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LABORATORY NOTEBOOK

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If after further thought and discussion, or a few experiments, you decide that the idea may have value, you may draw up a formal description, either as a memo or on the invention record form provided, for submission to the Patent Division.

It is a function of the Patent Division to review the yellow sheets in this notebook as forwarded by you and to cull from them all ideas having patentable novelty, but the invention record form submitted in this way will serve to accent those ideas considered by you to be most important.

PROCEDURE FOR
KEEPING LABORATORY NOTEBOOKS

1. Make all entries in ink and as legible and complete as possible.

2. Every pair of sheets is consecutively numbered, and all entries should be made on the white sheets with a carbon paper between the white sheet and the next succeeding yellow sheet. The yellow sheets, which are perforated for easy removal, are torn out daily and forwarded to the administrative office of the Research Corporation. No white pages (containing the original of the entry) should be torn out under any circumstances.

3. All experimental data is to be entered in this book, and as far as possible, all calculations, graphs, drawings and notes of any character should be entered herein. When patents are referred to, include patent number. When literature is referred to, include complete journal references (Author, Volume, Page, Date).

4. Make all entries in this notebook **first.** It is extremely important that this notebook be what the law calls a "notebook of original entry." Make your first notes about anything—weight, temperature, time, ideas for future work, drawings, sketches—**in this notebook.** If you want to keep a separate tabulation in special cases, do your transcribing of data from the notebook into the tabulation, but not the other way.

5. When starting a problem use the following heading:

Subject:_____Project No._____

Experiment No._____Test No._____

If work done on the problem for the day fills more than one page, continue on next page and start heading:

Project No._____Continued

If work done on the problem for the day fills part of page, use the remainder of page for the next problem worked on that day and fill in the complete heading. Make all your entries in strict chronological order. If you are working on several experiments at once and making alternate observations, use the full heading at the start of each experiment each day, and for later entries on that day you need only note the experiment number beside the data.
Date and sign every page or after each problem on the same page.
The full date should be entered, e.g., July 1, 1946, not 7/1/46.

Do not leave blank spaces after experiments. If there is only a small amount of space, 1 to 2 inches, left at the end of the page when an experiment is finished, draw diagonal lines through this space.
Have an associate, if possible a superior, witness the record of the entire experiment and the signature of each experiment. He should be someone qualified to understand the record.

6. Erasures should never be made. Draw a line through the incorrect work or entry. Never make corrections on a page after signing and dating. If it is at any time observed that a cancellation or addition should have appeared in an earlier entry, a new entry should be made, stating what should be cancelled and what addition should have appeared in the earlier entry.

7. When an experiment must be started on one day and completed some days or a week later, space should not be left at the first entry for subsequent results, but when the results are finally received, the data from the original day's work may (but need not) be brought forward (with suitable reference to the page where they first occur) and the whole experiment summarized on the day on which the final data are obtained.

8. **Do your thinking in the notebook.** Whenever you have an idea of any kind on future experimental work, whether it bears on your own problems or those of other laboratory or research sections or of others with whom you may be engaged during the course of your work, jot it down in your notebook at once, with as much detail as possible. Do the same for proposed apparatus layouts and experimental procedures.
After any discussion, formal or informal, that bears on the work of the Research Corporation or other organizations engaging the services of Research Corporation, note what it was about and who was present. If you watch someone else carry out an experiment, make a note of what you saw and who was there.
In short, make this notebook a diary so that anyone can go back to it years later (five to ten years is not unusual in patent actions) and report without question what you did, what you saw, what you thought, who was present, whom you spoke to, what you discussed and **when all this occurred.**

9. Index experiments on index pages in front of book.

10. When describing experimental work in weekly or monthly reports, always include the notebook and page numbers on which the work can be found.

11. When a book is filled, it is checked and approved by your section or division head and returned to the Patent Division for filing, unless it is to be retained for reference. However, only one finished book may be retained, and all other finished books must be returned to the Patent Division unless specific permission for their retention is obtained from the Director of Research. If other notebooks are later needed for reference, they can be charged out.

INDEX

ANALYTICAL INSTRUMENTS USED

①	Top Loading Balance	- SRB/00004
②	Top Loading Balance	- SRB/00011
③	Spectrodenistomer	- 938
④	DU-7 - Spectrophotometer	- SRB/00007
⑤	Rotary Evaporation RE/51	- SRB/00009
⑥	1310 - Infrared Spectrophotometer	- SRB/00014
⑦	Mettler AE - 160 - Balance	- SRB/00006
⑧	Mettler AE - 200 - Balance	- SRB/00012
⑨	pH Meter	- SRB/00145 or - SRB/00019
⑩	Temperature and Humidifer	- 00159
⑪	% Water	- 00515
⑫	Oven	- SRB/00003
⑬	Brookfield Viscometer Model LVTDV-II	- 00099
⑭	Brookfield Viscometer Model LVTDV-II	- 00035 (CYPRESS)
⑮	Atomic Absorption	- 00001 (CYPRESS)
⑯	Top Loading Balance	- 00516

unless otherwise indicated

* Unless otherwise specified, Mettler AE-200-Balance (Analytical # SRB/00012) is used on all analytical and DV weights

INDEX

DV METHODS USED

DV-478	Color Value
DV-1	Pan Solids
DV-288	Color Value
DV-516	Color Value
DV-407	Color Value
* DV-12	Acid #
DV-5	pH
DV-300	Microwave
DV-13	% H_2O
DV-162	IR
DV-26	Viscosity
DV-316	ppm/Nat (Atomic Absorption)
DV-400	IR/Hydrolysis Completion
DV-86	Flash Point
DV-4	Shear Stability
* DV-169	Hydroxyl Number (1 hr. cook time)
* DV-9	Hydroxyl Number (5 min. cook time)

* On specified DV Methods, Analytical Balance #SRB/00006

- Data plate #520 is used to monitor temperature unless otherwise specified; it is routinely checked with a calibrated thermometer to assure accuracy

INDEX

PREPARATION OF EXPERIMENTAL MAGENTA	(Amy's exp. coupler, phthalic anhydride)	10022-01
pH MEASUREMENT OF OVEN STABILITY 1892s	(pH of Lot #'s 10055-64 and 10055-70)	10022-02
PREPARATION OF EXPERIMENTAL INK BLUE 1892	(capped with DDSA)	10022-04
PREPARATION OF EXPERIMENTAL INK BLUE 1892	(3 rxns. at 1M, 2M, 3M of DDSA)	10022-06
SAMPLE REQUEST #10-1042 PREPARATION		10022-08
DRAWDOWN PREPARATION OF EXP. IB 1892 WITH DDSA	(NH_4OH H_2O-bleed test on all 3 #10022-06)	10022-09
OVEN STABILITY TESTING ON EXP. IB 1892's	(DDSA: 3 wks; DDSA-B: 2 wks)	10022-10
PREPARATION OF EXPERIMENTAL MAGENTA COUPLER	(MAP, Glydexx, dry ice)	10022-11
PREPARATION OF EXPERIMENTAL INK BLUE 92 COUPLER	(aniline oil, Glydexx, CO_2)	10022-12
PREPARATION OF EXPERIMENTAL MAGENTA	(Coupler, PA, 1-MI, ODCB; CV = 15.9)	10022-13
PREPARATION OF EXPERIMENTAL INK BLUE 92	(Coupler, OFBSA, H_2SO_4, DCM + benz. per.)	10022-14
PREPARATION OF LIQUITONE BLUE 452 W	(color value = 42; decreased from 48)	10022-16
PREPARATION OF EXPERIMENTAL INK BLUE	(H_2O-based IB 452; aniline 2,6, muriatic; CV = 7)	10022-17
PREPARATION OF EXPERIMENTAL INK BLUE	(H_2O-based IB 452; aniline 2,6, H_2SO_4, benzoquinone CV = 42)	10022-18
PREPARATION OF EXPERIMENTAL INK BLUE 1892	(4M of DDSA reacted - same as #04)	10022-19
PREPARATION OF EXPERIMENTAL INK BLUE	(H_2O-based IB 452; aniline 2,6, H_2SO_4; CV = 11)	10022-22
PREPARATION OF EXPERIMENTAL INK BLUE	(H_2O-based IB 452; CV = 34.2)	10022-24
PREPARATION OF EXPERIMENTAL INK BLUE 1892	(capped with DDSA (4M) - #04 x 4)	10022-26
PREPARATION OF EXPERIMENTAL INK BLUE	(H_2O-based 452; PDEA, muriatic; CV = 0.8825)	10022-27
OVEN STABILITY TESTING ON EXP. IB 1892's	(DDSA: 5 wks; DDSA-B: 3 wks)	10022-28

INDEX

PREPARATION OF SAMPLE #10-1049 (Eagle Int. : Liq. Red 897W + Liq. Yellow 800W)	10022-29
PREPARATION OF EXPERIMENTAL INK RED (Reactint Red X64 + ODSA: sent to Independent Ink)	10022-30
PREPARATION OF EXPERIMENTAL INK BLUE (Uncut Bullseye Violet, Glydexx, KOH; soluble in H_2O)	10022-31
PREPARATION OF EXPERIMENTAL INK BLUE (H_2O-based IB 452 : Glydexx, KOH : CV = 5)	10022-32
GPC TESTING OF EXPERIMENTAL INK BLUE 1892 (made-up sample, sent to Eric Stephens)	10022-32
PREPARATION OF EXPERIMENTAL INK BLUE 18452 (capped with DDSA (4M))	10022-33
TEST OF UNCUT INK BLUE 452	10022-33
PREPARATION OF EXPERIMENTAL INK BLUE (H_2O-based IB452 : ASA - soluble in H_2O)	10022-34
PREPARATION OF EXPERIMENTAL INK BLUE (H_2O-based IB 452: powder intermediate - soluble in H_2O)	10022-35
PREPARATION OF EXPERIMENTAL INK BLUE (H_2O-based IB 452 : XR-4913 - soluble in H_2O)	10022-37
PREPARATION OF ETHANOLAMINE CUT 1892 (Sample to 3 different customers)	10022-38
ANALYSIS OF INK RED 97 FOR CUSTOMER (TEST NIPFORM T1 SPECS)	10022-39
PREPARATION OF EXPERIMENTAL INK BLUE 452 (aniline 5: low CV, CV = 6.6)	10022-40
PREPARATION OF SAMPLE # 10-1056 (Sun Chemical : IR 97 + IB 452)	10022-42
** PREPARATION OF EXPERIMENTAL INK BLUE 452 (aniline 5: high CV, CV = 55; ODSA-capped; H_2O INSOLUBLE)	10022-43
PREPARATION OF EXPERIMENTAL INK BLUE 452 (aniline 5: low CV; NO AMV)	10022-46
PREPARATION OF EXPERIMENTAL INK BLUE 92 (aniline 5: low CV; NO AMV)	10022-47
PREPARATION OF EXPERIMENTAL INK BLUE 452 (aniline 5: low CV, CV = 19.8; oxidation incomplete)	10022-50
RESIMENE REACTIONS (resin reactions with Uncut IB 92 and IY 850)	10022-51
RESIMENE REACTIONS WITH INK YELLOW 850	10022-52

INDEX

SEPARATION TESTING ON INK YELLOW 850 (aniline 5, Silwet L-722) 10022-53

SEPARATION TESTING ON INK YELLOW 850 (aniline 2106, aniline 213, m-tol 2148DA) 10022-54

SEPARATION TESTING ON INK YELLOW 850 (Syn Fac: 334-10, 8337, 8210, 8216) 10022-55

COLOR VALUE OF EXPERIMENTAL INK BLUE 452 (Lot # 10022-43) 10022-56

PREPARATION OF EXPERIMENTAL INK BLUE 92 (H_2O-based: aniline 5, OFBSA, dimethylaniline) 10022-57

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based : low c.v. - prob. w/oxidiation) 10022-59

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based : ↑ HCl to 1.5M; low c.v.) 10022-61

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: ↑ AMV to 0.9%; low c.v.) 10022-62

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based : procedure same as #43; low c.v.) 10022-64

SEPARATION TESTING ON INK YELLOW 850 (dodecyl sulfate; sodium stearate; SynFac: 8145, 8118) 10022-66

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: ↑ p-DMAB to 0.228 M; low c.v.) 10022-67

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: ↑ aniline 5 to 0.44 M; low c.v.) 10022-68

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: ↑ H_2O_2 to 0.61 M; low c.v.) 10022-70

★★ PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: no initial cond. H_2O; high c.v. = 50) 10022-71

PREPARATION OF EXPERIMENTAL INK BLUE 452 (H_2O-based: SAME AS #71, except ↑ cond. time (6 hrs) 10022-73

PREPARATION OF EXPERIMENTAL INK BLUE 1892 (H_2O-based : cut with ethanolamine) 10022-74

PREPARATION OF EXPERIMENTAL MAGENTA

① EQUIPMENT: 1-500 mL roundbottom, three-neck flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #'s
experimental coupler	30.1 g	9981-68
phthalic anhydride, 99%	18.5 g	09809HN
1-methylimidazole	0.5 g	K9037
1,2-dichlorobenzene	30.0 g	06801MZ

③ PROCEDURE:
1 Charge coupler to the flask and begin stirring
2 Charge phthalic anhydride
3 Charge 1-methylimidazole
4 Charge 1,2-dichlorobenzene and equip flask with a condenser
11:40am 5 Begin heating flask to 110°C
6 Continue condensing reaction at 110°C for 3 hours
2:40pm 7 Run IR Test

