in urine and feces. As shown in Table II, approximately 0.5% of Benhepazone administered was excreted in urine during 48 hours, and less than 3% in feces. In addition to this fact, it has been observed that only 0.69% of the dose was detected in the digestive tract 24 hours after the administration. Thus, a large portion of Benhepazone administered could not be recovered.

Table II. Excretion[a] of Benhepazone in Urine and Feces of Rats after Oral Administration (100 mg./kg.)

Rat No.	Urine		Feces	
	0~24 hr.	24~48 hr.	0~24 hr.	24~48 hr.
1	0.53	0.038	2.32	0.20
2	0.33	0.14	2.80	0.096
3	0.29	0.019	0.30	0.19
4	0.53	0.24	0.25	0.67
5	0.43	0.050	1.10	0.30
6	0.34	0.040	0.39	0.10
7	0.44	0.025	0.69	0.086
Means±SD[b]	0.41±0.10	0.080±0.082	1.12±1.04	0.23±0.22

a) Expressed in per cent of dose. *b*) Standard deviation.

From these results, Benhepazone is considered to be almost completely absorbed from the gastrointestinal tract, and then transformed into related compounds. This assumption is supported by the fact that various metabolic products were found in urine.[1]

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Ikuo Matsumoto[*1] : Potassium Carbonate as a Base in the Alkylation of Ethyl Acetamidocyanoacetate.

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Alkylation of ethyl acetamidocyanoacetate followed by hydrolysis is a useful preparative method for α-amino acids and their derivatives. The condensation with alkyl halides has usually been carried out in the presence of sodium ethoxide[1] or sodium hydride.[2] We wish to describe a convenient procedure for the alkylation by using a common and weak base, potassium carbonate as a condensing agent. Its use avoids the need for strictly anhydrous conditions.[1,2]

The reaction is effected in boiling acetone in the presence of anhydrous potassium carbonate and alkylated esters are obtained generally in good yields with reactive primary alkyl halides such as ethyl bromide, allyl chloride, and many benzyl-type halides including

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1) N. F. Albertson, B. F. Tullar : J. Am. Chem. Soc., **67**, 502 (1945); N. F. Albertson : *Ibid.*, **68**, 450 (1946).
2) J. Shapira, R. Shapira, K. Dittmer : *Ibid.*, **75**, 3655 (1953).

several heterocyclic compounds. Addition of a catalytic amount of sodium iodide is favorable to enhance the reactivity of alkyl chlorides or bromides, which are more readily available than the corresponding iodides and used in many cases. Attempts to alkylate ethyl acetamidomalonate in a similar manner failed to give isolable products with many halides other than *p*-nitrobenzyl chloride, which gave an alkylation product in a yield of 84%.

Potassium carbonate has been a familiar condensing agent for N- or O-alkylation of various compounds, such as phthalimide[3] and phenols,[4] with alkyl halides or sulfates. In C-alkylation of active methylene compounds, however, its use is less popular and there seem to be but a few examples, especially in cyanoacetic and malonic esters. In an extensive review appeared in 1957,[5] potassium carbonate was classified as a base of limited use for "the alkylation of esters and nitriles." Although enolizable 1,3-diketones are successfully C-monoalkylated[6] with alkyl iodides and potassium carbonate, the long-known Claisen carbonate method for alkylation of β-ketocarboxylic esters[7] often leads to O- as well as C-alkylation. A recent paper[8] describes benzylation of ethyl malonate in a dipolar aprotic solvent with less basic sodium bicarbonate as a base, where the yields reported are not promising for preparative use even at a reaction temperature of 125°. Potassium carbonate has been used advantageously in monoalkylation of cyanoacetic esters but at rather high temperatures and, apart from the problem of dialkylation, with a large excess of the neat ester as a reaction medium.[9] In another case for second alkylation of a monosubstituted cyanoacetic ester, a yield of 60% was attained only by heating for 12~20 hours with twice the equivalent amount of a reactive alkyl halide.[10]

