

lower rate. The curves also show a break point that occurs after about 1.5 to 2 hr of reaction. Variations in the propionaldehyde concentration did not change the time needed to reach the change in velocity at the break point. However, the amount inhibited at the break point did depend upon the aldehyde concentration, as did the total inhibition at the end of 6 hr. The early portion of the reaction may represent a reaction of a particular functional group imparting some inhibition, perhaps of asteric nature. The latter portion of the curve, where the lines are nearly parallel, may represent predominantly a continuation of the inhibition due to factors other than any aldehyde present seen in the control. It is evident that the inhibition imparted by propionaldehyde is not as great as that with MA-Na, its dialdehyde counterpart, for any comparable concentration.

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Amines from Aldehydes Derived from the Ozonization of Soybean Esters¹

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Abstract

Investigations were carried out on reductive amination of caproaldehyde, pelargonaldehyde and azelaaldehyde esters, obtainable from ozonolysis of soybean oil products, with ammonia and hydrogen in the presence of nickel catalyst. A solvent system giving good yields of primary amine while suppressing amide formation was devised. Excess ammonia and homogeneous solutions suppressed secondary amine formation. Nonpolar solvents suppressed ammonolysis. Optimum conditions for reaction varied with the aldehyde.

Excellent yields of hexylamine (91%), nonylamine (90%), methyl 9-aminononanoate (92%) and butyl 9-aminononanoate (93%) were obtained from caproaldehyde, pelargonaldehyde, methyl azelaaldehyde and butyl azelaaldehyde, respectively, when aminated in anhydrous ammonia and either cyclohexane or methyl cyclohexane.

Introduction

THE CLASSICAL METHOD of preparing primary amines by reductive alkylation of ammonia with aldehydes in the presence of hydrogen and a hydrogenation catalyst has been the subject of numerous publications and patents for nearly half a century. Ammonolysis of carboxylic esters with ammonia and amines to form amides is equally familiar. In addition, these routes to amines and amides have been the subject, in part, of at least three reviews (3,5,16). However, little mention is made in these reviews of the N-alkylation of compounds containing both

functional groups in the same molecule, such as $\text{OHC}(\text{CH}_2)_x\text{COOH}$ or $\text{OHC}(\text{CH}_2)_x\text{COOR}$. We previously reported on the preparation of such bifunctional materials in excellent yield by the ozonization of soybean esters (2,13,14). The excellent yields and potential of such compounds as fiber-forming intermediates prompted us to explore the possibility of devising an economic route to nylon-9 (a fiber-forming polymer exhibiting toughness, wet strength and lack of water absorbency) from unsaturated fatty acids.

Although primary amines can, and have been, prepared in good yield by other methods, such as the reduction of oximes and phenylhydrazones, our investigations were confined to reductive alkylation because it involved but one operation and would therefore be preferable to other methods.

Otsuki and Funahashi (10,11) have described in British and U.S. patents the ozonization of oleic, erucic, ricinoleic and undecylenic acids to their respective ω -formyl acids. In the best example cited, the ozonide of oleic acid was reduced with sodium sulfite and the resulting aldehydic acid reductively aminated in liquid ammonia and absolute alcohol to give a 78% yield of 9-aminononanoic acid. In another example, ethyl azelaaldehyde was prepared and aminated in the same manner as the aldehydic acid to give a 73% yield of ethyl 9-aminononanoate. Likewise, Carpenter and Reeder (4) reductively aminated ethyl azelaaldehyde to give a 51% yield of the amino ester. Pollart and Miller (12) obtained by direct reductive amination of the ozonolysis products of oleic acid a 29% yield of 9-aminononanoic acid and, of methyl oleate, a 52% yield of methyl 9-aminononanoate, with no ammonolysis of the ester reported. However, in another

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example 10-aminodecanamide was obtained in 70% yield from ethyl undecenoate, illustrating simultaneous ammonolysis. In a Japanese patent, Otsuki and Funahashi (9) describe the preparation of methyl azelaaldehyde in 48% yield by chemical reduction of the ozonolysis products of methyl oleate, and a 93% conversion of the aldehyde ester to methyl 9-aminononanoate by reductively aminating in aqueous ammonia. However, our attempts to repeat this work, as discussed later, resulted in considerable secondary amine formation.

