solution which was approximately 0.001 *N* with respect to $HClO_4$. It was standardized by titrating it against 0.00102 g (5.0×10^{-6} mole of potassium acid phthalate in 50 ml of glacial AcOH) using a Corning Model 12 pH meter equipped with glass and calomel electrodes. Readings on the + mV scale were recorded for the corresponding milliliters of titrant. Milliliters of titrant were then plotted against + mV readings, and the end point was determined graphically. The standardized $HClO_4$ solution was then used for similar potentiometric titrations of bisbenzimidazoles (5.0×10^{-6} mole of bisbenzimidazole in 50 ml of glacial AcOH). The titration was carried out until the first break in the curve was obtained and this value corresponded to the protonation of one of the N atoms in the molecule. Because of the ease of solvent trapping in these compounds, titration with this method is considered the most reliable method for purity determination.

HeLa Cell Alkylation Study.—HeLa cells were harvested in Leighton tubes containing cover slips and inoculated with *trans*-Cl-Me-DBE. At a concentration of 10^{-5} to 10^{-6} *M* ml, the effect could clearly be seen *in vivo*. At 2–8 hr the nuclear membrane started to show fluorescence as well as about 15% of the nuclei of the cell population (variation depending on original culture), and, at the end of 24 hr, numerous cells appeared fluorescent in their nuclei (Figure 1). Thus, it was felt that in spite of the low solubility of the compound, the incorporation of a fluorescent alkylating agent into the nucleus was found to be possible. Efforts are being made to correlate fluorescence with various mitotic phases in synchronous population. Our present contention is satisfied with the knowledge that *in vivo* alkylation of nucleus, indeed, occurred with a fluorescent-labeling alkylating agent.

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Derivatives of Fluorene. XXX.[1] Rearrangement and Antitumor Activities of Some 9-Oxofluorene Oximes. 6(5H)-Phenanthridinones. I

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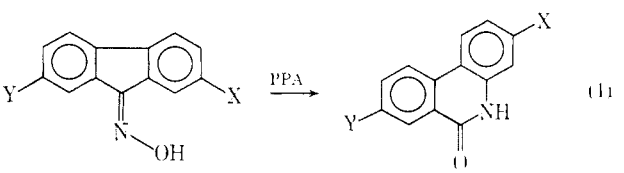
Rearrangement of 9-oxofluorene oximes in polyphosphoric acid (PPA) to the corresponding 6(5H)-phenanthridinones is described. Reaction of 1-iodo- and 1-nitro-9-oxofluorene oxime with PPA gave, instead of the expected phenanthridinones, the corresponding 9-oxofluorenes. Results of screening for antitumor activities are presented.

Because interesting antitumor activity was shown earlier[2] by a number of polyhalogenated fluorene derivatives, we have prepared a number of structurally related compounds with altered properties, *e.g.*, increased hydrophilicity, which might enhance the biological effects of these compounds. One such series is a group of oximes of 9-oxofluorenes (Table I).[3] Several of these have shown activity against Walker carcinosarcoma 256 (see Table II). A further reason for our interest in these oximes is that rearrangement to phenanthridinones (Table I) gives a heterocyclic system which has had few derivatives screened for antitumor activity. We are particularly interested in polyhalogenated phenanthridinones analogous to the active compounds in the fluorene series.[2] This paper is the beginning of such a study.

The oximes were prepared in DMSO, by an improved procedure,[3] or in the conventional way by treating the 9-oxofluorene with 2 equiv of hydroxylamine hydrochloride in refluxing 70% EtOH. The rearrangement of the 9-oxofluorene oximes was carried out in polyphosphoric acid[4] (PPA) at elevated temperatures.

Although the oxime of 3-nitrofluorenone in PCl_5–$POCl_3$ rearranged to a single compound, 2-nitrophenanthridinone,[5] monosubstituted 9-oxofluorene oximes, in general, rearrange to a mixture of the two isomers, difficult to separate. Even in PPA such mixtures are to be expected; however, in our work, 2-nitrofluorenone oxime gave a fair yield of only one product, 3-nitrophenanthridinone.

A series of 2,7-disubstituted fluorenone oximes, with both substituents the same, gave good yields of 3,8-disubstituted phenanthridinones (eq 1) when they were heated for 15 min at temperatures above 180°. It was reported earlier[6] that fluorenone oximes did not rearrange at temperatures of 100–150°, effective for many oximes.