TEST DATA AND OBSERVATIONS:
7A) A peak appears at 1830 cm⁻¹, indicating that the phthalic anhydride has not completely reacted with the experimental coupler.
8 Continue condensing at 110°C for 1.25 hours
3:55pm 9 Run IR Test

TEST DATA AND OBSERVATIONS:
9A) A peak appears at 1830 cm⁻¹, indicating that the phthalic anhydride has not completely reacted with the experimental coupler.
10 Continue condensing at 110°C ~~for~~ gd 5/6/96
6:00pm 11 Stop overnight

OBSERVATIONS:
Became very thin when ODCB was added
Actual Temp = 119°C
Turned "magenta color" almost a cranberry color 1 hr into experiment

As time goes by, color becomes a deeper, darker maroon - less yellow and bright indicates reactivity

Date May 6, 1996 Signature Gloria G. Wickens
Date May 14, 1996 Witness [illegible]

((CONTINUE TO PAGE 10022-03))

pH MEASUREMENT ON OVEN STABILITY 1892s

① EQUIPMENT AND SOLVENTS: pH meter (Omega) #SRB/00019
methanol
distilled H_2O } FOR CLEANING PURPOSES

② pH MEASUREMENT AND PROCEDURE:

1 – Remove all 6 oven stability testing vials and let cool to room temperature
2 – Measure and record pH of each; ph, DV-S

DATA: pH, DV-S: pH:

① Lot# 10055-64 : DDSA w/ NH_4OH : 9.69

② Lot# 10055-64 : DDSA w/ ethanolamine : 7.83

③ Lot# 10055-64 : DDSA w/ diethanolamine : 7.88

④ Lot# 10055-70 : DDSA-B w/ NH_4OH : 9.34

⑤ Lot# 10055-70 : DDSA-B w/ ethanolamine : 8.50

⑥ Lot# 10055-70 : DDSA-B w/ diethanolamine : 9.45

③ ~~CONCLUSIONS AND NOTES~~: gd 5/9/96

Date May 6, 1996 Signature Gloria G. Wickem
Date May 14, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-01

((PREPARATION OF EXPERIMENTAL MAGENTA))

8:30am [12] Continue condensing at 110°C for 2 hours
10:30am [13] Run IR Test

TEST DATA AND OBSERVATIONS:

13A) A peak still appears at 1830 cm⁻¹. The phthalic anhydride was added in excess so obviously it is not going to react 100% - there will always be a small anhydride peak.

NEUTRALIZATION:

(4) RAW MATERIALS:

	Weight (g)	Lot #
98% Sulfuric acid	2.7g	C4615
93% Sulfuric acid	1.0g + 1.0g	B4206

(5) PROCEDURE: [1] Add/charge 98% H_2SO_4 slowly through the condenser
[2] Raise temperature to 150°C
11:00am [3] Continue heating at 150°C for 1.5 hours
12:30pm [4] Run color value, DV-478

OBSERVATIONS: Turned a darker maroon when H_2SO_4 was added - did not exotherm until temp. was raised, then it exothermed to 161°C

TEST DATA AND OBSERVATIONS:

4A) *Color value, DV-478 ⟹ *USE ACETONE AS SOLVENT

Sample weight (sw) = 0.2322g
absorbancy (A) = 0.241
wavelength (λ) = 543
color value = (A/sw) × 5
*Color value = (0.241/0.2322) × 5 = 5.1895 = 5.19

4B) extremely low - it is expected theoretically to be appx. 37

1:00pm [5] Charge 93% H_2SO_4 extremely slowly through the condenser
[6] Continue heating at 150°C for 1 hour
2:00pm [7] Run color value, DV-478

H_2SO_4 turned to vapor and came 1" up the condenser (in gas form)

TEST DATA AND OBSERVATIONS:

7A) *color value, DV-478 ⟹ USE METHANOL AS SOLVENT

Sample weight (sw) = 0.2572g
absorbancy (A) = 0.825
wavelength (λ) = 539
Color value = (A/sw) × 5
*Color value = (0.825/0.2572) × 5 = 16.0381 = 16.04

Date May 07, 1996 Signature Clara G. Wickens
Date May 14, 1996 Witness [illegible]

((CONTINUE TO PAGE 10022-05))

PREPARATION OF EXPERIMENTAL INK BLUE 1892

① EQUIPMENT: 1- 1000 mL three-neck, roundbottom flask
mechanical agitator
thermal controller
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 92	205.9g	A1017
DDSA	106.4g	TRL-6
1-methylimidazole	3.0g	K9037

③ PROCEDURE:
1 Charge Uncut Ink Blue 92 and begin stirring
2 Charge DDSA
3 Charge 1-methylimidazole and equip flask with a condenser
12:45pm 4 Heat flask to 90-98°C
5 Continue heating flask at 90-98°C for 3 hours
3:45pm 6 Run IR

TEST DATA AND OBSERVATIONS:
6A) No peak appears at 1830 cm^{-1} - this indicates that the DDSA has reacted fully with the OH groups to form the standard ester blue compound. The IR data indicates the reaction is complete.
7 Stop reaction; retain full sample.
⟹ GIVE 8 oz. TO CRAIG COLLINS
GIVE 3 oz. TO KEVIN GILDER
FOR DRAWDOWN/STABILITY TESTING!

Date May 07, 1996 Signature Gloria G. Dickens
Date May 14, 1996 Witness Kevin Gilder

CONTINUED FROM PAGE 10022-03

((PREPARATION OF EXPERIMENTAL MAGENTA))

[8] Charge 1.0g of 93% H_2SO_4 extremely slowly through the condenser
[9] Continue heating at 150°C for 2 hours
[10] Run color value, DV-478

TEST DATA AND OBSERVATIONS

10A) Color value, DV-478
⟹ use methanol as solvent
Sample weight (sw) = 0.2386g
Wavelength (λ) = 539
absorbancy (A) = 0.725
Color value = (A/sw) x 5
Color value = (0.725/0.2386) x 5
Color value = 15.1928 = 15.2

10B) Color value decreased with the last H_2SO_4 addition - obviously it was not necessary and actually hurt the reaction

[11] Stop reaction; retain full sample for further testing.

OBSERVATIONS:
A viscous dark burgandy liquid with apparent anhydrous foam throughout (tiny, foamy bubbles - like ODSA)

Date May 07, 1996
Date May 14, 1996

Signature Gloria G. Dickens
Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 1892

REACTION I: 1M OF DDSA

① EQUIPMENT: 1-500mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #'s
Uncut Ink Blue 92	102.95g	PP001
DDSA	13.3g	TRL-6
1-methylimidazole	1.5g	K9037

③ PROCEDURE: [1] Charge Uncut Ink Blue 92 and begin stirring
[2] Charge DDSA
[3] Charge 1-methylimidazole and equip flask with a condenser
11:00am-2:00pm [4] Heat flask to 90-98°C for 3 hours
[5] Run IR

TEST DATA AND OBSERVATIONS:
5A) No peak appears at 1830cm^{-1}, indicating the reaction is complete - the anhydride has reacted with 1 OH group to form 1 ester structure
[6] Stop reaction; retain full sample in a 4 oz. jar for future testing.

REACTION II: 2M OF DDSA

① EQUIPMENT: 1-500mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #'s
Uncut Ink Blue 92	102.95g	PP001
DDSA	26.6g	TRL-6
1-methylimidazole	1.5g	K9037

((CONTINUE TO PAGE 10022-07))

Date May 8, 1996 Signature Gloria G. Dickens
Date May 14, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-06

((PREPARATION OF EXPERIMENTAL IB1892))

③ PROCEDURE: 1 Charge Uncut Ink Blue 92 and begin stirring
2 Charge DDSA
3 Charge 1-methylimidazole and equip flask with a condenser
11:00am-2:00pm 4 Heat flask to 90-98°C for 3 hours
5 Run IR

TEST DATA AND OBSERVATIONS:
5A) No peak appears at 1830 cm^{-1}, indicating the reaction is complete - the anhydride has reacted with 2 OH groups to form 2 ester structures.
6 Stop reaction; retain full sample for further testing.
⟹ END PRODUCT HAS FOAMY BUBBLES, UNLIKE 1M DDSA (a few slight bubbles) and 3 M DDSA (extremely smooth)

REACTION III : 3M OF DDSA

① EQUIPMENT: 1- 500mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight(g)	Lot #'s
Uncut Ink Blue 92	102.95g	PP001
DDSA	39.9g	TRL-6
1-methylimidazole	1.5g	K9037

③ PROCEDURE: 1 Charge Uncut Ink Blue 92 and begin stirring
2 Charge DDSA
3 Charge 1-methylimidazole and equip flask with a condenser
11:00am-2:00pm 4 Heat flask to 90-98°C for 3 hours
5 Run IR

TEST DATA AND OBSERVATIONS:
5A) No peak appears at 1830 cm^{-1}, indicating the reaction is complete - the anhydride has reacted with 3 OH groups to form 3 ester structures.
6 Stop reaction; retain full sample in 8oz. jar for future testing.

Date May 8, 1996 Signature Gloria G. Wickam
Date May 14, 1996 Witness [illegible]

⟹ CONCLUSIONS ON PAGE 10022-09

SAMPLE REQUEST # 10-1042 PREPARATION

RAW MATERIALS:	Lot #'s
Uncut Ink Blue 1892	RD07
Uncut Ink Red 1897	RD04
Uncut Ink Yellow 1800	RD05
gd 5/8/96 ~~Uncut~~ Ink Blue 18452 : H_2O-cut	D1013
ammonium hydroxide	T2166
distilled water	—
propanediol	04309HN

SAMPLES : MIX WELL (ON MAGNETIC STIRPLATE)

① LiquiTone Blue 892W : 267.86 g Uncut IB 1892 } 90/10 ratio
26.79 g ~~NH_4OH~~ gd 5/8/96 propanediol

color value, DV-478: use methanol as solvent
Sample weight (sw) = 0.3631g
Wavelength (λ) = 435
absorbancy (A) = 1.418
Color value = $(A/sw) \times 5 = (1.418/0.3631) \times 5 =$ 19.53

② LiquiTone Yellow 800 W: Uncut Ink Yellow 1800
⟹ NO BASE ADDED
⟹ JUST CHECKED COLOR VALUE AND WROTE ON 5-gal. PAIL

③ LiquiTone Red 897 W: 200.0g Uncut IR 1897
33.6g distilled H_2O
16.48g NH_4OH

④ LiquiTone Blue 452 W: 200g H_2O-cut ink blue 18452
28.6g distilled H_2O

color value, DV-478: use methanol as solvent
Sample weight (sw) = 0.1148g
Wavelength (λ) = 592
absorbancy (A) = 0.976
Color value = $(A/sw) \times 5 = (0.976/0.1148) \times 5 =$ 42.51

PAIL C.V. :
SW = 0.1037g
λ = 591
A = 0.998
CV = $(.998/.1037) \times 5$
CV = 48.196
⟹ WRITE ON PAIL

SEND ALL 4 SAMPLES TO:
Mr. Walter Burkert
Sun Chemical
1701 Westinghouse Blvd.
Charlotte, N.C. 28273

SHIP DATE: 05/08/96
AIRBORNE, AB# 5989276683

Date May 8, 1996 Signature Gloria G. Wicker
Date May 14, 1996 Witness [illegible]

DRAWDOWN PREPARATION OF EXP. IB 1892 WITH DDSA

① RAW MATERIALS:

	Lot #'s
Exp. IB 1892 w/ 1M DDSA	10022-06 : Rxn. I
Exp. IB 1892 w/ 2M DDSA	10022-06 : Rxn. II
Exp. IB 1892 w/ 3M DDSA	10022-06 : Rxn. III
ammonium hydroxide (NH_4OH)	T2166
distilled water (H_2O)	

② DRAWDOWN PREPARATION AND WATER-BLEED RESULTS:

⟹ prepare inks : add H_2O, base : MIX WELL, then add color

⟹ dry drawdown with hot air blower directly after preparation, drop with water and let it run, then immediately dry it again with hot air blower - check for bleed

A	B	C
1M color : 4g	2M color : 4g	3M color : 4g
H_2O : 15g	H_2O : 15g	H_2O : 15g
NH_4OH : 0.13g	NH_4OH : 0.23g	NH_4OH : 0.30g
(C) ⟹ good appearance and lay down; good color strength; PARTIALLY BLEEDS in H_2O	⟹ good appearance and lay down; good color strength; BLEEDS in H_2O	(A) ⟹ fair appearance and lay down; good color strength; BLEEDS VERY BAD in H_2O

SWAP OBSERVATIONS!

③ CONCLUSION:

Obviously all 4 OH groups in Uncut Ink Blue 92 need to be capped with an anhydride in order to become a water-bleed resistant 1892. Maybe 1892 is bleed-resistant at a molar ratio of DDSA between 3 and 4 - it is not known with this experiment. As expected a 1 molar ratio ~~was~~ is less bleed-resistant than a 4 and 3 molar ratio.

