in 30 cc. of MeOH was refluxed for 4 hr. After cooled, precipitates were filtered (174 mg. 58%). After removal of MeOH by distillation, the residue was recrystallized from Me_2CO giving white needles, m.p. 198° (decomp.), 90 mg. (37%). The IR spectrum was identical with that of 3-pyridazinol 1-oxide.

Ethyl-3-pyridazinecarbamate 2-Oxide (IIe)——A mixture of 1.34 g. of (IId), 50 cc. of EtOH, and 1 cc. of 28% NH_4OH, was hydrogenated with Pd-C, prepared from 0.2 g. of charcoal and 10 cc. of 1% $PdCl_2$ solution. After removal of the catalyst by filtration, the filtrate was neutralized with 10% HCl, and evaporated to dryness. The residue was extracted with $CHCl_3$, and purified on Al_2O_3 chromatography, yielding 1.10 g. (98%) of m.p. 84~85°, which was recrystallized from Et_2O into colorless needles m.p. 84~85°. *Anal.* Calcd. for $C_7H_9O_3N_3$: C, 45.90; H, 4.95; N, 22.94. Found: C, 45.76; H, 4.75; N, 22.39.

2*H*-[1,2,4]oxadiazolo[2,3-*b*]pyridazin-2-one (VI)——Four hundred seventy miligrams of (IIe) was heated at 115±2° for 18 hr. The weight reduced was 61 mg. (52% to the calculated amount of 1 mole of EtOH). The residue was extracted with 10 cc. each of Et_2O three times, and 240 mg. of the starting material was recovered from the extracts (51%). Further, the residue was extracted with 100 cc. each of Et_2O five times on warming, filtered from white flocculents, and the filtrate was concentrated to about 50 cc. When cooled, white needles, m.p. 139.5~140°, 110 mg. (32%) were afforded. *Anal.* Calcd. for $C_5H_3O_2N_3$: C, 43.80; H, 2.25; N, 30.65. Found: C, 43.78; H, 2.52; N, 30.60.

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Summary

On N-oxidation of 3-aminopyridazine and its three derivatives, it was found that their main products were 2-oxides, and 3-acetamidopyridazine gave a small amount of 1-oxide, besides. Further, diazotization of 3-amino-6-chloropyridazine 2-oxide, and thermal cyclization of ethyl-3-pyridazinecarbamate 2-oxide were investigated for additional confirmation of the position of N-oxide.

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150. Zen-ichi Horii, Chuzo Iwata, and Yasumitsu Tamura: Studies on Ergot Alkaloids and Related Compounds. IV.*[2] Preparation and Lithium Aluminum Hydride Reduction of Some Vinylogous Lactams.

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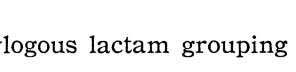
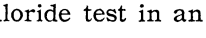
In the course of works on syntheses of Clavine alkaloids,[1] i. e., Agloclavine, Festuclavine, Pyroclavine, Costaclavine, etc., it was observed that condensation of 5-phthalimido-2-tetralone (Ic)[2] with methy 2-methyl-3-methylaminopropionate[3] according to the procedure of Nelson, Ladburg, and Hsi[4] yielded a small amount of by-product besides

*1 Toneyama, Toyonaka, Osaka (堀井善一, 岩田宙造, 田村恭光).
*2 Part III: Yakugaku Zasshi, **82**, 706 (1962).
1) H. Auterhoff: Angew. Chem., **71**, 84 (1959).
2) L.L. McCory, R.C. Terrell: J. Org. Chem., **24**, 131 (1959).
3) D.R. Howton: *Ibid.*, **10**, 277 (1945).
4) N.A. Nelson, J.E. Ladburg, R.S.P. Hsi: J. Am. Chem. Soc., **80**, 6633 (1958).