(1)

Ia. X = NO_2; Y = H
b. X = Y = F
c. X = Y = Cl
d. X = Y = Br
e. X = Y = NO_2

In spite of the two paths followed in the Beckmann rearrangement of many of these monosubstituted oximes, it was hoped that a bulky substituent, such as iodo or nitro, at the 1 position of the fluorene nucleus would lead to a single product, hopefully a 4-substituted 6(5H)-phenanthridinone. However, the only identifiable product obtained from each of these reactions was the corresponding 1-substituted 9-oxofluorene (eq 2).

(1) This work was supported in part by Grant CA-01744 and in part by Career Development Award 9-K3-CA-14,991 (T. L. F.) from the National Cancer Institute, National Institutes of Health.

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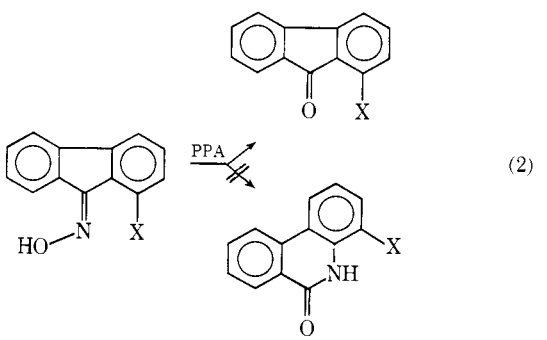
TABLE I: SUBSTITUTED 9-OXOFLUORENE OXIMES AND 6(5H)-PHENANTHRIDINONES

Substituent(s)	Mp, °C	Reaction solvent or temp, °C	Reaction time, hr	Yield, %	Formula	Analyses
			9-Oxofluorene Oxime			
1-NO_2	205–206	A[a]	44	98	$C_{13}H_8N_2O_3$	C, H, N
2-Br	197–198[b]	A	1	98	$C_{13}H_8BrNO$	C, H, N
2-NH_2-3-Cl	232–233	A	2	100	$C_{13}H_9ClN_2O$	C, H, N
2-NH_2-3-Br	226–227	A	2	98	$C_{13}H_9BrN_2O$	C, H, N
2-NH_2-7-Br	226–227	A	3	97	$C_{13}H_9BrN_2O$	C, H, N
2-NH_2-3-Br-7-NO_2	272–273	A	19	99	$C_{13}H_8BrN_3O_3$	C, H, N
2-$NHCOCH_3$-3-NO_2	275–276	B[c]	0.2	100	$C_{15}H_{11}N_3O_4$	C, H, N
2-F-7-NO_2	248–249	A	2	98	$C_{13}H_7FN_2O_3$	C, H, N
2-Cl-7-NO_2	264–264.5	A	24	100	$C_{13}H_7ClN_2O_3$	C, H, N
2-Br-7-NO_2	246–247	A	1.5	100	$C_{13}H_7BrN_2O_3$	C, H, N
3-Br-2-NO_2	235–236	A	3	81	$C_{13}H_7BrN_2O_3$	C, H, N
2,7-F_2	251–252	B	0.2	98	$C_{13}H_7F_2NO$	C, H, N
2,7-$(NO_2)_2$	288–289[d]	B	0.2	100	$C_{13}H_7N_3O_5$	N
2,7-Cl_2-4-NH_2	264–265	A	2	100	$C_{13}H_8Cl_2N_2O$	C, H, N
2,3-Cl_2-7-NO_2	260–261	A	2	96	$C_{13}H_6Cl_2N_2O_3$	C, H, N
2-NH_2-1,3,4,7-Cl_4	262–262.5	B	0.2	89	$C_{13}H_6Cl_4N_2O$	N
2-$NHCOCF_3$-3-Br	228.5–229.5	B	0.2	100	$C_{15}H_8F_3BrN_2O_2$	C, H, N
2-$NHCOCF_3$-7-NO_2	288.5–289.5	B	0.2	100	$C_{15}H_8F_3N_3O_4$	C, H, N
			6(5H)-Phenanthridinone			
3,8-F_2	311–312	185–190	0.1[e]	60	$C_{13}H_7F_2NO$	C, H, N
3,8-Cl_2	348–349	180–185	0.1[e]	46	$C_{13}H_7Cl_2NO$	C, H, Cl, N
3,8-Br_2	320–321	195–200	0.25[e]	60	$C_{13}H_7Br_2NO$	C, H, N
3,8-$(NO_2)_2$	354–355[f]	220–225	0.25[e]	100	$C_{13}H_7N_3O_5$	C, H, N

