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DICARBOXYLIC ACID ANHYDRIDE CONDENSATION
WITH COMPOUNDS CONTAINING ACTIVE METHYLENE GROUPS.
IV. SOME 4-NITROPHTHALIC ANHYDRIDE CONDENSATION REACTIONS

V. Oskaja and J. Rotberg

Translation of "Kondensatsiya angidridov dikarbonovykh kislot s soyedineniyami, soderzhashchimi aktivnyye metilenovyye gruppy IV. Nekotoryye reaktsii kondensatsii 4-nitroftalevogo angidrida," Latvijas PSR Zinatnu Akademijas Vestis, Kimijas Serija, LZAKAM, No. 1, 1967, pp. 63-69.

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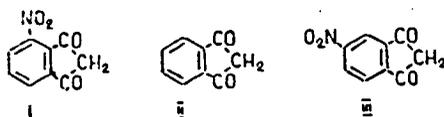
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| 16. Abstract 1. By 4-nitrophthalic anhydride condensation with acetoacetate in acetic anhydride and triethylamine solution with subsequent breakdown of the intermediate condensation product, 5-nitroindanedione-1,3 was obtained. 2. 4-Nitrophthalic anhydride with acetic anhydride, according to reaction conditions, may yield two products: in the presence of potassium acetate and at high temperatures 4-(or 5)-nitro-2-acetylbenzoic acid is formed; in the presence of triethylamine and at room temperature 5-(or 6)-nitrophthalic acetic acid is isolated. 3. 4-Nitrophthalic anhydride and malonic acid in pyridine solution according to temperature yield either 5-(or 6)-nitrophthalic acetic acid or 4-(or 5)-nitro-2-acetylbenzoic acid. | | |
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DICARBOXYLIC ACID ANHYDRIDE CONDENSATION
WITH COMPOUNDS CONTAINING ACTIVE METHYLENE GROUPS.
IV. SOME 4-NITROPHthalic ANHYDRIDE CONDENSATION REACTIONS

V. Oskaja and J. Rotberg

4-Nitroindandione-1,3 (I) [1,2] readily yields 2,4-dinitroindandione-1,3 and other derivatives. We set out to prepare yet another nitro analog of indandione-1,3 (II)--5-nitroindandione-1,3 (III), whose synthesis is not described in the literature [3].

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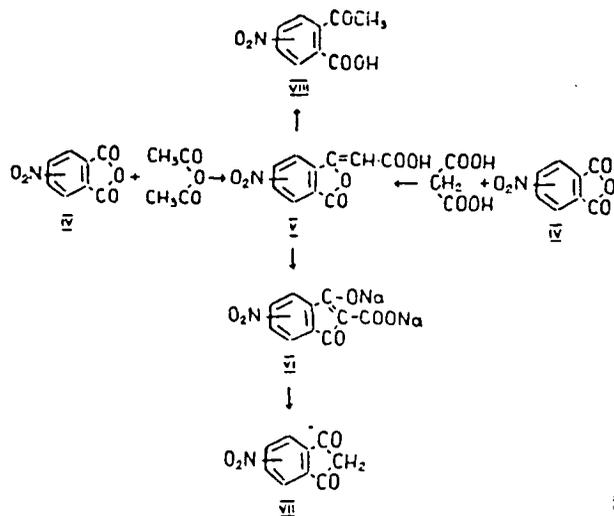


5-Nitroindandione-1,3, just like 4-nitroindandione-1,3 [1], cannot be made by the main method of synthesizing unsaturated indandione-1,3, that is, by the method of ester condensation of the esters of phthalic and acetic acids [3]. The diethyl ester of 4-nitrophthalic acid with the ethyl ester of acetic acid in the presence of sodium alcoholate or sodium amide yields resinous, dark-colored noncrystalline substances.

The inability of the esters of 4-nitrophthalic acid to enter into ester condensation was also noted in the literature [5].