the expected 2,4-dimethyl-7-phthalimido-3,4,5,6-tetrahydrobenzo[*f*]quinoline-1(2*H*)-one (IIIc). The by-product was shown to have the same molecular formula as that for the main product (IIIc), and its characteristic infrared and ultraviolet spectra suggest that it has a vinylogous lactam grouping  . From survey of literature,[5] it is conceivable that in 2-tetralone derivative not only the 1-positioned carbon atom but also the 3-positioned is activated towards certain reagents. Thus, structure of the by-product was assigned as 1,3-dimethyl-6-phthalimido-2,3,5,10-tetrahydrobenzo[*g*]-quinolin-4(1*H*)-one (IVc). In order to give further support to this assignment, several vinylogous lactams having similar structures to (IIIc) and (IVc) were prepared, and their infrared and ultraviolet spectra compared. Examination of chemical characters of those vinylogous lactams revealed further interesting problems concerning their behavior towards reduction with lithium aluminum hydride. The present paper describes the preparations, infrared and ultraviolet spectra and lithium aluminum hydride reductions of the several vinylogous lactams.

Preparation and Spectrum of Vinylogous Lactam

The vinylogous lactams were prepared by refluxing the ketones (Ia, b, c, V or VIIa, b) and the aminoester (XIV) in toluene followed by refluxing the resulting enamines in ethylene glycol. In the case of employing 5-phthalimido-2-tetralone (Ic) the products were (IIIc) (37% yield) and (IVc) (8% yield) as mentioned above. 2-Tetralone[6] (Ib) similarly yielded 2,4-dimethyl-3,4,5,6-tetrahydrobenzo[*f*]quinolin-1(2*H*)-one (IIIb) in 50% yield, 1,3-dimethyl-2,3,5,10-tetrahydrobenzo[*g*]quinolin-4(1*H*)-one (IVb) in 11% yield and small amounts of uncharacterizable compounds. By employing the same procedure, 2-indanone[7] (Ia), 1-tetralone[8] (V), cyclohexanone (VIIb) and cyclopentanone (VIIa)[9] were converted to 1,3-dimethyl-1,2,3,9-tetrahydro-4*H*-indeno[2,1-*b*]pyridin-4-one (IIIa), 1,3-dimethyl-2,3,5,6-tetrahydrobenzo[*h*]quinolin-4(1*H*)-one (VI), 1,3-dimethyl-2,3,5,6,7,8-hexahydro-4(1*H*)-quinolone (VIIIb), 1-methyl-2,3,5,6,7,8-hexahydro-4(1*H*)-quinolone (VIIIa) (from (VIIb) and ethyl 3-methylaminopropionate[10]), 1,3-dimethyl-1,2,3,5,6,7-hexahydro-4*H*-1-pyridin-4-one (VIIIc) in 80%, 30%, 30%, 63%, and 38% yield, respectively. Treatment of (IIIc) with hydrazine hydrate in boiling ethanol gave 2,4-dimethyl-7-amino-3,4,5,6-tetrahydrobenzo-[*f*]quinolin-1(2*H*)-one (IIId) in 78% yield. Of the vinylogous lactams thus obtained, compound (IIIa,b,c,d) and (VI) gave neither 2,4-dinitrophenylhydrazone nor picrate, and a negative ferric chloride test in an ethanolic solution. These compounds only have one strong absorption band corresponding to a vinylogous amide grouping  [11] in the infrared (Table I). These spectral data coincided with that of 4-methyl-3,4,5,6-tetrahydrobenzo[*f*]quinolin-1(2*H*)-one (IIIb: 9-H insitead of 9-CH_3) reported by Nelson, *et al.*[4] In the ultraviolet (Table I), these compounds exhibited two absorption maxima at near 230 mμ and 310 mμ. Thus, it would be concluded that these five compounds have a similar structure, and the absorption in the infrared and ultraviolet regions mentioned above are characteristic for a vinylogous lactam grouping cross-conjugated with a phenyl group.

5) Haco. Gesellschaft A.G. Swiss. 290,291 Aug. 1, 1953 (C.A., **49**, 2513). M.D. Soffer, *et al.*: J. Am. Chem. Soc., **74**, 1556 (1952).
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11) N.H. Cromwell *et al.*: *Ibid.*, **71**, 3337 (1949); N.J. Leonard, J.A. Adamcik: *ibid.*, **81**, 595 (1959); J. Weinstein, G.M. Wyman: J. Org. Chem., **23**, 1618 (1958); H.F. Holtzclaw *et al.*: J. Am. Chem. Soc., **80**, 1100 (1958); O. Cervinka: Collection Czechoslov. Chem. Commun., **25**, 1183 (1960).