DDSA

gd 5/9/96

Date May 9, 1996 Signature Gloria G. Dickens

Date May 14, 1996 Witness Steve Gilder

OVEN STABILITY TESTING ON EXP. IB 1892's

⟹ PREPARE A DRAWDOWN AND RECORD RESULTS ON EACH OF THE OVEN-TESTED INKS : CHECK FOR WATER-BLEED AND COLOR STABILITY

⟹ OVEN-AGED @ 135°F SINCE : Lot# 10055-64 : 04/19/96
Lot# 10055-70 : 04/29/96

(A) Lot # 10055-64: DDSA with NH_4OH :
- extremely blueish-red color - turned a redder blue
- no water-bleed

(B) Lot # 10055-64: DDSA with ethanolamine:
- turned a slightly reddish blue
- partial water-bleed

(C) Lot # 10055-64: DDSA with diethanolamine:
- nice lay-down and color
- water-bleed

(D) Lot # 10055-70: DDSA-B with NH_4OH:
- extremely blueish-red color - turned a redder blue
- no water-bleed

(E) Lot # 10055-70: DDSA-B with ethanolamine:
- a slight reddish-blue
- water-bleed

(F) Lot # 10055-70 : DDSA-B with diethanolamine:
- nice color strength and laydown
- extreme water-bleed

⟹ RETURN INKS TO OVEN @ 135°F

→ DDSA: 3 WEEKS
DDSA-B: 2 WEEKS

Date May 9, 1996 Signature Gloria G. Wickum
Date May 14, 1996 Witness Steve Gilch

PREPARATION OF EXP. MAGENTA COUPLER

① RAW MATERIALS: 1-500mL three-neck, round bottom flask
thermal controller
mechanical agitator
condenser
vs.
250 ml addition funnel

② EQUIPMENT:

	Weight (g)	Lot #'s
meta-aminophenol (MAP)	58.6g	Y1028
water	13.6g	—
dry ice	6.5g	—
Glydexx ND-101	133g	26761-45-5

③ PROCEDURE: 1 Charge MAP to the flask and begin stirring
2 Charge H_2O
3 Charge dry ice to the flask and equip with a condenser
4 Heat flask to 100-110°C and maintain temperature
9:20am 5 Add Glydexx slowly through an addition funnel, maintaining temperature at 100-110°C
9:45am 6 After addition is complete, equip flask with a condenser and continue heating at 100-125°C
3:45pm 7 Heat at 100-125°C for 6 hours
8 Stop reaction; run % H_2O, DV-13 and if necessary, strip on rotovap

TEST DATA AND OBSERVATIONS:

8A) Sample weight = 0.1105g
% H_2O = 2.842% ⟸ MUCH TOO HIGH TO RUN HYDROXYL NUMBER,

8B) Rotary Evaporator @ 99°C/180RPM STRIP OFF EXCESS H_2O
for 1.5 hours ⟸ AFTER ROTOVAP, RUN % H_2O AGAIN

8C) sample weight = 0.1077g
% H_2O = 0.121% ⟸ STILL THEORETICALLY TOO HIGH TO RUN OH # (must be <0.1%) BUT THAT IS THE MOST H_2O-STRIPPED TO MAX OF H_2O

9 Retain full sample in an 8 oz. jar for future reactions and testing

Date May 10, 1996 Signature Gloria G. Dickens
Date May 14, 1996 Witness [illegible]

PREPARATION OF EXP. INK BLUE 92 COUPLER

① EQUIPMENT: 1-500mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser
1-250mL addition funnel

② RAW MATERIALS:

	Weight (g)	Lot #'s
aniline oil	50.0g	Y8178
water	13.6g	—
dry ice	6.5g	—
Glydexx ND-101	133g	26761-45-5

③ PROCEDURE: [1] Charge aniline oil to the flask and begin stirring
[2] Charge H_2O
[3] Charge dry ice to the flask and equip with a condenser
[4] Heat flask to 100-110°C and maintain temperature
10:40am [5] Add Glydexx slowly through an addition funnel, maintaining temperature at 100-110°C
11:00am [6] After addition is complete, equip flask with a condenser and increase temperature to 100-125°C
[7] Heat at 100-125°C for 6 hours
5:00pm [8] Stop reaction; run % H_2O, DV-13 and if necessary, strip H_2O on rotovap

TEST DATA AND OBSERVATIONS:

8A) Sample weight = 0.1007g
% H_2O = 2.013 %

⇐ % H_2O MUCH TOO HIGH FOR OH # ANALYSIS AND FUTURE RXNS. - STRIP OFF AS MUCH H_2O AS PHYSICALLY AND TECHNICALLY POSSIBLE

8B) Rotary Evaporator @ 99°C / 180 RPM for 1.5 hours

8C) Sample weight = ~~0.97~~ gd 5/10/96 0.0970g
% H_2O = 0.061 %

⇐ % H_2O LOW ENOUGH TO RUN OH # ANALYSIS AND FUTURE REACTIONS!

[9] Retain full sample in an 8oz. jar for future reactions and testing

Date May 10, 1996 Signature Gloria G. Dickens
Date May 14, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL MAGENTA

① EQUIPMENT: 1-500 mL roundbottom, three-neck flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #'s
experimental coupler	17.5g	10022-11
phthalic anhydride, 99%	18.5g	09809HN
1-methylimidazole	0.5g	K9037
1,2-dichlorobenzene (ODCB)	30.0g	06801M2

③ PROCEDURE:
1 Charge coupler to the flask and begin stirring
2 Charge phthalic anhydride
3 Charge 1-methylimidazole
4 Charge ODCB and equip flask with a condenser
12:45 pm 5 Heat flask to 110°C
6 Continue heating flask @ 110°C for 3 hours
3:45 pm 7 Run IR Test

OBSERVATIONS: Reaction was at first a bright yellow, then a vivid orange - when temp. reached 110°C it was a light, bright red - not maroon or burgandy or cranberry

TEST DATA AND OBSERVATIONS:
7A) A peak appears at 1830 cm⁻¹, indicating the phthalic anhydride has not totally reacted - the anhydride peak still appears indicating an incomplete reaction

3:45-5:00 pm; 8:15-9:30 am 8 Continue heating flask at 110°C for 3.5 hours
9:30 am 9 Run IR Test

TEST DATA AND OBSERVATIONS:
9A) A peak still appears at 1830 cm⁻¹ - the phthalic anhydride was charged in excess so the anhydride peak will always be present. Continue with reaction!

no printout available from IR - paper roll not working correctly

NEUTRALIZATION:

④ RAW MATERIALS:

	Weight (g)	Lot #
93% sulfuric acid	3.7g	B4206

⑤ PROCEDURE:
1 Charge 93% H_2SO_4 slowly through the condenser
2 Raise temperature to 150°C

changed color from light, bright red to a deep burgandy - looks much more magenta - no vapor from H_2SO_4 - exo. a little @ 150°C

Date May 13, 1996 Signature Gloria G. Wickens
Date May 14, 1996 Witness Vicki Gilder

(CONTINUE TO PAGE 10022-15)

PREPARATION OF EXPERIMENTAL INK BLUE 92

① EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser
2- 600 mL beaker
2- 100 mL beaker
stir plate
stir bars

② RAW MATERIALS:

	Weight (g)	Lot #'s
experimental coupler	34.0 g	10022-12
OFBSA	10.4 g	S8266
93% H_2SO_4	5.2 g	X3077
urea	3.0 g	G6819
dichloromethane	20.0 g	05816LG
benzoyl peroxide	3.0 g	02106BQ

③ PROCEDURE:
1 Charge experimental coupler to the flask and begin stirring
2 Charge OFBSA
3 Charge 93% H_2SO_4
4 Charge urea and equip flask with a condenser
3:40pm - 5:00pm 5 Heat reaction to 100-105°C
8:15am - 11:00am 6 Continue heating at 100-105°C for 4 hours
7 Remove 5g of sample and place it in a 100mL beaker
8 Stop reaction; retain in flask for future reaction (if necessary)
9 Charge dichloromethane to beaker (while stirring on stir plate); color must fully dissolve in dichloromethane
10 Charge benzoyl peroxide to the homogeneous liquid
11 Stir on stir plate for 20 minutes (should be completely homogeneous)

reaction at 25°C is like concrete - an extremely hard blue rock!

OBSERVATIONS:
mixture did not immediately turn blue - first white cream, light blue then dark blue
reaction is very hard and rubbery - like a dark blue rubber ball even when warm
not completely soluble in dichloromethane
began to thicken

OBSERVATIONS: WHEN BENZOYL PEROXIDE WAS ADDED AND THE STIRRING SESSION (20 MIN.) BEGAN, LIQUID BEGAN TO THICKEN - AT THE END OF 20 MIN., MIXTURE HAD BECOME A HARD, DARK BLUE SEMI-SOLID - MIXTURE IS EVENTUALLY SOLUBLE IN METHANOL (ALLOWED TO SIT OVERNIGHT) - THE COLOR IS A DARK, BROWNISH BLUE

12 Stopped overnight.

Date May 13, 1996 Signature Glenn G. Dickum
Date May 14, 1996 Witness [illegible]

((CONTINUED TO PAGE 10022-16))

CONTINUED FROM PAGE 10022-13

((PREPARATION OF EXPERIMENTAL MAGENTA))

9:45 am - 1:00 pm ³Continue heating at 150°C for 3.25 hours

1:00 pm ⁴Run color value, DV-478

TEST DATA AND OBSERVATIONS:

4A) color value, DV-478

Sample Weight (SW) = 0.2397g

absorbancy (A) = 0.763

Wavelength (λ) = 527

color value = (A/SW) x 5

color value = (0.763/0.2397) x 5 = 15.9

4B) color looks very weak, but the dye peak is a large one - has a theoretical color value of 37

⁵Run pH, DV-5 then neutralize to a pH of 6.5-7.5 if necessary

TEST DATA AND OBSERVATIONS:

5A) ① add 30.0g H_2O to color and mix well

② run pH; pH = 1.77

pH meter = Omega # SRB 00019

③ adjust pH to 6.5-7.5 with 50% Caustic, Lot# Q1570

④ adjusted pH = 7.57 @ 60°C

⁶If layers are present, separate in separatory funnel

⁷Discard water layer; separate water and color layers

⁸Retain full color layer/product for future analysis.

OBSERVATIONS:
dark cranberry colorant resisted mixing with the H_2O - after a time it became a homogeneous thick liquid - once caustic was added it thinned extremely and exothermed to 65°C - also the color changed dramatically to a very red liquid

A water layer is present - it is very light, thin and oily

End product is an orangish red - not really a magenta color at all - smells highly of dichloromethane (still even after separating)

Date May 14, 1996 Signature Gloria G. Dickson

Date May 20, 1996 Witness Kellie Pettison

CONTINUED FROM PAGE 10022-14

((PREPARATION OF EXPERIMENTAL INK BLUE 92))

[13] Dissolve product in acetone
[14] Stop reaction; discard all product.

OBSERVATIONS:

Product set-up and became a hard, dark blue rock of a substance - it does not dissolve in methanol or acetone. A color value test is not even possible because the solid product will not dissolve in anything. Cleaning is impossible - will not even come off in H_2SO_4 (98%)!

PREPARATION OF LIQUITONE BLUE 452 W (@ CV = 42)

LIQUITONE BLUE 452W, LOT # D1013 : Color value = 48.1196 (DV-478)
% solids = 68%

⟹ COLOR MUST BEAT A COLOR VALUE, DV-478 (in methanol) OF 42 (NOT 48) TO SAMPLE TO CUSTOMERS

PROCEDURE AND DATA FOR DILUTION OF PRODUCT, TO ACHIEVE COLOR VALUE OF 42:

Color + water = total color

$$\frac{(\text{Color}) \times 48}{42} = X$$

$X - (\text{Color})$ = amount of H_2O to add

$$\boxed{\frac{(2823.4) \times 48}{42} = (3226.74) - (2823.4) = 403.3\text{g of } H_2O}$$

[1] Add 403.3g H_2O to 2823.4g LiquiTone Blue 452W
[2] Mix thoroughly
[3] Run color value, DV-478:
Sample weight (sw) = 0.1566g
Wavelength (λ) = 592
absorbancy (A) = 1.308
Color value = (A/sw) x 5
Color value = (1.308/0.1566) x 5
Color value = 41.76

CV = 42

READY TO BE SAMPLED!!

Date May 15, 1996 Signature Glorie G. Dickson
Date May 21 1996 Witness

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
aniline 2,6	52.9 g	9831-21
p-DMAB	7.5 g	X3052
urea	3.0 g	G6819
muriatic acid	10.3 g	W2811
hydrogen peroxide	9.7 g	W2894
water	9.7 g	—

CONDENSATION:

③ PROCEDURE: 1 Charge aniline 2,6 and begin stirring
2 Charge p-DMAB
3 Charge urea
4 Charge muriatic acid and quickly equip flask with a condenser
10:15 am 5 Heat reaction to 100-110°C (105°C)
2:15 pm 6 Continue heating at 105°C for 4 hours

OBSERVATIONS: Turned a reddish blue - almost purple

OXIDATION:

④ PROCEDURE: 1 Charge the mixture of H_2O/H_2O_2 slowly through the condenser
2 Continue heating at 105°C for 2 hours
3 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

3A) color value, DV-478
sample weight (sw) = 0.1744 g
wavelength (λ) = 589
absorbancy (A) = 0.247
color value = $(A/sw) \times 5 = (0.247/0.1744) \times 5 = \underline{7.08}$

3B) Color value is extremely low - the UV/Vis also shows unreacted material in the rxn. Also, the color itself is not homogeneous - it has distinct layers (all blue) in the flask - thick on bottom, thin/liquidy on top

4 Stop reaction; dispose of final product.

Date May 17, 1996 Signature Gloria G. Wickens
Date May 21 1996 Witness

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
aniline 2,6	52.9g	9831-21
p-DMAB	7.5g	X3052
urea	3.0g	G6819
93% sulfuric acid	5.3g	B4206

CONDENSATION:

③ PROCEDURE:
1 Charge aniline 2,6 and begin stirring
2 Charge p-DMAB
3 Charge urea
4 Charge sulfuric acid and equip flask with a condenser
10:15am 5 Heat reaction to 100-110°C
6 Continue heating reaction at 105°C for 4 hours
2:15pm 7 Run color value, DV-288

OBSERVATIONS: When acid was added, it immediately turned dark blue

thin, reddish-blue (dark) liquid

REAGENTS:
methanol
CrO_3 (chromium (III) oxide) ⟶ Lot # 05227AG
distilled water

Sample weight (sw) = 0.2075g
absorbance at maximum (A) = 0.045
Condensation absorptivity = (A/sw) × 15.8
= (0.045/0.2075) × 15.8
Condensation absorptivity = 3.43

OBSERVATIONS:
Color value extremely low - condensation did not work - the product did not oxidize - the CrO_3 did not react with the product - the green CrO_3 powder was still physically present in the 100mL flask dilutions and upon cleaning beaker - it was a hetergeneous mixture. Oxidation was ineffective.