Initially, 4-nitroindandione-1,3 (VII) was prepared from 4- (or 7-)nitrophthalic acid (V), which was rearranged into the disodium salt of 4-nitroindandione-1,3-carboxylic-2 acid (VI) and the latter was decomposed with mineral acid into 4-nitroindandione-1,3 (VII) [1].

* Numbers in the margin indicate pagination in the foreign text.

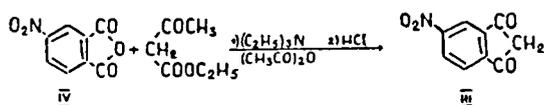


In attempts to prepare the corresponding nitrophthalylacetic acid (V) from 4-nitrophthalic anhydride (IV) and acetic anhydride by the Perkin's reaction in the presence of potassium acetate, we observed the formation of 4-(or 5-)-nitro-2-acetylbenzoic acid (VIII). This same acid (VIII) was obtained from the condensation of 4-nitrophthalic anhydride (IV) with malonic acid in a pyridine solution, if the reaction was carried out at the boiling point of a water bath. When 4-nitrophthalic anhydride was condensed at room temperature with malonic acid in the pyridine solution or with acetic anhydride in the presence of triethylamine as a catalyst, 5-(or 6-)-nitrophthalylacetic acid (V) was formed. Obviously, 4-(or 5-)-nitro-2-acetylbenzoic acid (VIII) is the decomposition product of 5-(or 6-)nitrophthalic acid (X).

From 4-(or 5-)-nitro-2-acetylbenzoic acid the following were prepared: its methyl ester, the oxime and 2,4-dinitrophenylhydrazone, and 5-(or 6-)-nitrophthalylacetic acid (V) was converted to 5-nitroindandione-1,3 (VII). To confirm the structure of the products obtained, their infrared spectra were recorded in the region

obtained, their infrared spectra were recorded in the region of absorption by carbonyl groups and were compared with the infrared spectra of already known compounds (see Table 1).

Since the yield of 5-nitroindandione-1,3 by this method was found to be unsatisfactory, other possibilities for its synthesis were studied. 4-Nitrophthalic anhydride was condensed in a solution of acetic anhydride and triethylamine with the diethyl ester of malonic acid and the ethyl ester of acetoacetic acid. With the malonic ester a colorless product forms; according to its infrared spectral data, this product can be given the structure of the diethyl ester of 5-(or 6-)-nitrophthalylmalonic acid



With the acetoacetic ester, 4-nitrophthalic anhydride (IV) in a solution of acetic anhydride and triethylamine yields an unstable intermediate product, which cannot be recrystallized, but which, on reacting with hydrochloric acid, yields 5-nitroindandione-1,3 (III) with a yield of 30.8%, based on the original 4-nitrophthalic anhydride. From 5-nitroindandione-1,3 derivatives, 2-benzal-5-nitroindandione-1,3 was prepared; its infrared spectrum is analogous to that for 2-benzal-4-nitroindandione-1,3.

Experimental Part¹

1. Condensation of 4-Nitrophthalic Anhydride with Acetic Anhydride

a. In the presence of potassium acetate. 4.8 g (0.025 mole) of 4-nitrophthalic anhydride and 1 g (0.01 mole) freshly dewatered potassium acetate (made by melting) in a small flask were covered with 10 g (0.1 mole) acetic anhydride. The flask was connected to a reflux condenser (a calcium chloride tube) and immersed in

¹The woman student M.E. Vitola participated in the experimental work.

