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Plastics

I.G. FARBENINDUSTRIE A.G.

ELBERFELD & LEVERKUSEN, 27 April 1945

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PLASTICS

I.G. FARBENINDUSTRIE AG  
ELBERFELD AND LEVERKUSEN, GERMANY

27 April 1945

Reported by

FRANCIS J. CURTIS  
CWS, HQ ETOUSA

18 May 1945

CIOS Black List Item 22  
Miscellaneous Chemicals

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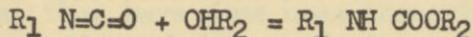
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## PLASTICS

I.G. FARBENINDUSTRIE AG  
LEVERKUSEN AND ELBERFELD, GERMANY

1. DIISOCYANATES AND POLYURETHANES - Leverkusena. GENERAL DATA

The fundamental reaction of an isocyanate with an alcohol to form a urethane was discovered by Wurtz in 1848:



The new development consists in reacting di- or poly-isocyanates with polyhydroxy compounds. In general, di- compounds form linear polyurethanes while tri- and higher make cross-linked products. Obviously, almost an infinite variety of properties can be produced by suitably varying constituents. Since the linkage is produced by transfer of the hydrogen of the hydroxyl to the nitrogen of the isocyanate, no water is split off.

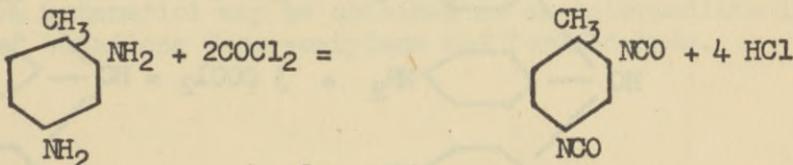
In general, the di-isocyanates react with compounds having a hydrogen atom replaceable by caustic soda, e.g., the groups OH, NH<sub>2</sub>, SO<sub>2</sub>NH<sub>2</sub>, COOH, etc. They, therefore, have uses by themselves outside the manufacture of polyurethanes.

The fundamental reaction was given to the U. S. in 1938; and the subject has been covered by German patents from 1939 on. British, Italian, and French patents have been published.

b. DI- AND TRI-ISOCYANATES - MANUFACTURE AND USE

The trade names for these products are Desmodur and Desmosit.

(1) Desmodur T (Tolylene di-isocyanate) M.W. 174

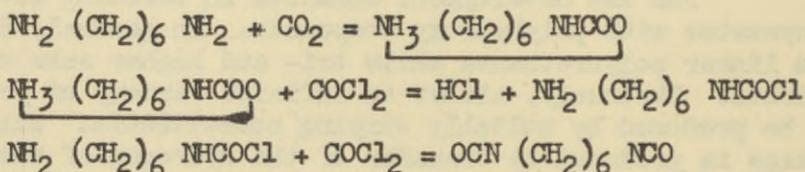


-3-

The tolylenediamine is a mixture of 60% of 2,4 and 40% 2,6 tolylenediamines, produced by nitration of o-nitrotoluene followed by reduction. A 33% solution of phosgene in o-dichlorobenzene is prepared at 0°C and a 33% solution of tolylenediamine in the same solvent at 110°C is run into the cold phosgene solution keeping the temperature under 50°C. The mixture is blown by nitrogen into more ortho-oil, heated to 160°C and phosgene passed in to completion as determined by formation of a clear solution. A rough take-over distillation is followed by a final distillation under 20 mm vacuum with a yield of 73% of theory on amine, 77-80% on phosgene and a loss of ortho oil of 5% of that used. The mixture of COCl<sub>2</sub> and HCl given off is scrubbed with ortho oil to dissolve COCl<sub>2</sub> which is re-used. Excess amine is recovered from the residue by cooking with caustic soda solution.

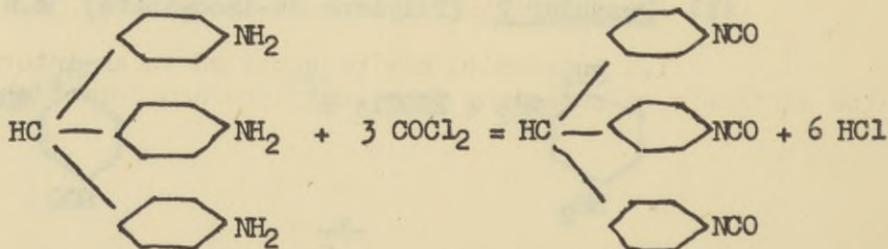
Production - 20 tons/mo.