Hasek (6) reported on the N-alkylation of ammonia with various ethyl and isopropyl esters of adipaldehydic and pimelaldehydic acids. The formation of amides was suppressed partly by use of an auxiliary alcoholic solvent corresponding to the ester used and partly by using a relatively inert amine such as *tert*-butyl or *tert*-octyl amine. When a primary amine or ammonia was used as the aminating agent, the ester of a secondary or tertiary alcohol was preferred. Best results were obtained in anhydrous media. In the best example mentioned, isopropyl adipaldehyde was reductively aminated in liquid ammonia and isopropyl alcohol to give a 70% yield of isopropyl 6-aminocaproate.

Our investigations show that compounds incorporating both aldehyde and ester functionalities in the same molecule can be successfully aminated in ammonia with nearly quantitative conversion of aldehyde to amine with little or no ammonolysis. The use of auxiliary nonpolar solvents to accomplish this result will be discussed in this paper.

Experimental

Materials

Methyl and butyl pelargonate were prepared by esterification of pelargonic acid (Emery) (17). Caproaldehyde, pelargonaldehyde and methyl azelaaldehyde (8-formyloctanoate) were obtained from the fractional distillation of bisulfite-purified ozonization products of soybean oil methyl esters (8); likewise, butyl 9-formyloctanoate was prepared from the bisulfite-purified ozonization products of butyl oleate (Swift Esterex 1B5). Raney nickel was made by the method of Adkins and Pavlic (1). Cyclohexane and methylcyclohexane were of spectro-grade. Sulfur compounds had to be removed from industrial grades before amination could proceed smoothly.

Analyses

Amine content was determined by titrating the amine with 0.1 *N* HCl to the bromphenol blue end point. Total amine and amide content was determined by the Kjeldahl method. Gas-liquid chromatographic (GLC) analyses of amine products was made on a 4 ft × 1/4 in. column packed with 20% nitrile silicon gum (General Electric) on Chromosorb W.

Amination of Aldehydes

Only aminations leading to optimum yield of primary amine will be described in detail:

A. Hexylamine. Caproaldehyde (19.0 g, 0.189 mole), cyclohexane (200 ml), liquid ammonia (70 g, 4.12 mole) and Raney nickel (7.0 g, settled weight, wet) were combined in a 400-ml Magna-Dash stainless-steel autoclave previously cooled below the boiling point of ammonia. The autoclave was then sealed and charged twice with 500 psi of hydrogen; the pressure was released after each charge. Next, the vessel was

TABLE I
Reductive Amination of Caproaldehyde in Anhydrous Ammonia

Auxiliary solvent	Reaction conditions			Yield of hexylamine, %
	Time, hr	Temp., C	Pressure, psi ^a	
Cyclohexane.....	0.5	90	1,800	91
Methanol.....	0.5	90	1,500	82
None.....	0.5	90	2,000	86
Cyclohexane.....	4	50	1,500	71
Cyclohexane.....	4	100	3,000	78
Cyclohexane.....	4	140	3,200	77
Methanol.....	4	140	2,800	72

^a Maximum working pressure.

charged to 1,200 psi with hydrogen and the temperature of the mixture was increased to 90C (the observed pressure at temperature was 1,800 psig). After stirring the mixture for 30 min at 90C, the system was allowed to cool down to 35C and excess pressure was vented. The contents of the autoclave were filtered and 50 ml of ether was added to the filtrate. Excess ammonia and ether were removed by heating the solution just high enough to evaporate the ether. The residue was diluted to a volume of 250 ml with cyclohexane and a 2-ml aliquot taken for titration with standard HCl. As determined by titration, aldehyde conversion to amine was 89.7%. To the remainder of the cyclohexane mixture were added 100 ml water and enough concentrated HCl to make the solution decidedly acidic. After this mixture was stirred for 2 hr, the cyclohexane was removed on a rotary evaporator. Potassium hydroxide pellets were added to the aqueous residue until basic to alk-acid paper. The regenerated amine was extracted with methylene chloride and the extracts were combined, dried, filtered and distilled. A 1 in. × 6 in. helices-packed column was used for fractionation and the following fractions taken: (a) bp 58–80C, 1.0 g; (b) 129.5–130.5C, 16.28 g; (c) 133–159C, 1.11 g, residue 0.5 g. Each fraction was analyzed by GLC: (a) 43% CH₂Cl₂, 57% hexylamine; (b) 99.8% hexylamine, Kjeldahl nitrogen for C₆H₁₅N; Calcd. 13.85. Found 13.74; (c) 45% hexylamine, 55% hexyl alcohol. Total yield of hexylamine in all fractions was 91.3%.