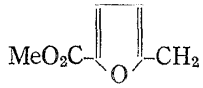
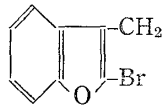
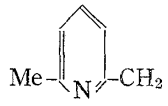
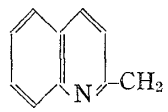
A casual case of C-methylation of ethyl acetamidocyanoacetate was reported[11] in an effort to prepare the N-methylated product by treatment with methyl iodide in acetone solution in the presence of powdered potassium hydroxide, but the yield was 50% as compared with 71% for the conventional method with sodium ethoxide. Amberlite IRA-400 was also a base for alkylation of an analogous compound, acetamidocyanoacetamide in aqueous ethanol and a highest yield of 50% was reported with allyl bromide.[12]

By contrast, potassium carbonate proved to be quite useful for alkylation of ethyl acetamidocyanoacetate; the reaction was performed in satisfactory yields with nearly molar equivalents of many alkyl halides under mild conditions. Although an attempt to alkylate with isopropyl bromide was not so successful and resulted in low yields, this method is probably of general applicability in the alkylation of ethyl acetamidocyanoacetate and its analogs with reactive primary alkyl halides. The use of potassium carbonate, combined with a choice of acetone as the reaction medium, eliminates the need for strictly anhydrous conditions, simplifies work-up of the reaction mixtures, and

3) H.R. Ing, R.H.F. Manske: J. Chem. Soc., **1926**, 2348; Org. Syntheses, Coll. Vol. **2**, 83 (1950); B. Vassel: U.S. Pat. 2,757,198 (1956) (C.A., **51**, 2024*a*).
4) L. Claisen, O. Eisleb: Ann., **401**, 29 (1913); H. Meerwein: "Methoden der organischen Chemie (Houben-Weyl)," **6/3**, 55, 57, 64 (1965); C.F.H. Allen, J.W. Gates, Jr.: Org. Syntheses, Coll. Vol. **3**, 140 (1955).
5) A.C. Cope, H.L. Holmes, H.O. House: Org. Reactions, **9**, 119 (1957).
6) K. von Auwers, H. Jacobsen: Ann., **426**, 227 (1922); A. Brändström: Acta Chem. Scand., **4**, 208 (1950); A.W. Johnson, E. Markham, R. Price: Org. Syntheses, **42**, 75 (1962).
7) K. von Auwers: Ber., **71**, 2082 (1938); H. Henecka: "Methoden der organischen Chemie (Houben-Weyl)," **8**, 603 (1952).
8) S. Sato, H. Sasaki, M. Nakamura: Yuki Gosei Kagaku Kyokai Shi, **24**, 120 (1966).
9) G.M. Robinson: J. Chem. Soc., **125**, 226 (1924); R. Robinson, J.S. Watt: *Ibid.*, **1934**, 1539; N.J. Leonard, W.C. Wildman: J. Am. Chem. Soc., **71**, 3092 (1949); P.E. Gagnon, G. Nadeau, R. Côté: Can. J. Chem., **30**, 592 (1950); J. Davoll: J. Chem. Soc., **1960**, 131.
10) K. Pettersson: Acta Chem. Scand., **4**, 395, 1319 (1950).
11) F.C. Uhle, L.S. Harris: J. Am. Chem. Soc., **78**, 381 (1956).
12) K. Shimo, S. Wakamatsu: J. Org. Chem., **28**, 504 (1963).

Table I. Alkylation of Ethyl Acetamidocyanoacetate with Various RX

$HC(CN)CO_2C_2H_5$ (I) + RX
$NHCOCH_3$

R	X	Molar Ratio of I–RX	Yield[a] (%)	m.p.[b] (°C)	Appearance
$C_6H_5CH_2$	Cl	1	91	132～133[l]	colorless scales
p-$ClC_6H_4CH_2$	Cl[c]	1.05:1	96	163～164	colorless scales
p-$FC_6H_4CH_2$	Cl[d]	1.05:1	82	162～163[m]	colorless leaflets
p-$O_2NC_6H_4CH_2$	Cl	1	98	185～186[n]	yellowish scales
o-$O_2NC_6H_4CH_2$	Br[e]	1	91	164～165[o]	yellowish needles
2,6-$Cl_2C_6H_3CH_2$	Cl[f]	1	90	151～152	colorless scales
3,4-$CH_2O_2C_6H_3CH_2$	Cl[f]	1	93	159～160	colorless scales
3,4-$O_2N(MeO)C_6H_3CH_2$	Cl	1.1:1	84	139～140.5	yellowish prisms
4-(*p*-$O_2NC_6H_4O$)$C_6H_4CH_2$	Cl[f]	1.1:1	96	150～151	yellowish scales
Et	Br	1:1.2	88	131[i,p]	colorless needles
CH_2:$CHCH_2$	Cl	1:1.2	87	88～89[j,q]	colorless needles
CH_2:$CMeCH_2$	Cl	1:1.2	86	80～81[j,r]	colorless scales
o-$C_6H_4(CO)_2N(CH_2)_3$	Br	1.05:1	97	211～212[k,s]	colorless needles
	Cl[g]	1.05:1	87	151～152	colorless prisms
	Br[h]	1.1:1	85	171～171.5[k]	colorless needles
	Cl[f]	1:1.1	84	135～135.5	colorless prisms
	Cl	1	93	140～141	colorless needles