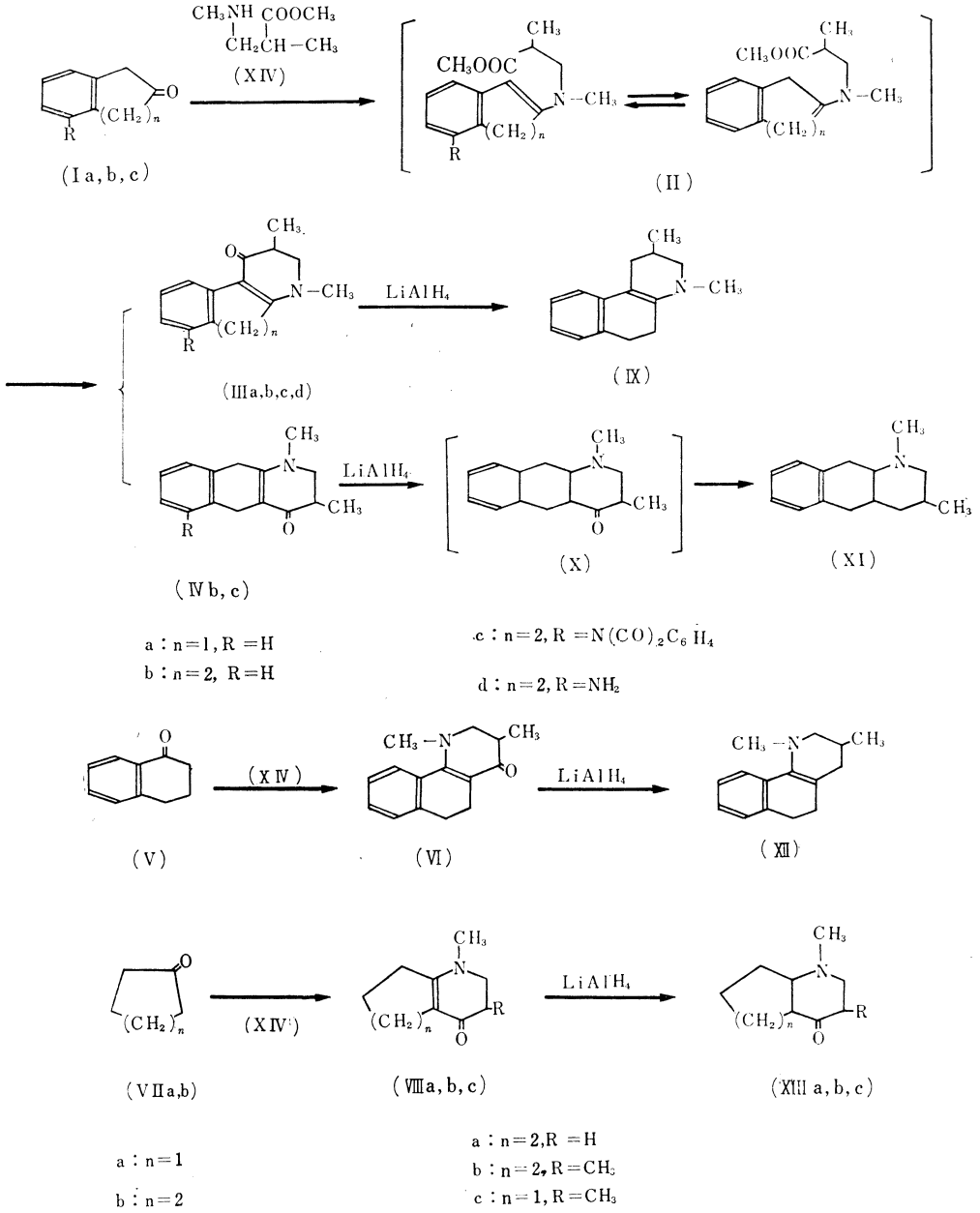

Chart 1.