Date May 20, 1996 Signature Gloria G. Dickens
Date MAY 20, 1996 Witness
8 Stopped overnight.

(CONTINUE TO PAGE 10022-20)

PREPARATION OF EXPERIMENTAL IB 1892

① EQUIPMENT: 1- 1000 mL three-neck, round bottom flask
mechanical agitator
thermal controller
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 92	205.9g	A1017
DDSA	106.4g	TRL-6
1-methylimidazole	3.0g	K9037

③ PROCEDURE:
1 Charge Uncut Ink Blue 92 and begin stirring
2 Charge DDSA
3 Charge 1-methylimidazole and equip flask with a condenser
4 Heat flask to 90-98°C
12:00pm 5 Continue heating flask at 90-98°C for 3 hours
3:00pm 6 Run IR

TEST DATA AND OBSERVATIONS:
6A) No peak appears directly at 1830cm^{-1}, but a peak appears between 1860-1850cm^{-1}, then steadies, then slopes down again ~ 1800cm^{-1}. Continue condensing just to be sure color is 100% capped (or DDSA has fully reacted). > NO IR GRAPH!
7 Continue condensing at 90-98°C for 2 hours
8 Run IR

TEST DATA AND OBSERVATIONS:
8A) No peak appears at 1830cm^{-1} - this indicates the aldehyde has reacted with all 4 OH groups to form the necessary ester groups. The IR data shows that the reaction is complete!
9 Stop reaction; retain full sample, distribute as such:
4 oz. sample jar } SAVE: possibly send to customer
4 oz. sample jar }
3.5 oz: Craig Collins for analysis!

Date May 20, 1996 Signature Gloria G. Wickham
Date May 20 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-18

((PREPARATION OF EXPERIMENTAL INK BLUE))

OXIDATION: REAGENT/OXIDIZER: BENZOQUINONE, LOT # Q1540

(4) PROCEDURE:
1 Heat reaction to 100-110°C
2 Remove 5g color to a beaker (with a stir bar)
3 Charge 1g benzoquinone to the beaker (while stirring)
4 Heat beaker to 70-90°C
5 Remove from heat once it reaches peak temperature
6 Run color value, DV-478

OBSERVATIONS:
Contents of beaker set-up quickly - turned rock hard - acetone is not dissolving/helping

TEST DATA AND OBSERVATIONS: COLOR VALUE, DV-478

6A) Sample weight (sw) = 0.3203g
wavelength (λ) = 592.0
absorbancy (A) = 2.725
color value = (A/sw) × 5
color value = (2.725/0.3203) × 5
color value = 42.54

6B) Oxidation with benzoquinone is totally effective - color and dye peak are extremely strong. Color yields a high color value after complete, effective oxidation. Continue with benzoquinone oxidation.

7 Lower reaction temperature to 60-65°C
8 Charge 10.8g benzoquinone to reaction
9 Heat reaction to 75-80°C
10 Continue heating at 75-80°C for 45 minutes
11 Run color value, DV-478

10:00am
10:45am
10:45am

OBSERVATIONS:
Rxn. exothermed to 122°C - flasks looks black but color still looks good

TEST DATA AND OBSERVATIONS: COLOR VALUE, DV-478

11A) Sample weight (sw) = 0.2177g
wavelength (λ) = 593
absorbancy (A) = 1.859
color value = (A/sw) × 5
color value = (1.859/0.2177) × 5
color value = 42.69

11B) Complete oxidation is very effective - a high color value indicates full oxidation! Great yield in color product thus far.

(5) RAW MATERIALS:

	Weight (g)	Lot #
Glydexx ND-101	44g	26761-45-5

(6) PROCEDURE: 1 Through an addition funnel, charge the Glydexx slowly to the reaction flask, when the temperature is raised to 100°C

Date May 21, 1996 Signature Gloria G. Wickens
Date May 21 1996 Witness [illegible]

((CONTINUE TO PAGE 10022-21))

CONTINUED FROM PAGE 10022-20

((PREPARATION OF EXPERIMENTAL INK BLUE))

2 Continue heating reaction at 100°C for 2 hours

3 Run color value, DV-478

TEST DATA AND OBSERVATIONS : COLOR VALUE, DV-478

3A) Sample weight (sw) = 0.1467g
Wavelength (λ) = 593.0
absorbancy (A) = 0.561
Color value = (A/sw) x 5
Color value = (0.561/0.1467) x 5
Color value = 19.12

3B) Appx. 10 units lower than theoretical color value - after Glydexx, reaction became somewhat thinner - Glydexx may have caused a loss of color

4 Run ph, DV-5 and neutralize to a pH of 5.0-7.0

TEST DATA AND OBSERVATIONS:

4A) BEGINNING pH = 8.23
⟹ neutralize with 93% H_2SO_4, Lot# B4206
ENDING pH = 6.10

4B) After neutralization, there appeared to be no separation - a sep. funnel separation of layers is not necessary

5 Stop reaction; retain full sample in an 8 oz. jar for future testing

⟹ FINAL PRODUCT OBSERVATION: A dark reddish blue liquid that has a peculiar liquid smell (not able to identify the odor - unlike any other odor)

Date May 21, 1996 Signature Gloria G. Wickem

Date May 22, 1996 Witness Kelly Pemberton

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
1-250 mL addition funnel
Condenser

② RAW MATERIALS:

	Weight (g)	Lot #
aniline 2,6	52.9 g	9831-21
p-DMAB	7.5 g	B4090
urea	3.0 g	G6819
93% sulfuric acid	5.3 g	B4206
Glydexx ND-101	44 g	26761-45-5

CONDENSATION: 10 hours

③ PROCEDURE:

1 Charge aniline 2,6 and begin stirring
2 Charge p-DMAB
3 Charge urea
4 Charge H_2SO_4 and equip flask with a condenser — 9^{45} am - 6^{00} pm
5 Heat reaction to 100-110°C — 8^{30} am - 10^{30} am
6 Continue heating reaction at 105°C for 10 hours — 10^{30} am
7 Charge Glydexx slowly, through addition funnel, at 100°C — 10^{45} am - 12^{45} pm
8 Continue heating reaction at 100°C for 2 hours

OXIDATION: BENZOQUINONE, LOT # Q1540

9 Charge 10.8 g of benzoquinone to reaction at 60-65°C
10 Heat to 75-80°C
11 Continue heating at 75-80°C for 1 hour
12 Run color value, DU-478

TEST DATA AND OBSERVATIONS: COLOR VALUE, DU-478

12A) Sample weight (SW) = 0.1525 g
Wavelength (λ) = 593.0
absorbancy (A) = 0.348
Color value = (A/SW) x 5
Color value = (0.348/0.1525) x 5 = 11.4

Date May 22, 1996 Signature Gloria G. Wickham
Date May 22, 1996 Witness Kelly Pearson

(CONTINUE TO PAGE 10022-25)

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
aniline 2,6	52.9g	9831-21
p-DMAB	7.5g	B4090
urea	3.0g	G6819
93% sulfuric acid	5.3g	B4208
benzoquinone	~~11.8g~~ 05/23/96	Q1540
	5.4g	

CONDENSATION: 10 hours

③ PROCEDURE:
1 Charge aniline 2,6 and begin stirring
2 Charge p-DMAB
3 Charge urea
4 Charge H_2SO_4 and equip flask with a condenser
5 Heat reaction to 100-110°C
6 Continue heating reaction at 105°C for 10 hours — 9^{45} am - 6^{00} pm
7 Lower reaction temperature to 60-65°C — 8^{20} am - 10^{30} am
8 Charge benzoquinone (5.4g)
9 Heat reaction to 75-80°C
10 Continue heating at 75-80°C for 1.0 hours
11 Run color value, DV-478

TEST DATA AND OBSERVATIONS: COLOR VALUE, DV-478

11A) Sample weight (sw) = 0.1499g
Wavelength (λ) = 593.0
absorbancy (A) = 0.902
color value = (A/sw) x 5
color value = (0.902/0.1499) x 5
color value = 30.08

11B) This reaction does not yield the highest of color values when just ½ of the full molar equivalence of benzoquinone is used. It seems to require the full (100%) molar ratio of benzoquinone.

12 Reduce heat to 60-65°C
13 Charge benzoquinone (5.4g)
14 Heat reaction to 75-80°C

Date May 22, 1996 Signature Gloria G. Wickens
Date May 22, 1996 Witness Kellie Johnson

(CONTINUE TO PAGE 10022-24)

CONTINUED FROM ~~TO~~ PAGE 10022-23
gd 5/22/96

((PREPARATION OF EXPERIMENTAL INK BLUE))

15 CONTINUE heating at 75-80°C for 1.0 hours
16 Run color value, DU-475

TEST DATA AND OBSERVATIONS: COLOR VALUE, DU-475

16A) Sample weight (sw): 0.1964g
Wavelength (λ): 593.0
absorbancy (A) = 1.343
color value = (A/sw) × 5
color value = (1.343/0.1964) × 5
color value = 34.19

16B) The reaction obviously needs the full amount of benzoquinone added all at once! It has not oxidized as well in 2 steps of benzoquinone addition as it appears to have in 1 step (cite previous reactions).

17 Stop reaction; discard final product down the drain.

(4) CONCLUSIONS: The benzoquinone oxidation needs to take place in 1 step and be at a full molar ratio. It does not yield as high a color value when oxidized in two steps as when oxidized in one step (at full molar equivalence).

Date May 22, 1996
Date May 22, 1996

Signature Gloria G. Wickham
Witness Kelly Pearson

CONTINUED FROM PAGE 10022-22

((PREPARATION OF EXPERIMENTAL INK BLUE))

13 Stop reaction; discard final product down the drain.

(4) CONCLUSIONS: When Glydexx is added (either before or after oxidation), the color appears to deteroriate. Glydexx reduces color value, thus dye peak/color yield of a product. It does not help a stable color - it hinders it.

Date May 23, 1996
Date May 24, 1996

Signature Gloria G. Richman
Witness Kelli Pearson

PREPARATION OF EXPERIMENTAL INK BLUE 1892

① EQUIPMENT: 1-3000mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 92	823.6 g	A1017
DDSA	425.6 g	TRL-6
1-methylimidazole	12 g	K9037

③ PROCEDURE:
1 Charge Uncut Ink Blue 92 and begin stirring
2 Charge DDSA
3 Charge 1-methylimidazole and equip flask with a condenser
4 Heat flask to 90-98°C
5 Continue heating flask at 90-98°C for 3 hours
6 Run IR

TEST DATA AND OBSERVATIONS:
6A) No peak appears at 1830 cm^{-1} - this indicates the aldehyde has reacted with all 4 OH groups to form the necessary ester groups. The IR data shows the reaction to be complete.

7 Test color and bleed (H_2O) through drawdown preparation

TEST DATA AND OBSERVATIONS:
DRAWDOWN/INK PREP:
Exp. IB 1892 - Lot# 10022-26 : 1.5g
distilled H_2O - ——— : 6.0g
ethanolamine - Lot# 0301SLN : 0.05g
→ drawdown appears bright and vivid with an acceptable lay-down; it is also bleed-resistant - DOES NOT BLEED!

8 Stop reaction; retain full sample in 5-8oz. and 1-4oz. jar

⟹ COLORANT IS ACCEPTABLE TO SAMPLE TO CUSTOMERS!

Date May 23, 1996 Signature Gloria G. Wichem
Date May 24, 1996 Witness Kellie Reaburn

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1- 250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
PDEA	36.2 g	F1030
p-DMAB	14.9 g	B4090
muriatic acid	11.8 g	W2811
urea	6.0 g	G6819
hydrogen peroxide	19.4 g	W2894
water	19.4 g	—

CONDENSATION: 3 HOURS

③ PROCEDURE:
1 Charge PDEA to flask and begin stirring
2 Charge p-DMAB
3 Charge muriatic acid
4 Charge urea and equip flask with a condenser
5 Begin heating flask to 100-105°C
6 Continue heating reaction at 100-105°C for 3 hours

OBSERVATIONS: product is a dark blue, thin liquid at the beginning - begins to thicken through time

OXIDATION:

④ PROCEDURE:
1 Reduce heat to 95-100°C
2 Mix H_2O_2 and H_2O together well
3 SLOWLY charge H_2O_2/H_2O mixture through the condenser, keeping temperature between 93-102°C
4 Continue heating at 95-100°C for 1 hour (to complete oxidation)
5 Run color value, DV-478

take appx. 30 minutes to charge H_2O_2/H_2O-mix do it slowly - if done too quickly, contents swell and threaten to come out of flask and condenser

TEST DATA AND OBSERVATIONS:

5A) Color value, DV-478
Sample weight (SW) = 0.1575 g
wavelength (λ) = 591
absorbancy (A) = 0.0278
Color value = (A/SW) x 5 = (0.0278/0.1575) x 5 = 0.8825

5B) Lowest color value I have ever seen - oxidation apparently did not/is not successful. The product (for SW) is in 2 layers - a thick product layer and a thin, H_2O layer.