a) Listed are crude yields, relative to I or RX depending on the molar ratio. The individual reaction product was once recrystallized or merely rinsed with an appropriate solvent, until the m.p. reached within 2° as compared with that of the analytical sample, and wegihed.
b) The values are given for analytical samples; recrystallization was carried out from 50～60% aq. EtOH unless otherwise specified.
c) Prepared by chloromethylation of chlorobenzene[13] and recrystallized from petr. ether, freezing point 27°.
d) Prepared by chlorination with SO_2Cl_2,[14] b.p_{15} 68～70°, n_D^{27} 1.5086.
e) Prepared by N-bromosuccinimide treatment of *o*-nitrotoluene[15] and recrystallized just before use, m.p. 44～46° (EtOH).
f) See Experimental section.
g) Prepared from methyl 2-furoate.[16]

13) F. Konishi, J. Nakazawa : Takamine Kenkyusho Nempo, **5**, 18 (1953).
14) F.L.M. Pattison, B.C. Saunders : J. Chem. Soc., **1949**, 2745.
15) G.H. Daub, R.N. Castle : J. Org. Chem., **19**, 1571 (1954); cf. A. Kalir : Org. Syntheses, **46**, 81 (1966).
16) A.L. Mndzhonian, M.T. Grigorian : "Syntheses of Heterocyclic Compounds," **1**, 29 (1959), Consultants Bureau, Inc., N.Y.
17) H. Erlenmeyer, W. Grubenmann : Helv. Chim. Acta, **30**, 297 (1947).
18) T. Ohta : Yakugaku Zasshi, **68**, 226 (1948).

Halides in the Presence of Anhydrous Potassium Carbonate

$$\xrightarrow[\text{in acetone}]{K_2CO_3} RC(CN)CO_2C_2H_5 \quad (NHCOCH_3)$$

Formula	Analysis					
	Calcd.			Found		
	C	H	N	C	H	N
$C_{14}H_{16}O_3N_2$	64.60	6.20	10.76	64.32	5.89	10.40
$C_{14}H_{15}O_3N_2Cl$	57.05	5.13	9.51	57.15	5.16	9.55
$C_{14}H_{15}O_3N_2F$	60.46	5.43	10.07	60.97	5.72	10.04
$C_{14}H_{15}O_5N_3$	55.08	4.95	13.76	55.14	5.04	13.81
$C_{14}H_{15}O_5N_3$	55.08	4.95	13.76	55.19	5.06	13.78
$C_{14}H_{14}O_3N_2Cl_2$	51.08	4.29	8.51	51.26	4.09	8.25
$C_{15}H_{16}O_5N_2$	59.20	5.30	9.21	59.15	5.37	9.22
$C_{15}H_{17}O_6N_3$	53.73	5.11	12.53	53.60	4.92	12.47
$C_{20}H_{19}O_6N_3$	60.45	4.82	10.58	60.94	4.90	10.67
$C_9H_{14}O_3N_2$	54.53	7.12	14.13	54.56	7.20	14.05
$C_{10}H_{14}O_3N_2$	57.13	6.71	13.33	57.14	6.79	13.27
$C_{11}H_{16}O_3N_2$	58.91	7.19	12.49	58.92	7.40	12.46
$C_{18}H_{19}O_5N_3$	60.49	5.36	11.76	60.31	5.28	11.35
$C_{14}H_{16}O_6N_2$	54.54	5.23	9.09	54.64	5.24	9.13
$C_{16}H_{15}O_4N_2Br$	50.67	3.99	7.39	50.63	4.04	7.39
$C_{14}H_{17}O_3N_3$	61.08	6.22	15.26	60.53	5.94	15.30
$C_{17}H_{17}O_3N_3$	65.58	5.50	13.50	65.23	5.22	13.24