B. Nonylamine. Pelargonaldehyde (10.0 g, 0.0704 mole), liquid ammonia (58 g, 2.93 mole) and Raney nickel (6.0 g) were combined, as described for hexylamine, and the autoclave was charged to 1,500 psig. The temperature was raised to 140C and its contents were stirred for 4 hr (maximum working pressure at temperature was 3,400 psi). The vessel was allowed to cool and its contents were filtered. After removal of solvent from the filtrate, the residue (10.0 g) was a water-white liquid requiring 6.99 meq of HCl per gram. Purity based on titration was 100%, bp 36C/0.18 mm Hg, *n*_D²⁰ = 1.4285.

TABLE II
Reductive Amination of Pelargonaldehyde

NHs Form	Auxiliary solvent	Reaction conditions			Yield of nonylamine, %
		Time, hr	Temp., C	Pressure, psi ^a	
Aqueous		4	140	1,600	b
Anhydrous		3	140	2,600	80
Anhydrous		4	140	3,400	100
Aqueous	Methanol	3	90	1,600	68
Aqueous	Methanol	4	140	1,900	70
Anhydrous	Methanol	3	90	2,000	61
Anhydrous	Methanol	4	140	2,400	87
Anhydrous	Cyclohexane	3	140	3,000	76
Anhydrous	Cyclohexane	4	140	2,900	90
Anhydrous	Cyclohexane	5	140	3,000	80
Anhydrous	Methyl cyclohexane	4	140	3,000	86

^a Maximum working pressure. Initial pressure before heating was 1,500 psi.

^b Heterogeneous conditions gave a mixture of primary and secondary amine products.

TABLE III
 Ammonolysis of Pelargonate Esters in Anhydrous Ammonia

Ester	Auxiliary solvent	Reaction conditions			Yield of pelargonamide, %
		Time, hr	Temp., C	Pressure, psi ^a	
Methyl	Methanol	4	140	2,300	48
Methyl	Methanol	4	140	3,400	21
Methyl	Cyclohexane	4	140	3,000	3.5
Methyl	Cyclohexane	5	140	3,100	4.4
Butyl	Methanol	4	140	2,400	24 ^b
Butyl	Butanol	4	140	2,900	1.4
Butyl	Cyclohexane	4	140	3,000	10
Butyl	Cyclohexane	4	140	2,900	0
Butyl	Methyl cyclohexane	4	140	3,100	0

^a Maximum working pressure. Initial pressure before heating was 1,500 psi.

^b Analysis of the unreacted material showed a 36% ester interchange.

Excellent yields of nonylamine were also obtained when auxiliary solvents were used.

Ammonolysis of Pelargonate Esters

The esters, methyl and butyl pelargonate, were subjected to conditions similar to those giving rise to maximum conversion of aldehyde to amine to determine the effect of amination conditions on ammonolysis of esters to amides; however, only the conditions giving minimum amide formation will be described:

A. Pelargonamide from Methyl Pelargonate. Methyl pelargonate (10.0 g, 0.0581 mole), cyclohexane (200 ml), liquid ammonia (38 g, 2.24 mole) and Raney nickel (4 g) were combined and reacted as described for nonylamine (maximum working pressure at temperature was 3,000 psi). After evaporation of the solvent the residue totaled 10.0 g Kjeldahl nitrogen for C₉H₁₉ON: Calcd. 8.91%. Found 0.31%. Yield of pelargonamide, 3.5%. Employment of polar auxiliary solvents resulted in greater conversion of ester to amide.

B. Pelargonamide from Butyl Pelargonate. Butyl pelargonate (10.0 g, 0.0438 mole), cyclohexane (200 ml), anhydrous ammonia (31.0 g, 1.82 mole) and Raney nickel (6.0 g) were combined and reacted as described for preparation of nonylamine (maximum working pressure at temperature was 2,900 psi). After evaporation of the solvent the residue totaled 10.0 g. Kjeldahl analysis of the residue produced no nitrogen.

Although neither nickel catalyst nor hydrogen gas in the two ammonolysis reactions was necessary, they were added to keep the reaction media the same during ammonolysis as during amination.