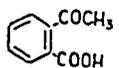
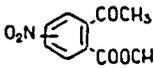
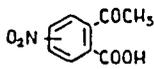
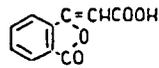
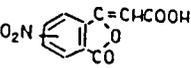
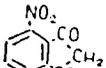
an oil bath, whose temperature was kept in the range 145-150°. The flask was heated for 90 min, after which the strongly darkening reaction mixture cooled somewhat, was diluted with water, and decolorized with charcoal. The solution was distilled off in a water bath, filtered, acidified with hydrochloric acid, and allowed to stand for a night. Brown crystals were isolated, which melted at 137-139°. The weight was 0.87 g (16.7%). After repeated recrystallization from water, colorless needles of 4-(or 5-)-nitro-2-acetylbenzoic acid had a melting point of 140-141°. They are readily soluble in water and in organic solvents. From a sodium bicarbonate solution they were extracted with acetic acid.

The following data were found, %: C, 51.81, 52.00; H, 3.56, 3.53; N, 6.96, 6.67. $C_9H_7O_5N$ (VIII). Calculated, %: C, 51.70; H, 3.37; N, 6.70.

b. In the presence of triethylamine. 9.6 g (0.05 mole) of 4-nitrophthalic anhydride was dissolved with heating in 20 g (0.2 mole) acetic anhydride; to the solution was added 5.0 g (0.05 mole) triethylamine; the flask was covered with a calcium chloride tube and allowed to stand for a night. The solution turned dark brown. The next day, the reaction mixture was poured onto 100 g ice and 30 ml concentrated hydrochloric acid. Intense cooling of the solution took place, and the separating oil solidified after some time. After decomposition of the acetic anhydride, a heterogeneous yellow precipitate was washed with water and crystallized from acetic acid. The yield was 1.85 g (15.8%). M.p.: 236-238°. Repeated crystallization from acetic acid yielded faintly yellowish crystals, m.p. 243-245°. From a sodium bicarbonate solution, they were isolated with acetic acid and the diluted solution was rapidly decolorized with potassium permanganate.

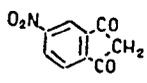
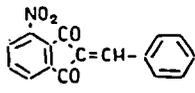
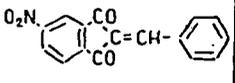
Found, %: C, 51.15; 51.48; H, 2.14, 2.32; N, 5.93, 5.86.
 $C_{10}H_5O_6N$ (V). Calculated, %: C, 51.08; H, 2.14; N, 5.96.

TABLE 1. INFRARED RED ABSORPTION SPECTRA OF PRODUCTS OBTAINED AND CERTAIN KNOWN COMPOUNDS

| (1) N | (2) Формула и наименование соединения | (3) Частота валентных колебаний в см ⁻¹ | | | | | | (8) Литературные данные |
|----------|--|---|---|---|-------------------------|---------------------------------|--------------------------|----------------------------|
| | | $\nu_{\text{C=O}}$ фталиид-го кольца (4) 3 | $\nu_{\text{C=O}}$ кетонная (5) 4 | $\nu_{\text{C=O}}$ карбок или (6) 5 | $\nu_{\text{C=C}}$ 6 | $\nu_{\text{аромат.}}$ (7) 7 | ν_{NO_2} 8 | |
| 1 |  o-Ацетилбензойная кислота (9) | — | 1721 (92) | 1705 (83) | — | 1613 (42) 1604 (41) | — | |
| 2 |  4-(Или 5-)-нитро-2-ацетилбензойная кислота (10) | — | 1741 (83) | 1705 (40) | — | 1617 (26) | 1529 (61) | |
| 3 |  Метилловый эфир 4-(или 5-)-нитро-2-ацетилбензойной кислоты (11) | — | 1720 (86) | | — | 1607 (56) | 1525 (71) | |
| 4 |  Фталилуксусная кислота (12) | 1800 | — | 1705 | | | | [6] |
| 5 |  5-(Или 6-)-нитрофталилуксусная кислота (13) | 1800 (71) | — | 1686 (75) | 1653 (77) | 1614 (57) | 1527 (68) | |
| 6 |  4-Нитроиндан-дион-1,3 (14) | | 1741 (60) 1709 (89) | | | 1600 (53) | 1535 (82) | [2] |

[Key to table, on next page]

TABLE 1 [Concluded]