On the other hand, both the side products, (IVb) and (IVc), showed two characteristic absorptions at near 1618 cm^{-1} (s) and 1540~1563 cm^{-1} (vs) in the infrared region and a single band at near 335 mμ in the ultraviolet region.[13] They gave neither 2,4-dinitrophenylhydrazone nor picrate, but were soluble in dilute hydrochloic acid. Compound (IVb) gave a dark green coloration with an ethanolic ferric chloride solution.[12] These

12) F. Wille, L. Saffer: Ann., **568**, 34 (1950); J. Weinstein, G.M. Wyman: J. Org. Chem., **23**, 1618 (1958); H. Fritz: Chem. Ber., **92**, 1809 (1959).

13) cf. K. Bowden *et al.*: J. Chem. Soc., **1946**, 45; N.H. Cromwell, W.R. Watson: J. Org. Chem., **14**, 411 (1949).

physical and chemical properties are most likely attributable to a normal vinylogous lactam and the spectral data presented are so characteristic that (IVb) and (IVc) can be distinguished from the cross-conjugated vinylogous lactams, (IIIa-d) and (VI). Compounds (VIIIa), (VIIIb), and (VIIIc) prepared as model compounds for (IVb) and (IVc) exhibited infrared absorption bands at near 1610 cm^{-1} (s) and 1550 cm^{-1} (vs), ultraviolet absorption maximum at near 335 mμ and gave a positive ferric chloride test. The similarity of the infrared and ultraviolet spectra and ferric chloride tests of (IVb) and (IVc) with those of (VIIIa), (VIIIb) and (VIIIc) would establish the structures of (IVb) and (IVc) as assigned. This conclusion seems to be reasonable from the chemical behavior of 2-tetralone derivative, i.e., considerable reactivity at the 3-position carbon atom.

Reduction of the Vinylogous Lactam with Lithium Aluminum Hydride

Nelson, *et al.*[4] reported that the reduction of 4-methyl-3,4,5,6-tetrahydrobenzo[*f*]-quinolin-1(2*H*)-one with lithium aluminum hydride yielded the enamine, 4-methyl-1,2,3,4,5,6-hexahydrobenzo[*f*]quinoline. When compound (IIIb) was reduced with an excess of lithium aluminum hydride in refluxing ether, 2,4-dimethyl-1,2,3,4,5,6-hexahydrobenzo-[*f*]quinoline (IX) was obtained in quantitative yield as in the case of Nelson, *et al.*[4] The same reduction of (VI) similarly gave 1,3-dimethyl-1,2,3,4,5,6-hexahydrobenzo[*h*]-quinoline (XII) in 80% yield.

On the other hand, the reductions of (VIIIa), (VIIIb), and (VIIIc) employing the same conditions yielded 1-methyl-2,3,4a,5,6,7,8,8a-octahydro-4(1*H*)-quinolone (XIIIa), 1,3-dimethyl-2,3,4a,5,6,7,8,8a-octahydro-4(1*H*)-quinolone (XIIIb) and 1,3-dimethyl-1,2,3,4a,5,6,7,7a-octahydro-4*H*-1-pyridin-4-one (XIIIc) in 40%, 50%, and 50% yield, respectively. The reduction of (IVb) afforded a 50% yield of 1,3-dimethyl-1,2,3,4,4a,5,10,10a-octahydrobenzo[*g*]quinoline (XI) and a 30% yield of the ketone, probably (X), which could not be characterized because of failure to make a crystalline derivative.

The formation of the enamine would be interpreted by 1,2-addition mechanism and the formation of the aminoketone by 1,4-addition mechanism.

Experimental

General Method of the Preparation of the Vinylogous Lactams[4]——A solution of one mole of the ketone and one mole of the aminoester in toluene was heated under reflux for 6~10 hr. in N_2 atmosphere. The water produced during the condensation reaction was removed from the mixture by means of a Dean-Stark apparatus. The solvent was removed under reduced pressure yielding the enamine which was used without further purification. The enamine thus obtained was dissolved in ethylene glycol and heated under reflux for 6~10 hr. The reaction mixture was cooled, extracted first with Et_2O and next with benzene. These two extracts were separately washed well water and dried over Na_2SO_4. The solvent was removed from each of the Et_2O and benzene extracts, and the residues were subjected to vacuum distillation or recrystallization.

Melting point, datum of elemental analysis and main absorptions in the infrared and ultraviolet spectra of the product are listed in Table I.

