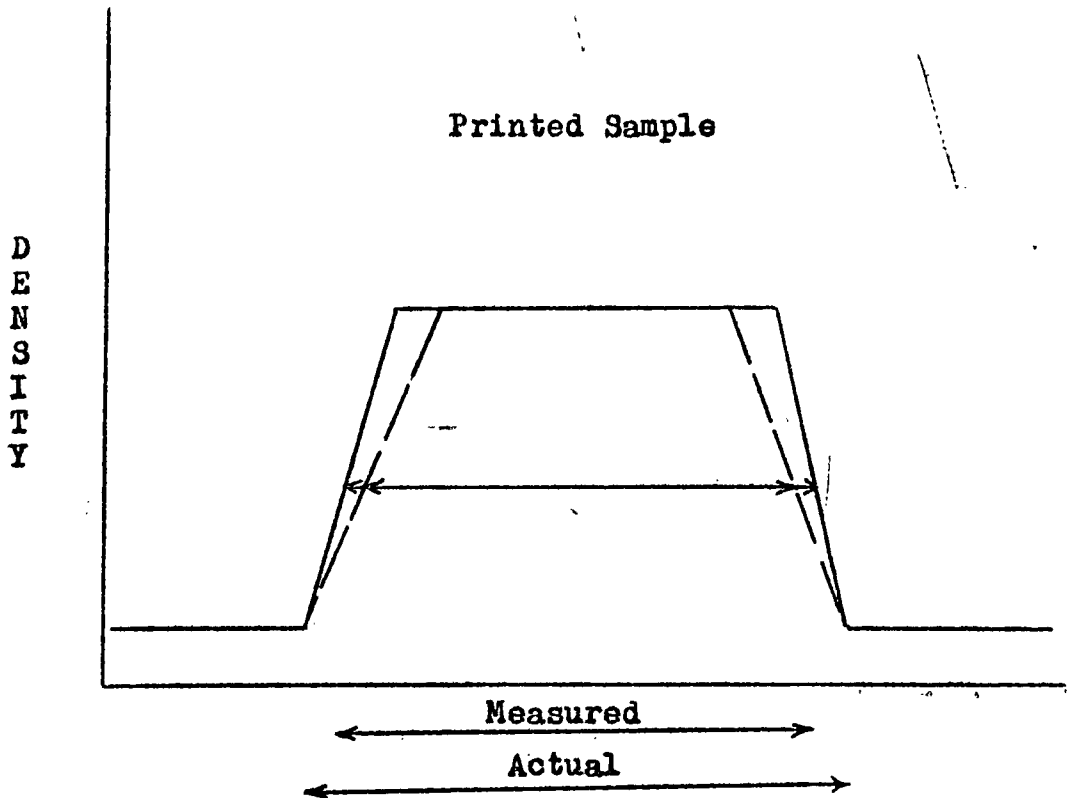
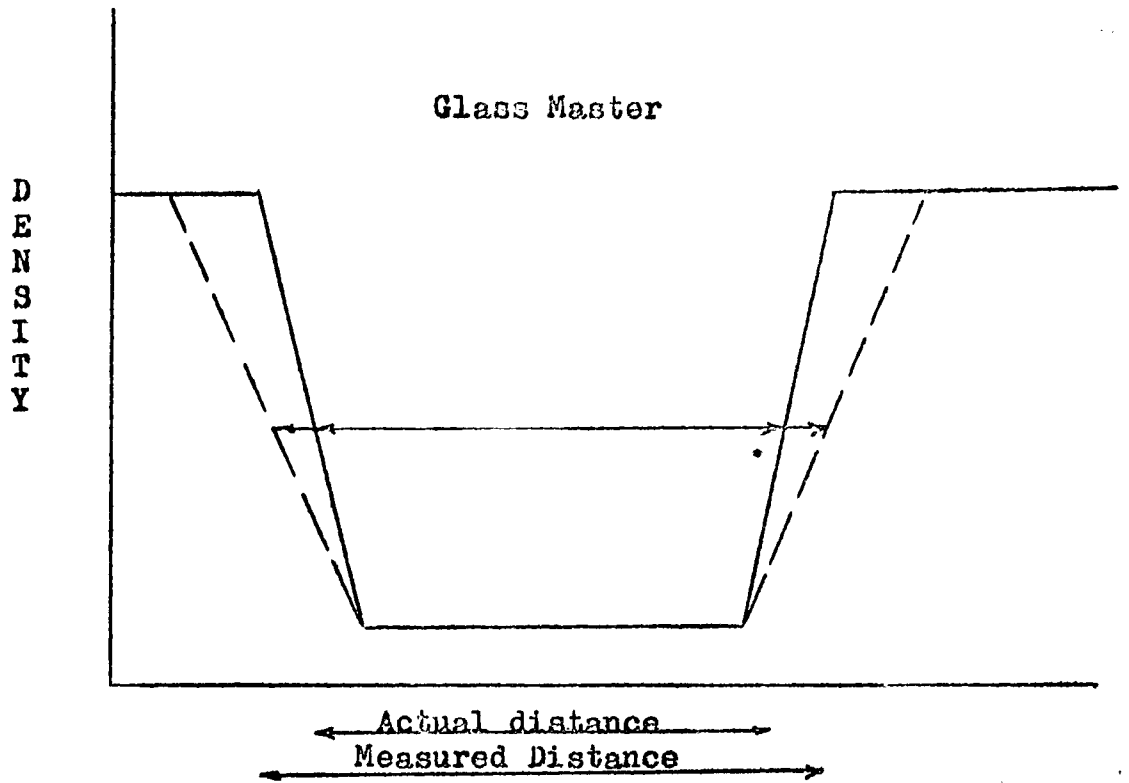