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 |
|---|---|---|------------------------|---|-----------|------------------------|-----------|---|
| 7 |  5-Нитроиндан- дин-1,3 (15) | | 1752 (49) 1711 (85) | | | 1607 (54) 1582 (44) | 1527 (68) | |
| 8 |  2-Бензаль-4-нитро- индандин-1,3 (16) | | 1729 (55) 1688 (84) | | 1654 (64) | 1591 (84) | 1528 (90) | |
| 9 |  2-Бензаль-5-нитро- индандин-1,3 (17) | | 1725 (26) 1685 (63) | | 1653 (40) | 1607 (42) 1588 (44) | 1529 (51) | |

Remark. All the spectra were recorded for suspensions of solid substances in paraffin oil. Intensities in absorption percentages are parenthesized.

Key: 1. Number

2. Formula and name of compound

3. Frequency of valency oscillations in cm^{-1}

4. of phthalic ring

5. ketonic

6. of carbonyl

7. ν_{aromatic}

8. Literature data

9. o-Acetylbenzoic acid

10. 4-(or 5-)-nitro-2-acetylbenzoic acid

11. Methyl ester of 4-(or 5-)-nitro-2-acetylbenzoic acid

12. Phthalylacetic acid

13. 5-(or 6-)-nitrophthalylacetic acid

14. 4-Nitroindandione-1,3

15. 5-Nitroindandione-1,3

16. 2-Benzal-4-nitroindandione-1,3

17. 2-Benzal-5-nitroindandione-1,3

2. Condensation of 4-Nitrophthalic Anhydride with Malonic Acid

a. In acetic acid. 4.8 g (0.025 mole) of 4-nitrophthalic anhydride, 3.1 g (0.030 mole) of malonic acid, 5 drops of piperidine, and 10 ml of glacial acetic acid in a flask with a reflex condenser were heated for 4 h on a water bath. The reaction mixture was diluted with an equal volume of water and acidified with 5 ml of concentrated hydrochloric acid. On standing, a white crystalline precipitate weighing from 2 to 2.5 g separated out. M.p. 138-140°. After recrystallization from water, fine colorless crystals were obtained, m.p. 140-141°. The substance did not depress the melting point with 4-(or 5-)-nitro-2-acetylbenzoic acid made from 4-nitrophthalic and acetic anhydrides in the presence of potassium acetate.

b. In pyridine at room temperature. In an erlenmeyer flask equipped with a plug and a gas vent tube, 19.3 g (0.1 mole) dry 4-nitrophthalic anhydride was dissolved in 25 ml of dried and freshly distilled pyridine. The solution was cooled to 20° and added to 10.5 g (0.1 mole) dry malonic acid. With mixing of the solution, at once intensive liberation of carbon dioxide gas occurred, and the temperature of the reaction mass rose by 10-12°. The solution was again cooled to 20° and allowed to stand until all the malonic acid had dissolved and the reaction products began to separate out as a precipitate. In 6-7 h, the liberation of carbon dioxide practically halted; the thickened solution was filtered off and the residue on the filter was washed twice with 15 ml ether. After drying, 20-25 g of a light yellow heterogeneous product was obtained, with a pyridine-like odor and melting in the range 85-110°. It was readily soluble in acetone, dioxan, and acetic acid, with more difficulty in alcohols, and is poorly soluble in water, ether, benzene, and chloroform. To prepare free 5-(or 6-)-nitrophthalylacetic acid from the pyridine salt, the reaction product was covered with 50 ml hydrochloric acid (1:1) and stirred. The reaction product was fully dissolved, and only then did 5-(or 6-)-nitrophthalylacetic

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acid began separating out as colorless crystals, whose weight after drying was 10-14 g. The product melted in a wide temperature range. After recrystallization from ethanol, the yield of 5-(or 6-)-nitro-phthalylacetic acid was 4-5.5 g (17-23.4%) and m.p. 237-240°. After recrystallization from alcohol, the m.p. was 243-245°. The product obtained did not depress the melting point with 5-(or 6-)-nitro-phthalylacetic acid prepared from 4-nitrophthalic anhydride and acetic anhydride in the presence of triethylamine.