Date May 24, 1996 Signature Gloria G. Wickham
Date May 24, 1996 Witness Kellie Lawson

((CONTINUE TO PAGE 10022-29))

OVEN STABILITY TESTING ON EXP. IB 1892's

⟹ *NOTE: THE H_2O-BASED DRAWDOWN PAPER HAS CHANGED - A NEW SHIPMENT HAS COME IN - IT IS BRIGHTER AND WHITER THAN THE PREVIOUS PAPER. A SHADE DIFFERENCE MAY BE OBSERVED BUT THE DENSITY OF THE TWO PAPERS SHOULD BE EQUAL!

⟹ PREPARE A DRAWDOWN AND RECORD RESULTS ON EACH OF THE OVEN-TESTED INKS: CHECK FOR WATER-BLEED AND COLOR STABILITY

⟹ OVEN-AGED @ 135°F SINCE: Lot # 10055-64 : 04/19/96
Lot # 10055-70 : 04/29/96

(A) Lot # 10055-64 : DDSA with NH_4OH :
- extremely reddish-blue color - became even redder
- no water-bleed

(B) Lot # 10055-64: DDSA with ethanolamine :
- same color - no color change from Lot # 10022-10
- partial water-bleed

(C) Lot # 10055-64: DDSA with diethanolamine :
- same color - no change from Lot # 10022-10
- water-bleed

(D) Lot # 10055-70 : DDSA-B with NH_4OH
- same color - no change from Lot # 10022-10
- partial water-bleed (* HAS BECOME LESS BLEED-RESISTANT THROUGH TIME)

(E) Lot # 10055-70 : DDSA-B with ethanolamine :
- a slight change in color - turned slightly redder
- water-bleed

(F) Lot # 10055-70 : DDSA-B with diethanolamine :
- a slight change in color - turned slightly redder
- extreme water-bleed

⟹ RETURN INK VIALS TO OVEN @ 135°F

→ DDSA : 5 WEEKS
DDSA-B : 4 WEEKS

Date May 24, 1996 Signature Gloria G. Wickham
Date May 28, 1996 Witness Kelle Pearson

PREPARATION OF SAMPLE # 10-1049

① SAMPLE REQUEST:

LiquiTone Yellow 800W	8 oz.
LiquiTone Red 897 W	8 oz.

⟹ for flexo use
⟹ SENT TO: Eagle International
ATTN: Dr. Benemeli's
⟹ SHIPPING DATE: 05/29/96 AB# 5989275983, AIRBORNE

A) RAW MATERIALS:

	Weight(g)	Lot #
Uncut Ink Red 1897	200g	RD04
Water	33.6 g	—
ammonium hydroxide	15.0 g	T2166

⟹ mix well on stir plate with stir bar; pour into an 8 oz. jar
⟹ MIXED WELL: LIQUITONE RED 897 W

B) RAW MATERIALS:

	Weight(g)	Lot #
Uncut Ink Yellow 1800	200g	RD05
Water ~~ammonium~~ gd 5/28/96	21 g	—
ammonium hydroxide	12 g	T2166

⟹ mix well on stir plate with stir bar; pour into an 8 oz. jar
⟹ MIXED WELL: LIQUITONE YELLOW 800W

CONTINUED FROM PAGE 10022-27

((PREPARATION OF EXPERIMENTAL INKBLUE))

6 Stop reaction; dispose of product

⑤ OXIDATION DISCUSSION: Reaction appears not to have oxidized in the H_2O_2/H_2O mixture - the product is in two layers. When 5g of color is oxidized with 0.5g benzoquinone (Lot# Q1540) (@ 75-80°C for 45 min.) the final oxidized product is a dark brownish-blue super solid that is insoluble in: ACETONE, METHANOL, TOLUENE, DMF, DICHLOROMETHANE. The product is rock hard and very dull in color. DISPOSE TOTALLY OF FINAL ROCK SOLID PRODUCT!

Date May 28, 1996 Signature Gloria G. Dickens
Date June 4, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK RED

① EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
mechanical agitator
thermal controller
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Reactint Red X64	170.0g	A1008
ODSA	99.0g	TMX-4
1-methylimidazole	2.7g	K9037

③ PROCEDURE:
1 Charge Reactint Red X64 to flask and begin stirring
2 Charge ODSA
3 Charge 1-methylimidazole and equip flask with a condenser
4 Heat reaction flask to 85-90°C
5 Continue heating flask at 85-90°C for 3 hours
6 Run IR

TEST DATA AND OBSERVATIONS:
6A) No peak appears at 1830 cm^{-1}, indicating the reaction is complete. The anhydride has fully reacted with the OH groups to produce the necessary ester groups.
7 Stop reaction; retain small portion in a vial for future testing.
8 Pour the rest into an 8 oz. jar to sample to a customer.

⟹ SHIP TO:
Independent Ink, Inc.
ATTN: Ramesh Babu

⟹ for ink jet use

⟹ PRODUCT NAME: Experimental Red 10022-30

AIRBORNE, AB# 5989275283

Date May 29, 1996 Signature Gloria G. Dickens

Date June 4, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Violet (Hi-Vis Bulbeye)	100g	B1054
Glydexx	82.6g	26761-45-5
KOH pellets	1.8g	07229BQ

③ PROCEDURE: [1] Strip Uncut Violet on rotovap for 2 hours - strip off all H_2O - % water needs to be less than 1.0%

TEST DATA AND OBSERVATIONS:

1A) ROTOVAP STRIP: TEMP. = 99°C
SPEED = 150 RPM
TIME = 2.0 hours

FINAL % WATER, DV-13: Sample weight = 0.1113g
% water = 0.640%

[2] Charge Uncut (Stripped) Violet to flask and begin stirring
[3] Charge 41.3g Glydexx
[4] Charge 1.4g KOH pellets and equip flask with a condenser
[5] Heat flask to 250°F
[6] Continue heating flask at 250°F for 2 hours
[7] Place a drop of product in a small beaker full of water - note results: PRODUCT DISSOLVED QUICKLY! RXN. NOT COMPLETE!
[8] Charge 41.3g Glydexx
[9] Charge 0.413g KOH pellets
[10] Continue heating flask at 250°F for 2 hours
[11] Test again - place a drop of product in a small beaker full of water - note results: PRODUCT DISSOLVED QUICKLY!
[12] Stop reaction; discard in waste colorant container.

④ CONCLUSION: Final product theoretically has a long polymer, which would cause it to become somewhat oily. Product theoretically should not have dissolved in H_2O - should has risen to the top in an oily bubble.

Date May 29, 1996 Signature Gloria G. Dickens
Date June 4, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1- 250-mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 452	66.8g	RD02
Glydexx	49.4g	26761-45-5
KOH pellets	1.15g	07229BQ

③ PROCEDURE:
1 Charge Uncut Ink Blue 452 and begin stirring
2 Charge Glydexx
3 Charge KOH pellets and equip flask with a condenser
4 Heat flask to 250°F
5 Continue heating at 250°F for 2 hours
6 Place 1 drop in a small beaker full of water - NOTE
RESULTS: SOLUBLE (COMPLETELY) IN H_2O!
7 Run color value, DV-478

TEST DATA AND OBSERVATIONS

7A) Sample weight (sw) = 0.1483g
wavelength (λ) = 593.0
absorbancy (A) = 0.156
Color value = $(A/sw) \times 5$
Color value = $(0.156/0.1483) \times 5$
Color value = 5.26
8 Stop reaction; retain small sample and dispose of the rest.

7B) Glydexx caused the color to deteriorate quickly - it is also completely soluble in water.

GPC TESTING OF EXP. IB 1892

PREPARATION OF SAMPLE:
1 Charge 0.5g sample to 100 mL volumetric flask
2 Fill to meniscus with THF
3 Mix well; submit to Eric Stephens for GPC testing

Sample weight: 0.4981g
Lot # 10055-58 (1892 w/ DDSA)
THF: Lot # 36106

Date May 30, 1996 Signature Gloria G. Wickham
Date June 4, 1996 Witness Debra L. Gilder

PREPARATION OF EXPERIMENTAL INK BLUE 18452

① EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 452	66.8 g	RD02
DDSA	53.2 g	TRL-6
1-methylimidazole	1.1 g	K9037

③ PROCEDURE: [1] Charge Uncut Ink Blue 452 and begin stirring
[2] Charge DDSA
[3] Charge 1-methylimidazole and ~~begin stirr~~ gd 5/30/96 equip flask with a condenser
[4] Begin heating flask to 85-90°C
[5] Continue heating at 85-90°C for 3 hours
[6] Run IR

TEST DATA AND OBSERVATIONS:

6A) IR MACHINE IS "DEAD"

[7] Place a drop of product into a small beaker full of water - note results: PRODUCT IS INSOLUBLE IN H_2O
[8] Place a drop of product into a small beaker full of Rotosolve solvent - note results: PRODUCT IS INSOLUBLE IN ROTOSOLVE (It should be soluble)
[9] Stop reaction; retain full product in a 4 oz. jar.

TEST OF UNCUT INK BLUE 452

① MIX: Uncut Ink Blue 452 - Lot # RD02 : 20 g
Adogen - Lot # 94095053 : 4 g
Methanol — : 6 g

② Discard product

⟹ Stir until soluble
⟹ Place a drop of mixture into a small beaker full of Rotosolve solvent: IS NOT SOLUBLE - VERY OILY

Date May 30, 1996 Signature Glorie G. Wickam
Date June 4, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1-250mL three-neck, roundbottom flask
mechanical agitator
thermal controller
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 452	66.8 g	RD02
ASA 1518	53.2 g	19968-184
1-methylimidazole	1.1 g	K9037

③ PROCEDURE:
1 Charge Uncut Ink Blue 452 and begin stirring
2 Charge ASA 1518
3 Charge 1-methylimidazole and equip flask with a condenser
4 Begin heating flask to 85-90°c
5 Continue heating at 85-90°C for 3 hours
6 Place a drop of product in a small beaker full of water - note results: PRODUCT IS SLIGHTLY SOLUBLE IN WATER
7 Stop reaction; discard entire final product.

Date May 31, 1996 Signature Gloria G. Wickum
Date June 4, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE

INTERMEDIATE SYNTHESIS

① EQUIPMENT : 1-250 mL three-neck, round bottom flask
thermal controller
mechanical agitator
condenser
Erlenmeyer flask
Buchner ~~flask~~ funnel
water vaccum system
filter paper

② RAW MATERIALS :

	Weight (g)	Lot #	MW
lauryl alcohol	37.2g	X3003	186
Sebacoyl chloride	47.8g	11729HG	239.14

③ PROCEDURE : 1 Charge lauryl alcohol and begin stirring
2 Slowly charge sebacoyl chloride and equip flask w/a condenser
3 Heat reaction to 45°C
4 Continue heating at 45°C for 2 hours

OBSERVATIONS: HCl gas comes off when chloride is added - rxn. exothermed to 60°C for ½ hour

④ RAW MATERIALS :

	Weight (g)	Lot #
Sodium bicarbonate (BAKING SODA)	15 g	ARM+HAMMER
Water	200g	—

⑤ PROCEDURE : 1 Mix sodium bicarbonate and water well
2 Slowly charge mixture to reaction, keeping temperature at 50°C
3 Mix well
4 Empty flask contents into a beaker
5 Adjust pH to 6.0-7.5 with sodium bicarbonate and water

exothermed to 60°C liquid became a light brown porous solid that held the excess H_2O - smoked somewhat and bubbled slightly

TEST DATA AND OBSERVATIONS : pH, DV-5
5A) BEGINNING pH = 1.47
ENDING pH = 6.45
6 Filter product using water vaccum system
7 Dry solid particulate matter until it becomes like a powder
8 LET DRY OVERNIGHT!

final solid product is a light brown powder

(CONTINUE TO PAGE 10022-36)

Date June 3, 1996 Signature Gloria G. Wickham
Date June 3, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-35

((PREPARATION OF EXPERIMENTAL INK BLUE))

COLORANT SYNTHESIS

(6) EQUIPMENT: 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

(7) RAW MATERIALS:

	Weight (g)	Lot #	MW
intermediate	37.07	—	388.64
Uncut Ink Blue 452	31.86	RD02	—

(8) PROCEDURE: [1] Charge Uncut Ink Blue 452 and begin stirring
[2] Charge 0.5 of total intermediate product (appx. 31.86g) and equip flask with a condenser (★ SAVE REMAINING ½ FOR FUTURE USE)
[3] Begin heating reaction to 45-50°C
[4] Continue heating reaction at 45-50°C for two hours
[5] Place one drop of product into a small beaker, NOTE RESULTS:
Product is partially soluble in H_2O; completely soluble in methanol
[6] Stop reaction; discard full product.