h) Prepared from 3-methylcoumarone.[17]
i) Recrystallized from aq. acetone.
j) From 25% aq. EtOH.
k) From 50% aq. AcOH.
l) Lit. m.p. 134°;[1] 133°;[18] 130~132°.[19]
m) Lit. m.p. 165~166°.[20]
n) Lit. m.p. 185~187°.[21]
o) Lit. m.p. 150~151°.[22]
p) Lit. m.p. 131°;[18] 130°.[1]
q) Lit. m.p. 89°.[1]
r) Lit. m.p. 81°.[1]
s) Lit. m.p. 214.5~215°;[23] 210°.[24]

19) J. M. Stewart : J. Org. Chem., **26**, 3360 (1961).
20) E. L. Bennett, C. Niemann : J. Am. Chem. Soc., **72**, 1800 (1950).
21) J. H. Burckhalter, V. C. Stephens : *Ibid.*, **73**, 56 (1951).
22) A. L. Davis, O. H. P. Choun, D. E. Cook, T. J. McCord : J. Med. Chem., **7**, 632 (1964).
23) M. Fields, D. E. Walz, S. Rothchild : J. Am. Chem. Soc., **73**, 1000 (1951).
24) R. Gaudry : Can J. Chem., **31**, 1060 (1953).

prevents a competitive etherification often encountered in reactions with reactive halides and sodium alkoxide in alcohols. The reaction conditions and yields are listed in Table I.

Experimental

The alkylation procedure is illustrated by typical examples. Ethyl acetamidocyanoacetate (I)[25] and ethyl acetamidomalonate (II)[26] were prepared by known methods. Reagent grade acetone was dried over K_2CO_3 and commercial anhyd. K_2CO_3 was used as received.

Ethyl Acetamido(3,4-methylenedioxybenzyl)cyanoacetate (III)——A stirred mixture of 34 g. (0.2 mole) of I, 34.1 g. (0.2 mole) of freshly distilled 3,4-methylenedioxybenzyl chloride (IV), 20 g. (0.145 mole) of K_2CO_3, and 1 g. of NaI in 100 ml. of acetone was refluxed on a water bath for 8 hr.; near the end of reaction a part of III crystallized out. The hot slurry was treated with 120 ml. of H_2O to dissolve most of the inorganic salts, cooled, and the precipitate collected and digested with a large amount of H_2O and then with a little MeOH to give 55~58 g. (90~95.5%) of almost colorless, crude III, m.p. 158~160°. Recrystallization as indicated in Table I afforded an analytical sample. Another run was carried out by using a dried benzene extract of IV (see below) from 32 g. (0.21 mole) of 3,4-methylenedioxybenzyl alcohol (V) instead of the distilled IV with or without stripping the benzene to give III in a reproducible yield of 96 or 91%, respectively. Previously, III was prepared in this laboratory with NaOEt as a base in an 81% yield;[27] the loss in yield may be due to the etherification of IV with the solvent EtOH.[28]

Ethyl 2-Acetamido-2-cyanobutyrate——A magnetically stirred mixture of 17 g. (0.1 mole) of I, 13 g. (0.12 mole) of EtBr, 15 g. (0.108 mole) of K_2CO_3, and 0.5 g. of NaI in 50 ml. of acetone was refluxed for 7 hr., where the bath temperature was raised slowly from 45° to 70° over the initial 2 hr. The hot mixture was clarified through a layer of Celite-carbon and the residue washed with hot acetone. The combined filtrate and washings were evaporated and the residue was triturated with H_2O to give 17.4 g. (88%) of crystals, m.p. 130~131°. Recrystallization yielded long needles (cf. Table I).