Found, %: C, 50.89; H, 2.09; N, 5.76. $C_{10}H_5O_6N$ (V). Calculated, %: C, 51.08; H, 2.14; N, 5.96.

c. In pyridine at water bath temperature. 9.6 g (0.05 mole) of 4-nitrophthalic anhydride and 5.2 g (0.05 mole) malonic acid, 15 ml pyridine, and 2 drops piperidine were kept for 1 h in a flask fitted with a reflux condenser on a boiling water bath. At the beginning of heating, the solution foamed strongly owing to the release of carbon dioxide. After cooling, the light-yellow pyridine solution was diluted with an equal volume of water, acidified with hydrochloric acid, and allowed to stand for a night. On the second day, the precipitating colorless crystalline residue was filtered off. The weight of the product obtained was 2.9 g (27.9%) and m.p. 137-139°. After one crystallization from water, m.p. 140-141. The substance did not depress the melting point with 4-(or 5-)-nitro-2-acetylbenzoic acid made by the condensation of 4-nitrophthalic and acetic anhydrides in the presence of potassium acetate.

Found, %: C, 51.90; H, 3.34; N, 6.70. $C_9H_7O_5N$ (VIII). Calculated, %: C, 51.70; H, 3.37; N, 6.70.

3. Derivatives of 4-(or 5-)-Nitro-2-acetylbenzoic Acid

a. Methyl ester. The above-prepared 4-(or 5-)-nitro-2-acetylbenzoic acid was boiled 2 h in methanol in the presence of concentrated sulfuric acid. After recrystallization from methanol, fine white crystals were obtained. M.p. 85°.

Found, %: C, 53.90; H, 4.10; N, 6.38. $C_{10}H_9O_5N$. Calculated, 5: C, 53.81; H, 4.06; N, 6.28.

b. Oxime. 0.2 g of 4-(or 5-)-nitro-2-acetylbenzoic acid, 15 ml of alcohol, 0.5 g of hydroxylamine hydrochloride, and 1.0 g of sodium acetate were heated on a water bath for 1 hr and the solution was then diluted with water. Colorless flakes crystallized out. After recrystallization from diluted alcohol, m.p. 144°.

Found, %: C, 52.23; H, 3.01; N, 13.35. $C_9H_6O_4N_2$. Calculated, %: C, 52.44; H, 2.93; N, 13.59. $C_9H_8O_5N_2$. Calculated, %: C, 48.21; H, 3.60; N, 12.50.

c. 2,4-Dinitrophenylhydrazone. 0.5 g of 4-(or 5-)-nitro-2-acetylbenzoic acid is dissolved in 20 ml of alcohol and to the mixture is added 0.4 g of 2,4-dinitrophenyl-hydrazine dissolved in a mixture of 2 ml concentrated hydrochloric acid, 3 ml water, and 10 ml alcohol. A finely crystalline orange precipitate formed, which after recrystallization from alcohol yielded fine orange crystals. M.p. 225-226°.

Found, %: C, 46.48; H, 2.84; N, 17.62. $C_{15}H_{11}O_8N_5$. Calculated, %: C, 46.27; H, 2.85; N, 17.99.