(2) Desmodur H (Hexamethylene di-isocyanate)



The formation of isocyanates from aliphatic diamines must be carried out in two steps with the intermediate formation of a carbamate. 1 part hexamethylenediamine in 6 parts ortho oil is saturated at 80°C with CO<sub>2</sub>, and cooled to 0° to minus 5°C. A solution of phosgene in three parts ortho oil, equivalent to 120 mol. % of that required for formation of carbamic acid chloride, is added at a rate to keep a temperature of 0-5°C. The batch is then gradually heated to 90°C and phosgene started in, the temperature finally reaching 155°C. The process is completed when the product gives a clear solution. Purification of the crude is similar to Desmodur T above with a crude yield of 85-90% theory. Production was 6-8 tons/mo.

(3) Desmodur R



Delta R Base

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A 40% solution of phosgene is prepared in o-dichlorobenzene at 5°C and likewise a 22% solution Delta R base in ortho oil and cooled to 5°C. The latter is run into the former with the addition of phosgene, temperature rising to 70°C, then further phosgene is added at 105°C with hourly tests for complete solubility. CO<sub>2</sub> or N<sub>2</sub> are passed through to remove excess COCl<sub>2</sub>, the batch is filtered and chlorobenzene distilled off and the remainder made into a 25% solution with methylene chloride.

Production - 2 tons/mo.

(4) Desmosit C

Desmosit C is 1 - chlorophenylene 2,4-di-isocyanate.

(5) Uses of Di- and Tri-isocyanates as such

While the chief use of di- and tri-isocyanates is for making polyurethanes, they have some uses themselves because of their properties.

Since skins have NH<sub>2</sub> groups with which the di-isocyanates can combine, they have been employed in place of natural tannins for tanning, needing 7 hours instead of the usual 80 - 100 hrs. Likewise acetate silk now melts if not carefully ironed whereas after treatment with di-isocyanates no further melting occurs.

Desmodur R is especially applicable to rubber and has been used to fasten rubber to tank treads so well that the rubber breaks before the joint between the rubber and the steel. Though not yet used widely in industrial practice, it provides the best method the Germans have found for sticking synthetic rubber to cords for any size tires. 2% on the rubber is used, and it may be either mixed with the rubber or impregnated on the cords. No natural rubber is necessary.

c. POLYHYDROXY ETC. COMPOUNDS

The chief polyhydroxy compounds now in use for combining with the di-isocyanates to form polyurethanes are:

(1) 1,4 butanediol

1,4 butanediol may be obtained as an intermediate in the synthesis of butadiene from acetylene and formaldehyde.

60

(2) Desmophen 900 ("Binde mittel 28")

3 mols adipic acid and 4.2 mols of trimethylol propane are heated in presence of  $\text{CO}_2$  at  $200^\circ\text{C}$ , temperature being held until an acid number of less than 2 results, while water distills off. Product is soluble in cold alcohol and in acetone. Yield 2.08 kg per 1 kg of adipic acid.

"Bindemittel 28 HH" is the corresponding ester of adipic acid and trimethylolpropane H,  $\text{CH}_3\text{CHOHCH}_2\text{CHOHCH}_3$

(3) Desmophen 1100

Desmophen 1100 is the polyester from 3 mols. adipic acid, 2 mols 1,3 butanediol, 2 mols trimethylolpropane.

(4) Desmophen 1200

Desmophen 1200 is the polyester from 3 mols adipic acid, 3 mols 1,4 butanediol and 1 mol trimethylolpropane.

d. LINEAR POLYMERS(1) Igamid U

1.004 mols of 1,6 hexamethylene di-isocyanate (Desmodur H) is condensed with 1 mol of 1,4 butanediol in a mixture of 90 pts. monochlorobenzene and 10 pts. o-dichlorobenzene. Quality of the components is of utmost importance. The di-isocyanate must be 99.5 - 100% by NCO content, with chlorine compounds as low as possible, e.g., not more than 0.05-0.1% from o-dichlorobenzene, 0.04% from chlorohexaneisocyanate and 0.004% from acid chloride. 1,4 butanediol must have a melting point of  $19-19.2^\circ\text{C}$ , hydroxyl number 37 and water content of not more than 0.05%.