Reaction of 2-Tetralone and Methyl 2-Methyl-3-methylaminopropionate——A solution of 29 g. (0.2 mole) of 2-tetralone and 26 g. (0.2 mole) of methyl 2-methyl-3-methylaminopropionate in 300 ml. of toluene was heated under reflux for 10 hr. in N_2 atmosphere. The water produced during the condensation reaction was removed from the mixture by means of a Dean-Stark apparatus. The solvent was removed under reduced pressure yielding the enamine. The crude enamine was dissolved in 240 ml. of ethylene glycol, and the solution was refluxed for 10 hr. The reaction mixture was extracted successively with Et_2O and benzene and each of the extracts was washed with water and dried. Concentration of the Et_2O extract and cooling gave 13 g. (29%) of crude 2,4-dimethyl-3,4,5,6-tetrahydrobenzo[*f*]quinolin-1(2*H*)-one(IIIb), m.p. 98~100°. Distillation of the residue from the crystallization filtrate gave an additional 10 g. (20%) of the crude (IIIb), b.$p_{0.2}$ 145~150°; m.p. 98~100°, and 1 g. (2%) of crude 1,3-dimethy-2,3,5,10-tetrahydrobenzo[*g*]quinolin-4(1*H*)-one (IVb), b.$p_{0.2}$ 160~170°; m.p. 100~120°.

Table I.

Comp.	m.p.[a] (°C)	Mol. formula	Analysis (%) Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N	λ_{max} mμ EtOH $\epsilon \times 10^3$		ν_{max} (1500~1700) cm^{-1} $CHCl_3$
(IIIa)	139~140[c]	$C_{14}H_{15}ON$	78.84	7.09	6.57	78.62	6.96	6.49	228.5 304	7.7 12.1	1653(vs), 1613(s), 1580(w)
(IIIb)	99~100[c]	$C_{15}H_{17}ON$	79.26	7.54	—	79.11	7.24	—	229.7 308.5	10.8 16.8	1654(vs), 1605(m), 1575(w)
(IIIc)	246~247[f]	$C_{23}H_{20}O_3N_2$	74.17	5.41	—	74.35	5.30	—	308	10.7	1664(vs), 1635(m), 1597(w), 1575(w)[b]
(IIId)	193~194[d]	$C_{15}H_{18}ON_2$	74.35	7.49	11.56	74.66	7.70	11.53	231 311	16.0 16.3	1650(vs), 1595(w), 1580(m)[b]
(VI)	97~98[e]	$C_{15}H_{17}ON$	79.26	7.54	6.16	79.21	7.53	6.40	226.8 285 298 (shoulder)	17.2 6.6	1653(vs)
(IVb)	120~121[c]	$C_{15}H_{17}ON$	79.26	7.54	6.16	79.31	7.37	6.29	331.2	12.2	1620(s), 1600(vs), 1553~1563(vs)
(IVc)	249[d]	$C_{23}H_{20}O_3N_2$	74.17	5.41	—	74.25	5.55	—	335	5.6	1618(s), 1600(s), 1563(vs)[b]
(VIIIa)	Picrate[d] 155~156	$C_{16}H_{18}O_8N_4$	48.73	4.60	14.21	48.70	4.69	14.41	337	10.1	1613(s), 1548(vs)
(VIIIb)	Picrate[d] 146	$C_{17}H_{20}O_8N_4$	50.00	4.94	13.72	50.19	5.23	13.91	335	12.8	1608(s), 1550(vs)
(VIIIc)	Picrate[d] 144~145	$C_{16}H_{18}O_8N_4$	48.73	4.60	14.21	49.10	4.64	14.43	331	15.0	1623(s), 1567(vs)

a) Uncorrected *b*) Nujol *c*) Recryst. solvent : EtOH-H_2O *d*) Recryst. solvent : EtOH
e) Recryst. solvent : petr. ether *f*) Recryst. solvent : benzene-petr. benzin

The benzene extract was washed with water and dried. Concentration of the benzene extract and cooling gave 4 g. (8%) of (IVb), m.p. 110~120°. Fractional distillation of the residue from the crystallization filtrate gave an additional 0.5 g. (1%) of (IIIb), b.$p_{0.2}$ 145~150°; m.p. 98~100°, 0.5 g. (1%) of (IVb), b.$p_{0.2}$ 160~170°; m.p. 100~120°, and 1 g. (2%) of uncharacterizable compound (A), b.$p_{0.2}$ 180~190°; m.p. 110~116°.