OBSERVATIONS

appearance like a dark blue icing

Date June 4, 1996 Signature Gloria G. Wickens
Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE

① EQUIPMENT: 1- 250 mL three-neck, roundbottom flask
mechanical agitator
thermal controller
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 452	66.8g	RD02
XR - 4913	30.0g	
1-methylimidazole	1.0g	K9037

③ PROCEDURE:
1 Charge Uncut Ink Blue 452 and begin stirring
2 Charge XR-4913
3 Charge 1-methylimidazole and equip flask with a condenser
4 Begin heating reaction to 90-95°C
5 Continue heating at 90-95°C for 3 hours
6 Place a drop of product in a small beaker full of H_2O -
NOTE RESULTS: Water soluble
7 Charge 10g XR-4913
8 Continue heating at 90-95°C for 1.0 hours
9 Place a drop of product in a small beaker full of H_2O -
NOTE RESULTS: Water soluble
10 Charge 10g XR-4913
11 Continue heating at 90-95°C for 1.5 hours
12 Place a drop of product in a small beaker full of H_2O -
NOTE RESULTS: Water soluble
13 Stop reaction; discard full product.

Date June 4, 1996 Signature Gloria G. Dickens
Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF ETHANOLAMINE CUT 1892

① EQUIPMENT: 1-1000 mL three-neck, round bottom flask
mechanical agitator
2 flask (rubber) stoppers

② RAW MATERIALS:

	Weight (g)	Lot #
LiquiTone Blue 892	600g	RD01
ethanolamine	43g	0301SLN
distilled water	77g	—

③ PROCEDURE: [1] Mix all three raw materials together by mechanical agitator
[2] Mix thoroughly (appx. 3 hours)
[3] Label product (final; homogeneous): LIQUITONE FLEXO BLUE (GS) 892W

④ SAMPLE: [1] Send 8 oz. sample to:

① THE INK COMPANY, TOM LUCK
② CPS, PAUL JELONEK
③ FLINT INK CORPORATION, ALAN MOSS

[2] Retain a full vial for future analysis.

Date June 5, 1996 Signature Gloria G. Wickens
Date July 2, 1996 Witness Bill Hopkins

ANALYSIS OF INK RED 97 FOR CUSTOMER

⟹ * INK RED 97
LOT# M1027

① DRAWDOWN FORMULATION
- density needs to be appx. 1.4/1.5
- read density on spectrodensitimeter
- COATED PAPER, AUTOMATIC K-PROOFER

A	B	C
ink : 2.0	ink: 4.0g	ink : 1.5g
TSR : 7.0	TSR: 7.0g	TSR : 7.0g
toluene : 2.0g	toluene = 2.0g	toluene : 2.0g
density = 1.880	density = 2.160	density = 1.772

* D	SEND TO CUST. AS:
ink : 1.0g	1.0g
TSR : 7.0g	3.73g Unirez 1144
toluene : 2.0g	5.27g
density = 1.556	D = 1.556

NIPFORM T1 SPECIFICATIONS

① appearance
② Color value, DV-516 : 31.6 - 33.4
Wavelength, DV-516 : 510 - 516
③ ppm Sodium, DV-316 : 300 max
④ % water, DV-13 : 1.0% max

② COLOR VALUE, DV-516
WAVELENGTH, DV-516
- use methanol as solvent

Sample weight (SW) = 0.2595g
Wavelength (λ) = 512
absorbancy (A) = 0.851
Color value = (A x 10)/SW
Color value = (0.851 x 10)/0.2595 = 8.51/0.2595
Color value = 32.79
Wavelength = 512
} BOTH ARE CENTER-CUT SPECS

③ % WATER, DV-13
A) sample weight = 0.0966g
% water = 0.369%
B) sample weight = 0.1039g
% water = 0.355 %

Date June 10, 1996
Date July 2, 1996

Signature Gloria G. Wichem
Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

(0.1M scale)

① EQUIPMENT: 1-250mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles (M)
Aniline 5	62.6g	10000-47	2.0M
dimethylaminobenzaldehyde	14.9g	B4090	1.0M
muriatic acid	6.39g	W2811	1.75M
urea	6.0g	G6819	1.0M
hydrogen peroxide	6.8g	W2894	2.0M
water	6.8g	—	—

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge muriatic acid
4 Charge urea and equip flask with a condenser
5 Begin heating reaction to 100-105°C
6 Continue heating at 100-105°C for 4 hours

OXIDATION

④ PROCEDURE:
1 Mix H_2O_2 and H_2O together
2 Lower reaction temperature to 95-100°C
3 SLOWLY charge H_2O_2/H_2O mixture through condenser, keeping temperature range steady at 93-101°C
4 Continue heating at 95-100°C for 1 hour
5 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

5A) Color value, DV-478
Sample weight (sw) = 0.1858g
Wavelength (λ) = 316.0
absorbancy (A) = 0.247

Color value = $(A/sw) \times 5$
Color value = $(0.247/0.1858) \times 5$
Color value = 6.65

Date June 12, 1996 Signature Glen G. Wickham
Date July 2, 1996 Witness Bill Hopkins

(CONTINUE TO PAGE 10022-41)

CONTINUED FROM PAGE 10022-40

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Stop reaction; discard full product.

(5) CONCLUSION: Color value extremely low - reaction did not produce the desired results.

Date June 12, 1996 Signature Gloria G. Dickson

Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF SAMPLE # 10-1056

① SAMPLE REQUEST:

(c)	Ink Red 97	8oz.	= Color value = 31.1	LOW END OF SPEC.
(d)	Ink Red 97	8oz.	= Color value = 34.3	HIGH END OF SPEC.
	Ink Red 97	32oz.	= Lot # M1027	
	Ink Red 97	32oz.	= Lot # M1027	
(b)	Ink Blue 452	8oz.	= Color value = 46.8	LOW END OF SPEC.
(a)	Ink Blue 452	8oz.	= Color value = 50.6	HIGH END OF SPEC.

⇒ for Gravure use
⇒ SENT TO: Sun Chemical Corp.
ATTN: Mr. Jack Miller
⇒ SHIPPING DATE: 06/18/96, UPS Ground (CONTAINS TOLUENE)

② PROCEDURE:

⇒ Kevin Gilder cut both IR97 and IB 452 to meet the required color values. A sample was sent of both the low end of the spec. range and the high end. The spectrophotometer printout for both IR97 color values (31.1; 34.3) are on file in G. Dickens' files; the IB452 printouts are on file in K. Gilder's files.

⇒ beginning Lot # for all IR97: M1027

Date June 14, 1996 Signature Gloria G. Dickens

Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask 1-250 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Aniline 5	125.2g	10000-47
p-DMAB	31.0g	B4090
muriatic acid	29.7g	W2811
urea	4.7g	G6819
water	51.0g	—
hydrogen peroxide	45.0g	W2894

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge water (6.0g)
4 Charge muriatic acid, slowly
5 Charge urea and equip flask with a condenser
6 Begin heating to 95-100°C
7 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Mix H_2O and H_2O_2 together
2 Keeping temperature range between 100-108°C, add H_2O/H_2O_2 mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
3 Continue heating at 100-108°C for 45 minutes
4 Run color value, DV-478

TEST DATA AND OBSERVATIONS: COLOR VALUE, DV-478

4A) Sample weight (sw) = 0.1389g
Wavelength (λ) = 590.0
absorbancy (A) = 1.5294
color value = (A/sw) × 5

color value = (1.5294/0.1389) × 5
COLOR VALUE = 55

5 Stopped overnight.

Date June 17, 1996 Signature Glorie G. Wickem
Date July 2, 1996 Witness [illegible]

(CONTINUE TO PAGE 10022-44)

CONTINUED FROM PAGE 10022-43

((PREPARATION OF EXP. INK BLUE 452))

⑤ RAW MATERIALS:

	Weight (g)	Lot #
ammonium hydroxide	10g	T2166
ammonium meta vanadate	0.14g	A382

⑥ PROCEDURE: [1] Charge amv before oxidation: BEFORE ADDITION OF H_2O_2/H_2O MIXTURE

[2] Charge ammonium hydroxide AFTER oxidation

[3] After oxidation is complete and NH_4OH is thoroughly mixed in, check pH and neutralize to 4.5-7.0

TEST DATA AND OBSERVATIONS:

3A) pH = 6.09

⟹ no neutralization necessary

[4] Strip excess water by way of rotovap for 1.5 hours - % H_2O on colorant needs to be less than 1%

OBSERVATIONS: color is very, very thick

TEST DATA AND OBSERVATIONS:

4A) Rotovap strip @ 99°C/180 rpm for 1.5 hours

⟹ % H_2O impossible to measure - colorant is extremely thick, too thick to place in % H_2O aquatester

[5] After water strip, run color value, DV-478

TEST DATA AND OBSERVATIONS:

5A) color value, DV-478

Sample weight (sw) = 0.1132g

wavelength (λ) = 591

absorbancy (A) = 1.4954

color value = (A/sw) × 5

color value = (1.4954/0.1132) × 5

color value = 66.05

((CONTINUE TO PAGE 10022-45))

Date June 18, 1996 Signature Glorie G. Wicker

Date July 2, 1996 Witness Bill Hopkins

CONTINUED FROM PAGE 10022-44

(PREP. OF EXP. INK BLUE 452)

(7) RAW MATERIALS:

	Weight (g)	Lot #
Colorant	50g	
ODSA	92.1g	TMX-4
1-methylimidazole	1.42g	K9037

(8) PROCEDURE:
1 Charge color to 250 mL three-neck, roundbottom flask and begin stirring
2 Charge ODSA
3 Charge 1-methylimidazole and equip flask with a condenser
4 Begin heating reaction to 90-98°C
5 Continue heating at 90-98°C for 3 hours
6 Test for solubility:
A) WATER: INSOLUBLE; COMPLETELY
B) ROTOSOLVE: SOLUBLE
C) METHANOL: SOLUBLE
D) ACETONE: SOLUBLE
* E) MAGIESOL OIL: SOLUBLE
7 Stop reaction; retain full sample.
⟹ END PRODUCT IS EXTREMELY THICK - ALMOST CLASSIFIED AS A SOLID!

(9) CONCLUSION: This marks the first time an Ink Blue 452 (or product thereof) has been water insoluble AND magiesol oil soluble. Reaction produced excellent results - a high, extremely high color value and a pass in the solubility tests. Recommendations are to repeat experiment!

Date June 19, 1996 Signature Gloria G. Wickham

Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500mL three-neck, round bottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Aniline 5	125.2g	10000-47
p-DMAB	31.0g	B4090
Water	51.0g	—
muriatic acid	29.7g	W2811
urea	4.7g	G6819
hydrogen peroxide	45.0g	W2894
ammonium meta vanadate	0.14g	A382

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0g water
4 Charge muriatic acid, slowly
5 Charge urea and equip flask with a condenser
6 Begin heating to 95-100°C
7 Continue heating at 95-100°C for 4 hours

gd 7/1/96

OXIDATION

④ PROCEDURE:
1 Mix H_2O_2 and H_2O together extremely well
2 Keeping temperature range between 100-108°C, add H_2O_2/H_2O mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
3 Once addition is complete, continue heating at 100-108°C for 45 minutes - 1 hour (OXIDATION SHOULD BE COMPLETE)
4 Run color value, DV-478

((CONTINUE TO PAGE 10022-48))

Date July 1, 1996 Signature Gloria G. Wickum
Date July 2, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 92

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Aniline 5	125.2 g	10000-47
~~p-DMAB~~ OFBSA	43.3 g	S8266
water	51.0 g	—
muriatic acid	29.7 g	W2811
urea	4.7 g	G6819
hydrogen peroxide	45.0 g	W2894
ammonium meta vanadate	0.14 g	A382

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge OFBSA
3 Charge 6.0 g water
4 Charge muriatic acid, SLOWLY
5 Charge urea and equip flask with a condenser
6 Begin heating to 95-100°C
7 Continue heating at 95-100°C for 4 hours

OXIDATION

④ PROCEDURE:
1 Mix H_2O_2 and H_2O together extremely well
2 Keeping temperature range between 100-108°C, add H_2O_2/H_2O mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
3 Once addition is complete, continue heating at 100-108°C for 1 hour

((CONTINUE TO PAGE 10022-49))

Date July 1, 1996 Signature Glori G. Dickens
Date July 16, 1996 Witness Bill Hopkins

CONTINUED FROM PAGE 10022-46

((PREPARATION OF EXPERIMENTAL IB 452))

TEST DATA AND OBSERVATIONS:

4A) Color value, DV-478
Sample weight (sw) = 0.1319g
Wavelength (λ) = 589.0
absorbancy (A) = 0.2952
color value = (A/sw) x 5
Color value = (0.2952/0.1319) x 5 = 11.19

5 Stop reaction; discard product, keeping a 4oz. retain sample.

(3) CONCLUSION: Extremely low color value - this may have been caused by an incomplete oxidation. AMV, the oxidation catalyst, was not added, therefore oxidation may not be complete.