Ethyl Acetamido(*p*-nitrobenzyl)malonate (VI)——A mixture of 22 g. (0.1 mole) of II, 17.2 g. (0.1 mole) of *p*-nitrobenzyl chloride, 15 g. (0.108 mole) of K_2CO_3, and 0.5 g. of NaI in 60 ml. of acetone was stirred and refluxed for 8 hr. Work-up as described for III yielded 29.4 g. (84%) of pale yellow crystals, m.p. 193~194°. The yield is comparable to those reported for similar reaction with more basic NaOEt (84[29] or 88%[21]). Recrystallization first from aq. AcOH and then from EtOAc gave practically colorless, fine needles, m.p. 193~194° (lit. 196~197°;[29] 193~194°;[21] 190°[30]). *Anal.* Calcd. for $C_{16}H_{20}O_7N_2$: C, 54.54; H, 5.72; N, 7.95. Found: C, 54.55; H, 5.68; N, 7.93.

N-Acetyl-3-(3,4-methylenedioxyphenyl)alanine——A mixture of 31 g. of the crude III and 14 g. of NaOH in 80 ml. of H_2O was heated for 1 hr. on a water bath with occasional shaking until dissolution (15~20 min.) and refluxed another 1 hr. Upon cooling the mixture was treated with some ice and 5 ml. of Ac_2O with swirling, decolorlized, and acidified carefully at 30~40° with dil. HCl. After standing overnight in a refrigerator the crystals were collected and washed with H_2O to give 23.5~24.5 g. (92~96%) of the acetamido acid, m.p. 178~180°. An analytical sample crystallized from 20% aq. AcOH as colorless needles, m.p. 177~179° (lit. 178~180°[31]). *Anal.* Calcd. for $C_{12}H_{13}O_5N$: C, 57.37; H, 5.22; N, 5.58. Found: C, 57.43; H, 5.45; N, 5.62.

N-Acetyl-*p*-nitrophenylalanine——Alkaline hydrolysis of ethyl acetamido(*p*-nitrobenzyl)cyanoacetate (VII) was not so successful as that of the malonate analog VI (with Na_2CO_3[32] or NaOH[33]). Heating 30.5 g. of crude VII with 5 volumes of 25% HCl at reflux gave a clear solution, which deposited a copious precipitate of fine needles in 1~2 hr. After 4 hr. reflux, the precipitate of *p*-nitrophenylalanine hydrochloride was collected (22.6 g., 91.5%; a second crop from the mother liquor is negligible) and acylated by alternate addition of 13 ml. of Ac_2O and aq. NaOH to give 22.2 g. (88%, based on VII) of yellowish, fine needles, m.p. 198~200°. Recrystallization from 25% aq. AcOH raised the m.p. to 200~201° (lit. 207~209°;[32] 203.5~204°;[29] 190~192°[34]). *Anal.* Calcd. for $C_{11}H_{12}O_5N_2$: C, 52.38; H, 4.80; N, 11.11. Found: C, 52.44; H, 4.28; N, 10.81.

25) S. Tatsuoka, T. Kinoshita, R. Nakamori: Yakugaku Zasshi, **71**, 702 (1951).
26) Y. Izumi, S. Konishi: Nippon Kagaku Zasshi, **74**, 22 (1953).
27) Y. Matsuda, I. Matsumoto: Yakugaku Kenkyu, **29**, 508 (1957).
28) S. Kobayashi: Sci. Papers Inst. Phys. Chem. Res., **6**, 165 (1927); cf. R. Schwarz, K. Capek: Monatsh. Chem., **84**, 595 (1953).
29) P. Block, Jr.: J. Org. Chem., **21**, 1237 (1956).
30) D. F. Elliott, C. Harington: J. Chem. Soc., **1949**, 1374.
31) S. Yamada, T. Fujii, T. Shioiri: This Bulletin, **10**, 680 (1962).
32) F. Bergel, V. C. E. Burnop, J. A. Stock: J. Chem. Soc., **1955**, 1223.
33) A. L. Davis, J. M. Ravel, C. G. Skinner, Wm. Shive: Arch. Biochem. Biophys., **76**, 139 (1958).
34) W. L. Bressler, L. S. Ciereszko: J. Am. Chem. Soc., **77**, 2330 (1955).