The analytical sample of (IIIb) was recrystallized from H_2O-EtOH, m.p. 99~100°. The analytical sample of (IVb) was recrystallized from H_2O-EtOH, m.p. 120~121°. The analytical samdle of the compound (A) was recrystallized from Et_2O, m.p. 116~117°. IR $\nu_{max}^{CHCl_3}$ cm^{-1} : 1618(s), 1540(vs). UV $\lambda_{max}^{EtOH}(\epsilon)$: 278 mμ (15.2×10^3), 353 mμ (10.8×10^3). From elemental analyses of this compound and its picrate it was shown that the compound (A) had a molecular formula of $C_{15}H_{17}ON$. *Anal.* Calcd. for $C_{15}H_{17}ON$: C, 79.26; H, 7.54; N, 6.16. Found : C, 79.43; H, 7.37; N, 6.01. The picrate of the compound (A) was recrystallized from EtOH, m.p. 180~181°. *Anal.* Calcd. for $C_{21}H_{20}O_8N_4$: C, 55.26; H, 4.42; N, 12.28. Found : C, 55.32; H, 4.39; N, 12.56.

In another run which employed 5 g. of 2-tetralone and 4.5 g. of methyl 2-methyl-3-methylaminopropionate, chromatographic purification of the benzene extract through an alumina column (300 g.) using benzene-*n*-hexane (1:1) and benzene as eluent gave three kinds of crystals : 0.7 g. of compound (B), m.p. 204~206°, 0.1 g. of compound (C), m.p. 82~84° and 3 g. (38%) of (IIIb). Recrystallization of the compound (B) from EtOH-H_2O gave a sharp melting point of 206° and the picrate was recrystallized from EtOH, m.p. 205~206° (decomp.).

These minor products of compound (A), (B) and (C) were not further characterized.

Reaction of 5-Phthalimido-2-tetralone and Methyl 2-Methyl-3-methylaminopropionate——A solution of 5 g. (0.017 mole) of 5-phthalimido-2-tetralone and 2.5 g. (0.017 mole) of methyl 2-methyl-3-methylaminopropionate in 50 ml. of toluene was heated under reflux for 10 hr. in N_2 atmosphere. The water produced during the condensation reaction was removed from the mixture by means of a Dean-Stark apparatus. The solvent was removed under reduced pressure yielding the enamine. The crude enamine was dissolved in 40 ml. of ethylene glycol and heated under reflux for 15 hr. The reaction mixture was extracted with Et_2O. The resulting solution was washed with water and dried. Concentration of the Et_2O solution and cooling gave 3 g. of the crude product. This crude product was extracted with 50 ml. × 2 of hot EtOH. Concentration of the EtOH solution and cooling gave 0.5 g. (8%) of 1,3-dimethyl-6-phthalimido-2,3,5,10-tetrahydrobenzo[*g*]quinolin-4-(1*H*)-one(IVc), m.p. 249° (decomp.). Crystallization of the residue, insoluble in hot EtOH, from petr. benzin-benzene gave 2.3 g. (37%) of 2,4-dimethyl-7-phthalimido-3,4,5,6-tetrahydrobenzo[*f*]quinolin-1(2*H*)-one(IIIc), m.p. 246~247°.

2,4-Dimethyl-7-amino-3,4,5,6-tetrahydrobenzo[*f*]quinolin-1(2*H*)-one (IIId)——A solution of 0.1 ml.

of 50% $NH_2NH_2 \cdot H_2O$ was added dropwise to a boiling mixture of 200 mg. of (IIIc) and 60 ml. of anhyd. EtOH, and then the mixture was refluxed for 4 hr. The solvent was removed under reduced pressure, the residual half-solid was washed with 1 ml. of 3% NH_4OH and crystallized from EtOH, to give 102 mg. (78%) of 2,4-dimethyl-1-oxo-7-amino-1,2,3,4,5,6-hexahydrobenzo[*f*]quinoline (IIId), m.p. 193~194°.