fig. 9

MANN-DATA MICRO-ANALYZER

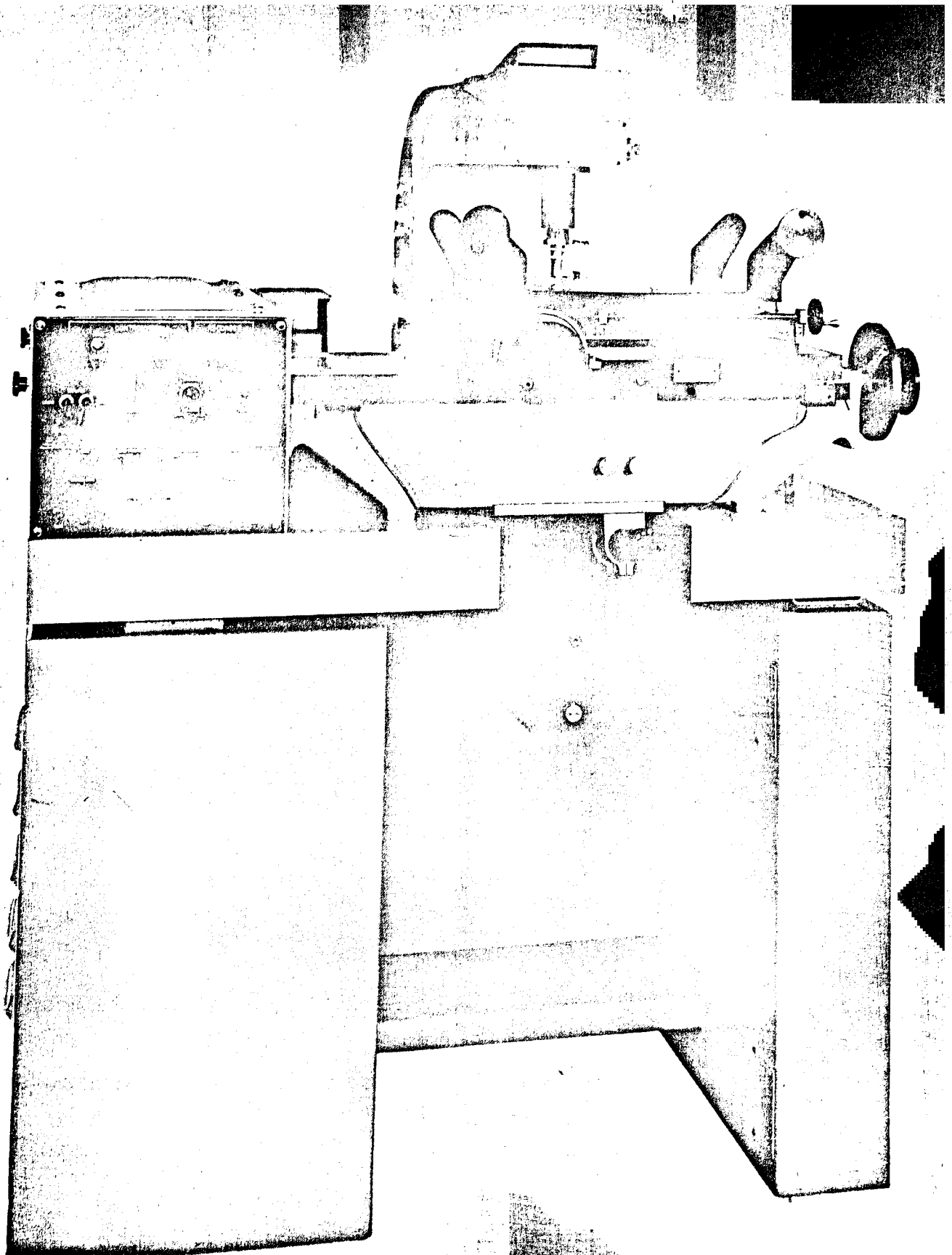


FIGURE 90

Alis

10/10/1998 Pittman

"A pH system -- " - poor wording

I still don't like the wording of "displacements over both very and short distances";

rich does not tell what it is about

A3 ^{Kudak} master was a High Resolution white (spec plates include many emulsions)

A5 ~~What was the pH?~~ Where were the pH values measured? At exit of dryer?

It would probably be better to take pH and Temp, at intake air, calculate pH at dryer temperature, which gives the state of the air reaching the films; the exit data indicate how much was taken up.

p14 The largest negative number in the "set" because it is the smallest ???

How can you say which? There probably was some spread of ~~image~~ the photographic image over the mask falling on the film, so that ~~an~~ final difference was result of two opposing factors