Date July 1, 1996 Signature Gloria G. Dickerson

Date July 16, 1996 Witness Bill Tompkins

CONTINUED FROM PAGE 10022-47

(PREPARATION OF EXPERIMENTAL IB 92)

4 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

4A) color value, DV-478

Sample weight (SW) = 0.1242g

Wavelength (λ) = 629.0

absorbancy (A) = 0.1861

color value = (A/SW) x 5

color value = (0.1861/0.1242) x 5

color value = 7.49

5 Stop reaction; discard product, keeping a 4 oz. retain sample.

OBSERVATIONS: color very nice - a nice, smooth yellowish blue - typical IB 92 shade

(5) CONCLUSION: Color value extremely low - this could be because the oxidation catalyst AMV was never added. Recommendation is to repeat experiment using AMV as oxidation catalyst.

Date July 1, 1996 Signature Glen G. Wickham

Date July 16, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
aniline 5	125.2g	10000-47
p-DMAB	31.0g	B4090
water (H_2O)	51.0g	—
muriatic acid (HCl)	29.7g	W2811
urea	4.7g	G6819
hydrogen peroxide (H_2O_2)	45.0g	W2894
ammonium meta vanadate (AMV)	0.14g	A382

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0g water
4 Charge muriatic acid, slowly
5 Charge urea and equip flask with a condenser
6 Begin heating to 95-100°C
7 Continue heating at 95-100°C for 4 hours

OXIDATION

*Charge AMV at 93°C before starting oxidation

④ PROCEDURE:
1 Mix H_2O_2 and H_2O together extremely well
2 Keeping temperature range between 100-108°C, add H_2O_2/H_2O mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
3 Once addition is complete, continue heating at 100-108°C for 1 hour
4 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

4A) sample weight (SW) = 0.1395 absorbancy (A) = 0.5530 C.V. = (0.5530/0.1395)x5
Wavelength (λ) = 589.0 Color value = (A/SW)x5 C.V. = 19.82

Date July 2, 1996 Signature Glenn G. Dickens
Date July 16, 1996 Witness Bill Hopkins

5 Stop reaction; discard full product.

** OBVIOUSLY OXIDATION WAS INCOMPLETE **

RESIMENE REACTIONS

① RAW MATERIALS:

	Lot #
Resimene resin	68002-20-0
Uncut Ink Blue 92	D1008
Ink yellow 850	X1004
toluene	—
Thinned Solution Resinate	—

REACTION 1

① 7:2 RATIO
Uncut IB 92 : 70g
Resimene : 20g

② Heat @ 65°C for 1 hour

③ RESULTS: product formed a thick, blue rubbery mass - resembles chunk of rubber cement - hard yet breakable and soft/moldable - rxn. set-up in flask!

SOLUBILITY TESTS: water:
acetone:
toluene:
methanol:
} SOLUBLE, YET NOT COMPLETELY

④ Retain sample for future analysis; discard remaining product.

⇐ Rxn. completed in 250mL three-neck, roundbottom flask with thermal controller and mechanical agitator

REACTION 2

① 7:2 RATIO
Ink Yellow 850: 70g
Resimene : 20g

② Heat @ 60-80°C for 1 hour on hotplate; used 800mL beaker and mag. stirbar

③ ADDITIONS: 15 min. : 25g toluene
1hr 10 min. : 25g toluene
1 hr 20 min. : 25g toluene

④ RESULTS: product started to thicken - toluene was added to prevent solid formation - even with toluene, crystalization occurred - crystals formed at bottom of beaker

DRAWDOWN PREP.: 10g color
27g TSR
12.5g toluene

⇒ prepared DD ink's viscosity @ 18 sec.

Date July 3, 1996 Signature Gloria G. Dickens

Date July 16, 1996 Witness [illegible]

⇒ drawdown very tacky, sticky - probably will block

⑤ Retain full sample; retain drawdown ink.

RESIMENE REACTIONS WITH INK YELLOW 850

① RAW MATERIALS:

	Lot #
Resimene resin	68002-20-0
Ink yellow 850	X1004
toluene	
Thinned Solution Resinate	

② EQUIPMENT: 600 mL beakers
stirplates/hot plates
thermometers
magnetic stirbars

REACTION 1

① 7:3 RATIO
Ink Yellow 850: 70g
Resimene: 30g
② Heat @ 60-80°C for 1.5 hours
③ ADDITIONS: 15 min.: 5g toluene
⟹ @ 55 min.: became thicker, formed a thick ball around stirbar

55 min.: 10g toluene
⟹ product is a viscous ink liquid - has a rubbery glob around stirbar - glob is soft and malleable
④ DRAWDOWN PREP.: ink is @ 18 sec.
color/product: 10g
thinned solution resinate: 27g
toluene: 19g
K-PROOFER DD: drawdown is very clean and not tacky - smears just a tad; appears to be a nice, bright color ink
⟹ product does settle out into layers

REACTION 2

① 7:4 RATIO
Ink Yellow 850: 70g
Resimene: 40g
② Heat @ 60-80°C for 1.5 hours
③ ADDITIONS: 15 min.: 5g toluene
⟹ @ 55 min.: became thicker, formed a thick ball around stirbar

55 min.: 10g toluene
⟹ product is very thick - has hard crystals collected in the bottom - has also a hard crystalline chunk around the stirbar
④ DRAWDOWN PREP.: ink is @ 18 sec.
color/product: 10g
thinned solution resinate: 27g
toluene: 23g
K-PROOFER DD: drawdown is very clean and bright; it is not tacky nor does it smears - appears to be very nice
⟹ product does settle out into layers

Date July 5, 1996 Signature Gloria G. Dickerson
Date July 16, 1996 Witness Lilli Hopkins

★ ⟹ RETAIN BOTH FULL PRODUCTS AND INKS
→ place tape around containers and save for future analysis

SEPARATION ~~EMULSION~~ TESTING ON INK YELLOW 850

gd 7/8/96

① EQUIPMENT: 4 vials with lids

② RAW MATERIALS:

	Lot #
Ink Yellow 850	X1004
Thinned Solution Resinate	—
Siegwerk Uncoated Yellow Line Ink	SAMPLE
toluene	—
aniline 5	10000-47
Silwet L-722	2147EK061192

③ PROCEDURE:
1. Prepare 4 inks in the following corresponding formulations
2. Seal vials and let stand in flammable cabinet
3. Test at a later date

④ FORMULATIONS:

(A)
Siegwerk Line Ink: 8g
TSR: 2g
Ink Yellow 850: —
toluene: 17g

(B)
Siegwerk Line Ink: 8g
TSR: 2g
Ink Yellow 850: 0.24g
toluene: 19g

(C)
Siegwerk Line Ink: 8g
TSR: 2g
Ink Yellow 850: 0.24g
toluene: 17g
aniline 5: 0.10g

(D)
Siegwerk Line Ink: 8g
TSR: 2g
Ink Yellow 850: 0.24g
toluene: 17g
Silwet L-722: 0.10g

Date July 5, 1996 Signature Gloria G. Dickens

Date July 16, 1996 Witness Sally Z. Hopkins

SEPARATION TESTING ON INK YELLOW 850

① EQUIPMENT: 4 vials with lids

② RAW MATERIALS:

	Lot #
Ink Yellow 850	X1004
Thinned Solution Resinate	—
Siegwerk Uncoated Yellow Line Ink	SAMPLE
toluene	—
Aniline 2106	PP101
Aniline 213	O1058
M-Toluidine 2148DA	PP03

③ PROCEDURE: 1 Prepare 4 inks in the following corresponding formulations
2 Seal vials and let stand in flammable cabinet
3 Test at a later date

④ FORMULATIONS:

(A)

Siegwerk Line Ink: 8.0g
TSR: —
Ink Yellow 850: 0.24g
toluene: 17.0g

(B)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Aniline 213: 0.10g

(C)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Aniline 2106: 0.10g

(D)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
M-Toluidine 2148DA: 0.10g

Date July 8, 1996 Signature Gloria G Wickens

Date July 16, 1996 Witness [illegible]

SEPARATION TESTING ON INK YELLOW 850

① EQUIPMENT: 4 vials and lids

② RAW MATERIALS:

	Lot #
Ink Yellow 850	X1004
Thinned Solution Resinate	—
Siegwerk Uncoated Yellow Line Ink	SAMPLE
toluene	—
Syn Fac 334-10	SAMPLE
Syn Fac 8337	SAMPLE
Syn Fac 8210	G1185
Syn Fac 8216	H1095

③ PROCEDURE: 1 Prepare 4 inks in the following corresponding formulations
2 Seal vials and let stand.
3 Test and observe separation at a later date.

④ FORMULATIONS:

(A)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
(17.9g) toluene: 17g → 17.9g
Syn Fac 334-10: 0.10g

(B)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
(17.8g) toluene: 17.8g
Syn Fac 8337: 0.10g

★ (C)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Syn Fac 8210: 0.10g

★ (D)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Syn Fac 8216: 0.10g

Date July 8, 1996 Signature Clovis G. Wickens
Date July 16, 1996 Witness Bill Hopkins

★ #C, D: prepared 07/10/96

COLOR VALUE OF EXP. IB452

⟹ EXP. INK BLUE 452, LOT # 10022-43

COLOR VALUE, DV-478

⟹ use acetone as solvent
Sample weight (sw) = 0.1266g
wavelength (λ) = 593.0
absorbancy (A) = 0.3571
color value = (A/sw) x 5
color value = (0.3571/0.1266) x 5
color value = 14.1

OBSERVATIONS:
Colorant is insoluble in methanol - even in acetone, some particulate matter is observable - mixed solution has small blue "crumbs" throughout

⟹ EXTREMELY LOW COLOR VALUE; COLOR HAS DETERIORATED SINCE LAST COLOR VALUE ANALYSIS

Date July 12, 1996 Signature Alan G. Dickson

Date July 16, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 92

① EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	weight (g)	Lot #
Aniline 5	31.3 g	10000-47
N,N-Dimethylaniline	12.1 g	15428HG
OFBSA	20.8 g	S 8266
muriatic acid	14.9 g	W2811
urea	2.4 g	G6819
ammonium meta vanadate	0.10 g	A382
Water	25.5	—
hydrogen peroxide	22.5	W2894

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge n,n-dimethylaniline
3 Charge OFBSA
4 Charge 3.0 g water
5 Charge amv
6 Charge urea and equip flask with a condenser
7 Charge muriatic acid
8 Heat reaction to 95-100°c
9 Continue heating reaction to 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Mix H_2O and H_2O_2 together
2 Keeping temperature range between 100-108°c, add H_2O/H_2O_2 mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
3 Continue heating at 100-108°c for 45 minutes
4 Run color value, DV-478

Date July 12, 1996 Signature Gloria G. Dickens
Date July 16, 1996 Witness Bill Hopkins

(CONTINUE TO PAGE 10022-58)

CONTINUED FROM PAGE 10022-57

((PREPARATION OF EXPERIMENTAL INK BLUE 92))

TEST DATA AND OBSERVATIONS

4A) Sample weight (sw) = 0.1736g
Wavelength (λ) = 624.0
absorbancy (A) = 0.5721
color value = (A/sw) x 5
color value = (0.5721/0.1736) x 5
color value = 16.47 = 16.5

5 Stop reaction; discard full product.

(5) CONCLUSION: Color value extremely low. Consistency extremely good - mixture is completely homogeneous.

Date July 15, 1996 Signature Alan G. Dickens
Date July 16, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	weight (g)	Lot #
Aniline S	125.2g	10000-65
p-DMAB	31.0g	B4090
muriatic acid	29.7g	W2811
urea	4.7g	G6819
ammonium meta vanadate	0.14g	A382
water	51.0g	—
hydrogen peroxide	45.0g	W2894

CONDENSATION

③ PROCEDURE:
1 Charge aniline S and begin stirring
2 Charge p-DMAB
3 Charge water (6.0g)
4 Charge urea
5 Charge amv and equip flask with a condenser
6 Charge muriatic acid through the condenser
7 Begin heating reaction to 95-100°C
8 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Mix H_2O_2 and remaining H_2O together
2 Keeping temperature range between 100-108°C, add H_2O/H_2O_2 mixture slowly through the condenser (WATCH FOR ENDO/EXOTHERM)
~~3 Continue heating at 1~~ gd 7/15/9
3 Run color value, DV-478

TEST DATA AND OBSERVATIONS:
3A) Sample weight (sw) = 0.1740g
Wavelength (λ) = 591.0
absorbancy (A) = 1.4815
color value = (A/sw) x 5 = (1.4815/0.1740) x 5 = 42.6

Date July 15, 1996
Signature Gloria G. Dickens
Date July 16, 1996
Witness Bill Hopkins

★ EXTREMELY HIGH COLOR VALUE: BEFORE OXIDATION

(CONTINUE TO PAGE 10022-60)

CONTINUED FROM PAGE 10022-59

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

4 Continue heating at 98°C for 1 hour

5 Run color value, DV-478

TEST DATA AND OBSERVATIONS

5A) Sample weight (sw) = 0.1327g

Wavelength (λ) = 590

absorbancy (A) = 1.1351

color value = (A/sw) × 5

color value = (1.1351/0.1327) × 5

color value = 42.769 = 43 ⟸ LOW COLOR VALUE - PROBLEM WITH OXIDATION??