Amination of Aldehyde Esters

Only aminations leading to optimum yield of amino ester will be described in detail:

A. Methyl 9-Aminononanoate. Methyl azelaaldehy-

date (25.0 g, 0.134 mole), methyl cyclohexane (200 ml), liquid ammonia (60 g, 3.53 mole) and Raney nickel (6 g) were combined and reacted as described for nonylamine (maximum working pressure at temperature was 2,800 psi). Evaporation of the solvent yielded 25 g of a water-white material requiring 4.94 meq of HCl/g, indicating a purity of 92.5%. GLC analysis of the amino ester was made to determine its chief impurity. A peak having the same retention time as methyl 9-hydroxynonanoate appeared. The presence of starting material was not detected. Yield of methyl 9-aminononanoate after distillation, having a purity of 91%, was 88%, bp 85–88C/0.2 mm Hg. Residue as polymer was 12%.

B. Butyl 9-Aminononanoate. Butyl azelaaldehyde (26 g, 0.114 mole), methyl cyclohexane (250 ml), liquid ammonia (75 g, 4.41 mole) and Raney nickel (7 g) were treated as described for nonylamine (maximum pressure at temperature was 3,500 psi). Titration of the residue (26 g), after evaporation of the solvent, required 4.05 meq HCl/g, indicating a purity of 92.7%, bp 118–119C/0.18 mm Hg, $n_D^{20} = 1.4433$; anal. calcd. for C₁₃H₂₇NO₂: C, 68.07; H, 11.81; N, 6.11. Found: C, 68.07; H, 11.91; N, 5.84; yield of butyl 9-aminononanoate after distillation was 86% and the purity, 97.6%. Residue as polymer was 14%. The only impurity found in the distillate was butyl 9-hydroxynonanoate, as determined by GLC.

Results and Discussion

Reductive Amination of Aldehydes

The conditions for N-alkylation with caproaldehyde and pelargonaldehyde were determined separately. Even though most members of a homologous series of compounds react in much the same manner to form new or different derivatives, or both, conditions for their conversion are not necessarily the same. For example, the conditions for optimum conversion of caproaldehyde to hexylamine were 30 min at 90C and 1,500–2,000 psi; whereas the conditions for optimum conversion of pelargonaldehyde to nonylamine were 4 hr at 140C and 2,600–3,400 psi. The conditions leading to the maximum conversion of pelargonaldehyde to primary amine were later applied to the conversion of aldehyde esters (compounds of similar chain length) to amino esters and gave excellent results.

N-alkylations conducted in aqueous ammonia were not practical from the standpoint of aldehyde conversion to primary amine because the heterogeneous conditions (arising from the insolubility of the aldehydes in aqueous ammonia) resulted in considerable secondary amine formation (ca. 30% by weight of distillable products) even in the presence of 70 mole excess of ammonia (all reductive alkylations were car-

 TABLE IV
 Reductive Amination of Azelaaldehyde Esters

Ester	NH ₃ Form	Auxiliary solvent	Reaction conditions			Conversion	
			Time, hr	Temp., C	Pressure, psi ^a	Amine, %	Amide, %
Methyl	Anhydrous	Methanol	4	140	2,500	^b	
Methyl	Anhydrous	Ethanol	3	90	1,800	79	
Methyl	Anhydrous	Ethanol	4	90	1,800	82	
Methyl	Anhydrous	Cyclohexane	4	140	3,000	91	<4
Methyl	Anhydrous	Methyl cyclohexane	4	140	2,800	92	<3
Butyl	Aqueous		3	90	1,600	^c	
Butyl	Aqueous	Ethanol	3	90	1,600	70	
Butyl	Anhydrous	Ethanol	5	90	1,600	65	
Butyl	Anhydrous	Butanol	4	140	2,500	59	0
Butyl	Anhydrous	Cyclohexane	3	140	3,000	80	0
Butyl	Anhydrous	Cyclohexane	4	140	3,100	86	0
Butyl	Anhydrous	Methyl cyclohexane	4	140	3,100	88	0
Butyl	Anhydrous	Methyl cyclohexane	4	140	3,500	93	0

^a Maximum working pressure. Initial pressure before heating was 1,500 psi.

^b Partially polymerized in autoclave during reaction.

^c Heterogeneous conditions gave a mixture of primary and secondary amine products.

ried out in excess ammonia in hopes of suppressing such side reactions). Homogeneous, aqueous, ammoniacal solutions, obtainable by using auxiliary solvents miscible with aqueous ammonia, such as methanol or ethanol, were successful in reducing secondary amine formation to a trace. However, the best conversions were only in the order of 70% in aqueous media.