1,3-Dimethyl-2,3,5,6,7,8-hexahydro-4(1*H*)-quinolone (VIIIb)——A solution of 7.8 g. (0.08 mole) of cyclohexanone and 10.4 g. (0.08 mole) of methyl 2-methyl-3-methylaminopropionate in 100 ml. of toluene was heated under reflux for 50 hr. in N_2 atmosphere. The water produced during the condensation reaction was removed from the mixture by means of a Dean-Stark apparatus. The solvent was removed under reduced pressure, and the residual oil was distilled to afford 9 g. (63%) of 1,3-dimethyl-2,3,5,6,7,8-hexahydro-4(1*H*)-quinolone (VIIIb), b.p_4 153~155°. The picrate of (VIIIb) was recrystallized from EtOH, m.p. 146°.

General Method of the Reduction of the Vinylogous Lactams with $LiAlH_4$——One mole of $LiAlH_4$ was added to a stirred solution of one mole of vinylogous lactam in an appropriate volume of anhyd. Et_2O. The mixture was then refluxed for 4~6 hr. before decomposing the excess hydride by addition of AcOEt followed by ca. 50% KOH. The Et_2O solution was dried over anhyd. K_2CO_3 and evaporated to give an oil which was distilled under reduced pressure.

Table II.

Comp.	b.p.[a] (°C/mm.)	m.p.[a] (°C) Picrate	Mol. formula	Analysis (%) Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N
(IX)	135/0.05	——	$C_{15}H_{19}N$	84.45	8.98	—	84.63	8.61	—
(XII)	(m.p. 77~78°)	——	$C_{15}H_{19}N$	84.45	8.98	6.57	84.81	8.84	6.45
(XI)	——	179~180	$C_{21}H_{24}O_7N_4$	56.75	5.44	—	56.99	5.05	—
(XIIIa)	135~136/17	180~181[b]	$C_{16}H_{20}O_8N_4$	48.48	5.09	14.14	48.72	4.86	14.41
(XIIIb)	117~121/12	201~202[b]	$C_{17}H_{22}O_8N_4$	49.75	5.40	13.65	50.11	5.63	13.58
(XIIIc)	92/4	158~159[b]	$C_{16}H_{20}O_8N_4$	48.48	5.09	14.14	48.18	5.14	14.36

a) Uncorrected *b*) Decomposed

2,4-Dimethyl-1,2,3,4,5,6-hexahydrobenzo[*f*]quinoline (IX)——A solution of 1.5 g. (0.0066 mole) of (IIIb) in 60 ml. of anhyd. Et_2O was reduced with $LiAlH_4$ (600 mg.) according to the general method affording a quantitative yield of (IX), b.$p_{0.05}$ 130~135°. The analytical sample of 2,4-dimethyl-1,2,3,4,5,6-hexahydrobenzo[*f*]quinoline (IX), b.$p_{0.05}$ 135°, was distilled.

1,4-Dimethyl-1,2,3,4,4*a*,5,10,10*a*-octahydrobenzo[*g*]quinoline (XI)——A solution of 340 mg. of (IVb) in 100 ml. of anhyd. Et_2O was reduced with $LiAlH_4$ (400 mg.) according to the general method affording 170 mg. (50%) of (XI), b.$p_{0.03}$ 120~130° (bath temp.) and 100 mg. of the by-product, b.$p_{0.03}$ 145~150° (bath temp.). The picrate of (XI) was crystallized from EtOH, m.p. 179~180°. The by-product did not give the picrate and the 2,4-dinitrophenylhydrazone, but its infrared absorption at 1706 cm^{-1} in $CHCl_3$ seems to be ascribed to ketone group.

Summary

Condensation of 5-phthalimido-2-tetralone (Ic) and methyl 2-methyl-3-methylaminopropionate (XIV) by the method of Nelson, *et al.*[4] yielded (IIIc) as a major product and (IVc) as a minor product. The structures of both products were established mainly by comparison of their infrared and ultraviolet spectra with those of several compounds having similar structures. Consequently, it was found that nine vinylogous lactams newly prepared could be classified into two groups, normal vinylogous lactams (IVb, c), (VIIIa, b, c) and cross-conjugated ones (IIIa~d), (VI), from their spectral data and their behaviors towards lithium aluminum hydride reduction.

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