6 Run pH, DV-5

TEST DATA AND OBSERVATIONS

6A) temperature = 98°C

pH = 2.00

7 Stop reaction; retain full sample in an 8oz. jar.

Date July 15, 1996 Signature Alan G. Dickens

Date July 16, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles
Aniline 5	125.2g	10000-65	
p-DMAB	31.0g	B4090	0.2M
muriatic acid	37.06g	W2811	1.5M
urea	4.7g	G6819	
ammonium meta vanadate	0.14g	A382	0.45%
water	51.0g	———	
hydrogen peroxide	45.0g	W2894	

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge water (6.0g)
4 Charge urea and equip flask with a condenser
5 ~~Charge amv and equip flask with a condenser~~ gd 7/16/96
6 Charge muriatic acid through the condenser, slowly
7 Begin heating reaction to 95-100°C
8 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 gd 7/16/96 ~~Mix~~ Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture cautiously, watching for ENDO/EXOTHERM cycle
5 Heat at 98°C for 1 hour

(CONTINUE TO PAGE 10022-63)

Date July 16, 1996 Signature Glen G. Wickham
Date July 16, 1996 Witness Bill Hopkins

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1 - 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles
Aniline 5	125.2g	10000-65	
p-DMAB	31.0g	B4090	0.2M
muriatic acid	29.7g	W2811	1.2M
urea	4.7g	G6819	
ammonium meta vanadate	0.28g	A382	0.9%
Water	51.0g	—	
hydrogen peroxide	45.0g	W2894	

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge water (6.0g)
4 Charge urea and equip flask with a condenser
5 Charge muriatic acid slowly through the condenser
6 Begin heating reaction to 95-100°C
7 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture cautiously through the condenser, watching for ENDO/EXOTHERM cycle
5 Heat at 98°C for 1 hour

((CONTINUE TO PAGE 10022-63))

Date July 16, 1996 Signature Glorin G. Wickham
Date July 16, 1996 Witness Bill Hopkins

CONTINUED FROM PAGE 10022-61

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (sw) = 0.1568g
Wavelength (λ) = 590.0
absorbancy (A) = 1.2361
Color value = (A/sw) x 5
Color value = (1.2361/0.1568) x 5
Color value = 39.4

7 Stop reaction; discard full product.

CONTINUED FROM PAGE 10022-62

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (sw) = 0.1468g
Wavelength (λ) = 591.0
absorbancy (A) = 1.3170
Color value = (A/sw) x 5
Color value = (1.3170/0.1468) x 5
Color value = 44.8 = 45

7 Remove 5.0g of experimental colorant from reaction flask and let cool to room temperature

8 Add 0.5g NH_4OH (LOT# T2166) to color and mix well

9 Run color value, DV-478

TEST DATA AND OBSERVATIONS: Sample weight (sw) = 0.1321g absorbancy = 1.0740
Wavelength (λ) = 590.0 . Color value = (A/sw) x 5
Color value = $\left(\frac{1.0740}{0.1321}\right)$ 5
CV = 40.65

Date July 16, 1996 Signature Gloria G. Wickham

Date July 23, 1996 Witness [illegible]

10 Stop reaction; discard final/full product.

PREPARATION OF EXPERIMENTAL INK BLUE 452

(1) EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

(2) RAW MATERIALS:

	Weight (g)	Lot #
Aniline 5	125.2 g	10000-65
p-DMAB	31.0 g	B4090
muriatic acid	29.7 g	W2811
urea	4.7 g	G6819
ammonium meta vanadate	0.14 g	A382
water	51.0 g	—
hydrogen peroxide	45.0 g	W2894

CONDENSATION

(3) PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0 g water
4 Charge muriatic acid
5 Charge urea and equip flask with a condenser
6 Begin heating reaction to 95-100°C
7 Continue heating at 95-100°C for 3 hours

*CHANGE IN PROCEDURE FROM #61, 62

OXIDATION

(4) PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating @ 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture cautiously, watching for ENDO/EXOTHERM cycle
5 Heat at 98°C for 45 minutes

((CONTINUE TO PAGE 10022-65))

Date July 17, 1996 Signature Gloria G. Wickham
Date July 23, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-64

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DU-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (sw) = 0.1223g
Wavelength (λ) = 590.0
absorbancy (A) = 1.0424
color value = (A/sw) x 5
color value = (1.0424/0.1223) x 5
color value = 42.62 = 43.0

7 Stop reaction; discard full product.

⟹ AVERAGE TO LOW COLOR VALUE

Date July 17, 1996
Date July 23, 1996

Signature Gloria G. Dickens
Witness [illegible]

SEPARATION TESTING ON INK YELLOW 850

① EQUIPMENT: 4 vials with lids

② RAW MATERIALS:

	Lot #
Ink yellow 850	X1004
Thinned Solution Resinate	—
Siegwerk Uncoated Yellow Line Ink	SAMPLE
toluene	—
dodecyl sulfate; sodium salt 70%	00603TG
sodium stearate	—
Syn Fac 8145	9914-18
Syn Fac 8118	—

③ PROCEDURE: 1 Prepare 4 inks in the following corresponding formulations
2 Seal vials and let stand in flammable cabinet
3 Test at a later date

④ FORMULATIONS:

(A)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
dodecyl sulfate: 0.10g

(B)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Sodium stearate: 0.10g

(C)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Syn Fac 8145: 0.10g

(D)

Siegwerk Line Ink: 8.0g
TSR: 2.0g
Ink Yellow 850: 0.24g
toluene: 17.0g
Syn Fac 8118: 0.10g

Date July 17, 1996 Signature Gloria G. Wichern
Date July 23, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1- 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles
Aniline 5	125.2g	10000-65	
p-DMAB	34.1g	B4090	0.228M
muriatic acid	29.7g	W2811	
urea	4.7g	G6819	
ammonium meta vanadate	0.14g	A382	
water	51.0g	—	
hydrogen peroxide	45.0g	W2894	

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0g water
4 Charge muriatic acid
5 Charge urea and equip flask with a condenser
6 Begin heating reaction to 95-100°C
7 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture slowly
5 Heat at 105°C for 45 minutes

((CONTINUE TO PAGE 10022-69))

Date July 18, 1996 Signature Gloria G. Wickens
Date July 23, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles
Aniline 5	137.72g	10000-65	0.44M
p-DMAB	31.0g	B4090	
muriatic acid	29.7g	W2811	
urea	4.7g	G6819	
ammonium meta vanadate	0.14g	A382	
water	51.0g	—	
hydrogen peroxide	45.0g	W2894	

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0g water
4 Charge muriatic acid
5 Charge urea and equip flask with a condenser
6 Begin heating reaction to 95-100°C
7 Continue heating at 95-100°C for 3 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2 / H_2O mixture slowly
5 Heat at 105°C for 45 minutes

((CONTINUE TO PAGE 10022-69))

Date July 18, 1996 Signature Gloria G. Wickham
Date July 23, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-67

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (SW) = 0.1302 g
Wavelength (λ) = 590.0
absorbancy (A) = 0.9422
color value = (A/SW) x 5
color value = (0.9422/0.1302) x 5
color value = 36.18

7 Stop reaction; discard full product.

CONTINUED FROM PAGE 10022-68

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (SW) = 0.1351 g
Wavelength (λ) = 590.0
absorbancy (A) = 1.1214
color value = (A/SW) x 5
color value = (1.1214/0.1351) x 5
color value = 41.50 = 42

7 Stop reaction; discard full product.

Date July 18, 1996 Signature Gloria G. Wickham
Date July 23, 1996 Witness

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1 - 500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #	moles
Aniline 5	125.2g	10000-65	
p-DMAB	31.0g	B4090	
Muriatic acid	29.7g	W2811	
Urea	4.7g	G6819	
ammonium meta vanadate	0.14g	A382	
Water	65.5g	—	
hydrogen peroxide	59.5g	W2894	0.61M

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge 6.0g water
4 Charge muriatic acid
5 Charge urea and equip flask with a condenser
6 Begin heating reaction to 105°C
7 Continue heating at 105°C for 4 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 1.5 hours
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture slowly
5 Heat at 105°C for 45 minutes

((CONTINUED TO PAGE 10022-72))

Date July 19, 1996 Signature Gloria J. Wichum
Date July 23, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1 - 500mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser

② RAW MATERIALS:

	Weight (g)	Lot #
Aniline 5	125.2g	10000-65
p-DMAB	31.0g	B4090
muriatic acid	29.7g	W2811
urea	4.7g	G6819
ammonium meta vanadate	0.14g	A382
water	45.0g	—
hydrogen peroxide	45.0g	W2894

CONDENSATION

③ PROCEDURE:
1 Charge aniline 5 and begin stirring
2 Charge p-DMAB
3 Charge muriatic acid
4 Charge urea and equip flask with a condenser
5 Begin heating reaction at/to 105°C
6 Continue heating reaction at 105°C for 4 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 1.5 hours
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2/H_2O mixture slowly
5 Heat at 105°C for 45 minutes

((CONTINUE TO PAGE 10022-72))

Date July 19, 1996 Signature Gloria J. Wickens
Date July 23, 1996 Witness [illegible]

CONTINUED FROM PAGE 10022-70

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (sw) = 0.1284g
Wavelength (λ) = 590.0
absorbancy (A) = 0.9985
color value = (A/sw) x 5
Color value = (0.9985/0.1284) x 5
color value = 38.88 = 39.0

7 Stop reaction; discard full product.

CONTINUED FROM PAGE 10022-71

((PREPARATION OF EXPERIMENTAL INK BLUE 452))

6 Run color value, DV-478

TEST DATA AND OBSERVATIONS:

6A) Sample weight (sw) = 0.1476g
Wavelength (λ) = 591.0
absorbancy (A) = 1.4854
color value = (A/sw) x 5
Color value = (1.4854/0.1476) x 5
color value = 50.32

7 Stop reaction; discard full product.

(5) CONCLUSIONS: Excellent color value - reaction without additional beginning condensation water works very well (or appears to). Could also have been other factors: LONG COND. TIME, LONGER OXIDATION CATALYST REACTION TIME. Reccommendation is to run reaction while controlling "TIME" factors.

Date July 19, 1996 Signature Gloria G. Wickens

Date July 23, 1996 Witness [illegible]

PREPARATION OF EXPERIMENTAL INK BLUE 452

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller ~~mc~~ gl 7/23/96
mechanical agitator
condenser

② RAW MATERIALS:

	weight (g)	Lot #
Aniline S	125.2g	10000-65
p-DMAB	31.0g	B4090
muriatic acid	29.7g	W2811
urea	4.7g	G6819
water	45.0g	—
hydrogen peroxide	45.0g	W2894
ammonium meta vanadate	0.14g	A382

CONDENSATION

③ PROCEDURE:
1 Charge aniline S and begin stirring
2 Charge p-DMAB
3 Charge muriatic acid
4 Charge urea and equip flask with a condenser
5 Begin heating reaction to 105°C
6 Continue heating at 105°C for 6 hours

OXIDATION

④ PROCEDURE:
1 Reduce temperature to 93°C
2 Charge amv and continue heating at 93°C for 15 minutes
3 Mix H_2O_2 and remaining H_2O together thoroughly
4 Keeping temperature range between 100-108°C, charge H_2O_2 / H_2O mixture slowly
5 Heat at 105°C for 45 minutes
6 Run color value, DV-478: color value = (A/sw) x 5
Sample weight (sw) = 0.1392g
wavelength (λ) = 591.0
absorbancy (A) = 1.3318

COLOR VALUE = (1.3318/0.1392) x 5
= 47.84 = 48

Date July 23, 1996 Signature Gloria G. Dickens
Date July 23, 1996 Witness [illegible]

7 Stop reaction; discard full product.

PREPARATION OF EXPERIMENTAL INK BLUE 1892

① EQUIPMENT: 1-500 mL three-neck, roundbottom flask
thermal controller
mechanical agitator
condenser
automatic K-Proofer

② RAW MATERIALS:

	Weight (g)	Lot #
Uncut Ink Blue 92	51.35 g	G1011
ODSA	38.5 g	TMX-4
1-methylimidazole	1.25 g	K9037
ethanolamine	24.4 g	0301SLN
distilled water	131.0 g	—

③ PROCEDURE:
1 Charge uncut ink blue 92 and begin stirring
2 Charge ODSA
3 Charge 1-methylimidazole and equip flask with a condenser
4 Begin heating reaction to 100°C
5 Continue heating at 100°C for 4 hours
6 Run IR

TEST DATA AND OBSERVATIONS:
6A) An anhydride peak appears at 1830 cm^{-1}, indicating reaction is incomplete - ODSA has not fully reacted.
7 Continue heating at 100°C for 1 hour
8 Run IR

TEST DATA AND OBSERVATIONS:
8A) A small anhydride peak still appears at 1830 cm^{-1}
9 Reduce temperature to 50°C
10 Thoroughly mix ethanolamine and water
11 Charge ethanolamine/H_2O mixture to reaction flask (66 g H_2O)
12 Continue stirring until thoroughly mixed - until product is a homogeneous mixture
13 Charge 65.0 g of distilled H_2O and mix well
14 Run pH, DV-5

Date July 23, 1996 Signature Gloria G. Wickham
Date July 23, 1996 Witness [illegible]

((CONTINUE TO PAGE 10022-75))

CONTINUED FROM PAGE 10022-73

((PREPARATION OF EXP. INK BLUE 1892))

TEST DATA AND OBSERVATIONS:

14A) pH = 10.59

⟹ acidify with 93% H_2SO_4, Lot # B4206

⟹ after acid, run pH

pH = 1.35

⟹ separate layers of H_2O and colorant by decanting H_2O out of beaker

15 Stop reaction; retain full sample product (retain entire ink product layer)

Date July 23, 1996 Signature Gloria G. Dickens

Date July 23, 1996 Witness [illegible]