4. Rearrangement of 5-(or 6-)-Nitrophthalylacetic Acid, Isolation of 5-Nitroindandione-1,3

In an erlenmeyer flask, 11.7 g (0.05 mole) 5-(or 6-)-nitrophthalylacetic acid was dissolved in 450 ml absolute alcohol, cooled, and to it was added a sodium methylate solution made by dissolving 2.6 g (0.11 g-atom) metallic sodium in 75 ml absolute methanol. A dark-red noncrystalline precipitate formed, the disodium salt of 5-nitroindandione-1,3-carboxylic-2 acid. In 2 h the precipitate was filtered off, /68 washed with 96° alcohol and dried. The yield was 6.3-8.2 g. From the salt obtained, a solution that was saturated in water at room temperature was prepared. Intensive release of carbon dioxide took

place, and a light-yellow precipitate of 5-nitroindandione-1,3 was formed, which darkened with time. The precipitate was filtered off and washed with water. After recrystallization from 70° alcohol, to which a few drops of hydrochloric acid had been added, brown-violet crystals with m.p. 113° were obtained. The crystals were soluble in water and very soluble in the usual organic solvents. The color of the solutions was violet.

Found, %: C, 56.60; H, 2.47; N, 7.52. $C_9H_5O_4N$ (VII). Calculated, %: C, 56.55; H, 2.64; N, 7.33.

5. Condensation of 4-Nitrophthalic Anhydride with Diethyl Ester of Malonic Acid

9.7 g (0.05 mole) 4-nitrophthalic anhydride and 8.0 g (0.05 mole) diethyl ester of malonic acid with heating were dissolved in 30 g (0.3 mole) acetic anhydride, cooled to room temperature, and added to 5.0 g (0.05 mole) triethylamine. The solution gradually darkened. The flask was sealed with a calcium chloride tube and allowed to stand at room temperature for 2 days, after which its contents were poured onto 100 g ice and 25 ml concentrated hydrochloric acid. 15.7 g of a white, finely crystalline product was obtained, which was recrystallized from 30 ml acetic acid. The yield of the diethyl ester of 5-(or 6-)-nitrophthalylmalonic acid was 7.9 g (47.3%).

After recrystallization from glacial acetic acid, m.p. 121-123°.

Found, %: C, 54.00; H, 4.06; N, 4.06. $C_{15}H_{13}O_8N$. Calculated, %: C, 53.71; H, 3.91; N, 4.18.

6. Condensation of 4-Nitrophthalic Anhydride with Acetoacetic Ester, Preparation of 5-Nitroindandione-1,3 (III)

9.7 g (0.05 mole) 4-nitrophthalic anhydride and 6.5 g (0.05 mole) acetoacetic ester, with moderate heating, were dissolved in 35 g

(0.35 mole) acetic anhydride and to the still-warm solution 10.1 g (0.1 mole) triethylamine was added. The next day, the dark solution was treated according to the earlier-described method [2]. After the intermediate product was decomposed with an acid, 2.94 g (30.8%) of violet-gray crystals with m.p. 112-113° was obtained; the crystals did not depress the melting point with 5-nitroindandione-1,3 prepared, as above, from 5-(or 6-)-nitrophthalylacetic acid.

7. 2-Benzal-5-nitroindandione-1,3

This compound was obtained on analogy with 2-benzal-4-nitroindandione-1,3 (I). After recrystallization from acetic acid or from alcohol, yellow crystals, m.p. 189°, were obtained.

Found, %: C, 68.79; H, 3.38; N, 5.18. $C_{16}H_9O_4N$. Calculated, %: C, 68.77; H, 3.25; N, 5.02.

Conclusions

1. By 4-nitrophthalic anhydride condensation with acetoacetate in acetic anhydride and triethylamine solution with subsequent breakdown of the intermediate condensation product, 5-nitroindandione-1,3 was obtained.

2. 4-Nitrophthalic anhydride with acetic anhydride, according to reaction conditions, may yield two products: in the presence of potassium acetate and at high temperatures 4-(or 5-)-nitro-2-acetylbenzoic acid is formed; in the presence of triethylamine and at room temperature 5-(or 6-)-nitrophthalic acetic acid is isolated.

3. 4-Nitrophthalic anhydride and malonic acid in pyridine solution according to temperature yield either 5-(or 6-)-nitrophthalic acetic acid or 4-(or 5-)-nitro-2-acetylbenzoic acid.

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