Process

132 liters chlorobenzene (dry distilled) and 15 liters dichlorobenzene are placed in a stainless steel reaction kettle and 13.776 kg of the butanediol added. After warming to  $60-70^\circ$ , 25.428 kg Desmodur H is added slowly. The temperature rises to the boiling point and a reflux condenser and cooling are used. Although the reaction is ended in 5-6 hours, heating to boiling is continued 2-3 hours longer.

The product is filtered, washed with chlorobenzene and the solvent removed with steam. The polyurethane is dried at not more than 30 mm absolute vacuum and 65°C. The yield is 39 kg.

Uses

Igamid U does not combine with ordinary commercial plasticizers but can be injection and compression molded. The melting point is 184°C with a short period between melting and low viscosity. Products have good mechanical strength and electrical properties and high resistance to solvents and oils. Fibers are produced by hot melt, spinning and stretching. Bases for leather are made by sheeting. Used for bristles (Perlon U), it has a lower melting point than Nylon and therefore is easier to form, has a better resistance against acids, is harder, has better electrical properties, and has a maximum water absorption of 2% compared to 8% for Nylon.

Molecular weight - 10,000.

(2) Polystal

Polystals are resinous adhesives made from Desmodur T and Desmophen 1200. They are used for plywood in the normal way by pressing at 100°C for 1/2 hour at the usual pressures. They can be operated at room temperature but in that case 5 hours is required. These adhesives have the advantage of being more elastic than phenolics.

e. CROSS LINKED POLYMERS

When the di-isocyanates are reacted with alcohols or other compounds with more than 2 functional groups, cross-linked products are obtained which are infusible. By mixing the constituents with basic catalysts just before using, these can be employed as lacquers for wood, rubber, leather, paper, etc. with times of polymerization from 1 to 24 hours. Coatings are produced of great weather resistance, high gloss, good electrical properties and very impervious to gases.

Furthermore, when alkyd resins are condensed with extra glycol which is then combined with di-isocyanate, a wide range of improved properties can be obtained.

8a

Paper for Protection Against Mustard Gas

A paper plant near Dresden was equipped to impregnate paper with Desmodur-Desmophen combinations for the German Army, though only 50,000 sq m were produced. Samples of this paper have been secured for testing, as it was claimed to be the best protection the Germans had found against mustard gas.

Two formulae were used:

1

Ground coat	26	pts (wt)	Desmophen 1100
	2.6	"	Desmosit C
	3.9	"	Desmodur T
	67.5	"	Ethyl acetate
Top coat	27	"	Desmophen 1100
	2.7	"	Desmosit C
	8.1	"	Desmodur T
	62.2	"	Ethyl acetate

2

Ground coat	26.5	pts (wt)	Desmophen 1200
	6.5	"	Desmodur T
	67.5	"	Ethyl acetate
Top coat	27	"	Desmophen 1100
	10.8	"	Desmodur T
	62.2	"	Ethyl acetate

In the second the somewhat softer 1200 is used for the ground coat and the more poorly dissolving C replaced by T. The mixture must be used within a day.

For impregnation, crepe paper is passed continuously through the ground coat and is dried 5 minutes at 120°-130°C. The top coat is then put on in a similar manner.

Drs. Bayer, and Boehme were interviewed.

2. ETHYL CELLULOSE - Elberfeld

Production - 7 tons/mo.

Bill of Materials

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For 56 kg product:

- 628 kg 50% caustic soda
- 45 kg Beechwood cellulose NW 10 from Wolfen
- 220 kg ethyl chloride

Process

Caustic soda and cellulose are added during one hour to a nickel Werner-Pfleiderer mixer and kneaded for 3.5 hours without cooling at a temperature of 35 - 40°C.