2,6-Dichlorobenzyl Chloride——Reduction of 40 g. of 2,6-dichlorobenzaldehyde (m.p. 69～71°) in aq. EtOH at 20～25° with 4 g. of $NaBH_4$, instead of $LiAlH_4$,[35] yielded 39 g. of 2,6-dichlorobenzyl alcohol (Ⅶ), m.p. 97～98° (lit. 97～98.5°[35]). A solution of 33 g. of Ⅶ in 60 g. of $SOCl_2$ was treated with 0.5 ml. of pyridine, slowly heated and refluxed until the gas evolution practically ceased, poured into H_2O, and the solid recrystallized from MeOH to give 30 g. of colorless needles, m.p. 39～39.5° (lit. 39～40°;[36] 49～50°[37]), and 5 g. of a second crop, m.p. 37～39°. When the pyridine was omitted, Ⅶ was recovered unchanged even after prolonged heating. The reaction without pyridine as a catalyst[38] seems not to proceed beyond the chlorosulfite stage, presumably due to the steric factor.

3,4-Methylenedioxybenzyl Chloride——Hydrogenation of piperonal in MeOH at 120° over Raney Ni and trituration of the evaporated residue with petr. benzin gave colorless Ⅴ, m.p. 53～55°, in 90～96% yields. Addition of 58 g. of Ⅴ all at once at 20～25° to 116 ml. of stirred concd. HCl caused Ⅴ crystals to melt with a temperature drop of 2～3° and the emulsion, when kept at <20°, congealed in 5～10 min., remelting at 22°. After stirring for 30 min., the mixture was extracted with 50 and 30 ml. of benzene. The combined extracts were washed successively with 20% aq. NaCl and 10% cold aq. K_2CO_3, dried over anhyd. K_2CO_3, and immediately distilled to give 57～61 g. (88～94%) of colorless oil, $b.p_{13}$ 129°, which was used within 1～2 days because of the instability on storage. The alkali washing is important, because the presence of a little acid during the distillation is liable to cause the chloride Ⅳ to resinify with evolution of HCl,[39] which, once occurred, further accelerates the degradation.

4-Chloromethyl-4′-nitrodiphenyl Ether——Recrystallization twice from 95% EtOH gave pale yellow needles, m.p. 60.5～61° instead of the reported value 54～55°.[40] *Anal.* Calcd. for $C_{13}H_{10}O_3NCl$: C, 59.22; H, 3.82; Cl, 13.45. Found: C, 59.37; H, 3.97; Cl, 13.65.

2-Chloromethyl-6-methylpyridine Hydrochloride——The following modification is suitable for large-scale preparation. A solution of 60 g. of 6-methyl-2-pyridinemethanol (hygroscopic crystalline mass) in 30 g. of toluene was added dropwise to 70 g. of $SOCl_2$ with stirring and cooling below 20° and the temperature raised slowly to 75° in 30 min. to give a thick slurry, which was treated with 20 ml. of anhyd. EtOH at 50° to destroy the excess $SOCl_2$, refluxed another 30 min., cooled, and filtered. The cake was washed with acetone to give 73 g. (84%) of colorless needles, m.p. 154～155° (lit. 146～153°;[41] 152～154°;[42] 155～156°[43]). The acetone washing is very effective to remove resinous impurities. Unlike 2-chloromethylpyridine the free base,[44] m.p. 18～19°, can be stored without discoloration over 6 months when kept frozen at 10°.

35) R. N. Castle, J. L. Riebsomer: J. Org. Chem., **21**, 142 (1956).
36) P. R. Austin, J. R. Johnson: J. Am. Chem. Soc., **54**, 647 (1932).
37) H. C. McBay, O. Tucker, P. T. Groves: J. Org. Chem., **24**, 536 (1959).
38) W. Gerrard, K. H. V. French: Nature, **159**, 263 (1947); G. Machell: Chemical Products, **19**, 356 (1956).
39) E. M. Schultz, R. T. Arnold: J. Am. Chem. Soc., **71**, 1913 (1949).
40) P. L. Southwick, G. E. Foltz, W. E. McIntyre, Jr.: *Ibid.*, **75**, 5877 (1953).
41) T. Kato: Yakugaku Zasshi, **75**, 1236 (1955).
42) W. Baker, K. M. Buggle, J. F. W. McOmie, D. A. M. Watkins: J. Chem. Soc., **1958**, 3594.
43) W. Mathes, H. Schüly: Angew. Chem., **75**, 235 (1963).
44) E. Matsumura, T. Hirooka, K. Imagawa: Nippon Kagaku Zasshi, **82**, 616 (1961).