Reductive alkylations executed under strictly anhydrous conditions were superior to those conducted under aqueous conditions. At optimum conditions for each aldehyde, a 91% conversion of caproaldehyde to hexylamine was attained and quantitative conversion of pelargonaldehyde to nonylamine was achieved. Excellent conversions were also realized when auxiliary solvents (methanol, cyclohexane or methyl cyclohexane) were employed (Tables I and II).

Ammonolysis of Pelargonate Esters

Like pelargonaldehyde, the esters of pelargonic acid were chosen as the guide compounds for our study of the effect of amination conditions on ester ammonolysis because they contained the same number of methylene groups in their alkyl chain as did the aldehyde esters that were to be aminated and, therefore, should give parallel results under similar conditions.

Ammonolysis of butyl and methyl pelargonate in anhydrous ammonia showed the former to be the more stable ester of the two, for twice as much amide was realized from the methyl ester (21%) as from the butyl ester (10%).

The use of auxiliary primary alcohol solvents, corresponding to the ester functionality, promoted ammonolysis of the methyl ester but retarded it for the butyl ester. This effect may be caused by the difference in polarities of the two alcohol solutions. When methyl pelargonate was ammonolyzed in methanol, a 48% conversion of ester to amide took place, but when butyl pelargonate was ammonolyzed in butanol, only 1.4% yield of pelargonamide was obtained. Further, when butyl pelargonate was ammonolyzed in methanol, a 24% yield of pelargonamide was produced, and of the unreacted ester recovered, there was a 36% ester interchange.

Ammonolysis of these esters in the presence of non-polar auxiliary solvents, such as cyclohexane and methyl cyclohexane, gave a much reduced amide formation even at higher pressures and longer reaction times than those used for the polar solvents. Amide formation was negligible when the butyl ester was ammonolyzed in cyclohexane or methyl cyclohexane, and only a 4% yield of pelargonamide was realized from the methyl ester after 5 hr at 140C, even though this temperature is in the range where ammonolysis should proceed readily (15). Summary of results may be found in Table III.

Amination of Azelaaldehyde Esters

Previous knowledge acquired about the aldehyde and ester functionalities under amination conditions was applied to the N-alkylation of ammonia with methyl and butyl 9-formyloctanoate. The amino ester from butyl azelaaldehyde was preferred to the amino ester obtained from methyl azelaaldehyde because of the former's stability on standing at room temperature. Similar observations were made by Horn et al. (7) in their study of the esters of 7-aminoheptanoic acid (nylon-7). They observed the stability of the methyl ester against polymer formation to be less than 1 hr at room temperature, whereas the butyl

ester was stable up to 7 days. However in spite of the greater stability of butyl 7-aminoheptanoate, it formed polymers of lower molecular weight than did the methyl ester.

Our best conversions of aldehyde to amine were obtained under anhydrous conditions. In general, aminations employing auxiliary nonpolar solvents (cyclohexane or methyl cyclohexane) gave higher yields of amino ester than those employing polar auxiliary solvents (methyl, ethyl or butyl alcohol), because the latter system failed to prevent amide formation and gave less conversion of aldehyde to amine. Methyl and butyl azelaaldehyde gave excellent yields of their respective amino esters (91–93% before distillation) with a minimum of amino amide formation when reductively aminated in liquid ammonia and cyclohexane or methyl cyclohexane (Table IV). The yield of methyl and butyl 9-aminononanoate after distillation was 80 and 84%, respectively.

Methyl azelaaldehyde dimethyl acetal could not be aminated under the conditions used for aminating the aldehyde, as would be expected from the known stability of acetals in basic media.

Miscellaneous

The activity of the catalyst used for reductive amination was an important factor affecting the yield of amine. Whenever the catalyst is too active, as with industrial-grade nickel catalyst (Grace) and on occasion with the catalyst prepared from the alloy (1), the aldehydes are reduced to their corresponding alcohols before having a chance to form an addition compound or Schiff's base with the ammonia and then be reduced to the amine. These active catalysts gave excellent results after being deactivated slightly by subjecting them once to reductive amination and then reclaiming and washing the catalysts thoroughly with ethanol.

Another factor affecting amine yields was the quality of the auxiliary solvents. Commercial-grade cyclohexane or methyl cyclohexane contained impurities (believed to be sulfur derivatives, such as thiophene) that poisoned the catalyst thus giving lower yields of primary amine. The commercial-grade solvents had to be subjected to laborious washing with sulfuric acid to make them suitable. The problem of solvent purity was solved by using spectro-grade cyclohexane or methyl cyclohexane and reclaiming the solvent.

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