The alkali cellulose is then transferred to a nickel plated 1300 liter autoclave and air removed. 220 kg ethyl chloride is added at 2-3 atmospheres pressure, stirring is started and temperature raised during 1 to 1.5 hours to 60°C at 7 atmospheres. The steam is turned off and the temperature rises to 80°C; when the temperature falls, heat is turned on until 120-130°C is reached with final heating at 140°C for 1 hour. The total heating period is 12 hours and highest pressure 20-22 atm. during ethylation. 120 liters of ether-alcohol is distilled off during 2 hours. The autoclave is cooled to 90°C and contents blown to a suction filter.

The product is washed on the filter 3 times, acidulated with 1 liter HCl in an enameled kettle for 2 hours and then given 6 washes with softened water. It is dried at 90°C in a tray drier.

The ethoxyl content is 47-48%.

Four viscosities are made and tested by an IG test in which a 5% solution in 9 parts benzene - 1 part alcohol is placed in a 50 cm high tube and the time taken for rise of an air bubble. The test is said to be independent of the size of the air bubble.

The following grades were made:

Very low	10 sec.
Low	14 sec.
Middle	20 sec.
High	30 sec.

Drs. Lutter and Rietz were interviewed.

10 a

3. BENZYL CELLULOSE - Elberfeld

Production - 10-15 tons/mo.

Bill of MaterialsFor 130 kg:

306 kg 50% caustic soda  
 150 kg regenerated 18% caustic soda  
 79 kg Beech cellulose  
 330 kg benzyl chloride (from Verdingers)

All the caustic soda and the cellulose are added to an 800 liter nickel Werner-Pfleiderer mixer and kneaded 6 hours with cooling, keeping temperature below 30-35°C.

The alkali cellulose is transferred to a 1000 liter nickel autoclave, the benzyl chloride added, and heated to 70°C; steam is then shut off and temperature rises to 105°C. Stirring is started 1/2 hour after heating. At 106°C, cooling is begun and temperature must not go above 113°C. Total heating period is 8 hours.

The batch is transferred to a 1000 liter nickel kettle and caustic drained off. Two batches are united and put into a Nirosta (V<sub>2</sub>S) kneader and washed with water 4 times and kneaded 13 times with 90% methanol. During the 7th to 10th wash the product becomes granulated and must have 3 more washes. It is disintegrated and put into a nickel kettle with salt in the bottom and given 15 washes with methanol for 1/2 hour each. It is then suction filtered and washed with cold water 5 times and with hot water 10 times in an enameled kettle. It is then dried in a chamber and again washed with hot water 10 times at 60°C, cooled, centrifuged, ground and dried.

The dibenzyl ether and benzyl alcohol are recovered in the methanol which is distilled. From a double batch are recovered 135-140 kg. The mixed ether-alcohol proportions were not known exactly.

Benzyl content - 51%

Viscosity grades -

5% solution in benzyl alcohol by IG method (see under ethyl cellulose)

High	22 sec.
Middle	16 sec.
Low	12 sec.
Very low	10 sec.

Same solubility tests as ethyl cellulose with toluol and alcohol-benzene (1:9)

Drs. Lutter and Rietz were interviewed.

11a

4. CELLAPRIT (cellulose acetate 50% water wet) - Elberfeld

Production - 20 tons/mo.

Bill of materials

For 790 kg:

- 167 liters water
- 272 kg 50% caustic soda
- 130 kg Beech cellulose
- 162 kg monochloroacetic acid in 65 liters water

The water and caustic are mixed in a 800 liter nickel Werner-Pfleiderer mixer, cooled with brine to 20°C, and during 3 hours the cellulose is added and, while cooling, kneaded for 2 hours.

Monochloroacetic acid in water is added slowly to the kneader, being careful to clean off gummy deposits at least 3 times. The addition takes 16 hours. Brine is cut off and kneading continues for 4 hours.

The product is used as a stiffening and sizing agent for cotton textiles.

Drs. Lutter and Rietz were interviewed.

5. LIST OF DOCUMENTS

The following documents have been sent to MIRS for filing:

- a. Folder No. III I.G. Leverkusen  
"Method of Preparation of a Number of Products."
- b. Document No. XIII I.G. Leverkusen  
"Theoretical Explanation of Use of Desmophen - Desmodur for Coatings."





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