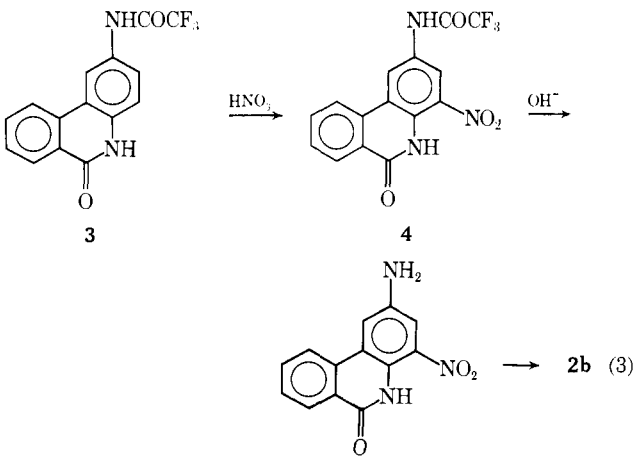
[a] A = 70% EtOH. [b] C. Courtot and C. Vignati, *Bull. Soc. Chim. France*, **41**, 58 (1927), reported mp 194–195°. [c] B = DMSO. [d] J. Schmidt and K. Bauer, *Ber.*, **38**, 3737 (1905), reported mp *ca.* 285–286°. [e] In polyphosphoric acid. [f] C. L. Arcus, M. M. Coombs, and J. V. Evans, *J. Chem. Soc.*, 1498 (1956), reported mp >380°.



(2)

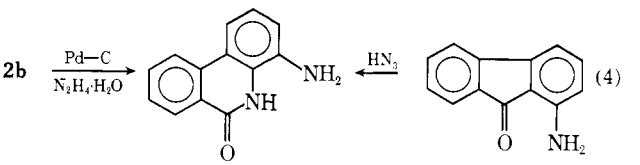
2a, X = I
b, X = NO_2

Compound **2b** was prepared from 2-amino-6(5H)-phenanthridinone by trifluoroacetylation of this amine to **3** (eq 3). The latter was nitrated with HNO_3 in AcOH to 4-nitro-2-trifluoroacetamido-6(5H)-phenanthridinone (**4**). Alkaline hydrolysis of **4** gave 2-amino-4-nitro-6(5H)-phenanthridinone (**5**) which was deaminated to give **2b**.



3 **4** **5** **2b** (3)

Reduction of **2b** gave 4-amino-6(5H)-phenanthridinone (**6**) which was also obtained from a Schmidt reaction on 9-oxofluoren-1-amine (eq 4).



(4)

6

The oximes and 6(5H)-phenanthridinones were tested in mice (with L1210 and S180) or in rats (with W256) for antitumor activity through the Cancer Chemotherapy National Service Center, National Institutes of Health. Significant results are given in Table II. All the other compounds tested were inactive.

Experimental Section[7]

Substituted 9-Oxofluorene Oximes. A.—A mixture of the 9-oxofluorene and $HONH_2 \cdot HCl$ (2 equiv) was refluxed in 70% EtOH (0.2–0.5 l./0.01 mole of the ketone) then most of the solvent was distilled off. After H_2O dilution and basification with dilute $NaHCO_3$ the product was isolated and recrystallized, if needful, from EtOH, Me_2CO, or PhMe.

(7) All melting points below 250° were taken on a Fisher-Johns block and are corrected to standards. The melting points above 250° were taken with a Hoover capillary melting point apparatus and are uncorrected. Where analyses are indicated only by symbols of the elements, analytical results obtained for those elements were within ±0.4% of the theoretical values. Analyses were performed by A. Bernhardt, Elbach über Engelskirchen, West Germany, and by Schwarzkopf Laboratories, Woodside, N. Y.

TABLE II

ANTITUMOR ACTIVITY OF SUBSTITUTED 9-OXOFLUORENE OXIMES AND 6(5H)-PHENANTHRIDINONES[a]

Substituent(s)	Test system	Daily dose, mg/kg	Survivors	Tumor wt, mg, or survivor days (T/C's	% T/C	Confidence, %[b]
		9-Oxofluorene Oximes				
2-Br	L1210[c]	200.0	4/4	10.0 8.5	117	
	W256[d]	400.0	6/6	4.0/5.3	75	
2,7-Cl_2	S180[e]	90.0	6/6	518/901	57	
		60.0	6/6	555/901	61	
		40.0	6/6	643/901	71	
		26.6	5/6	695/901	77	
						95.0
	W256	150.0	6/6	5.0 7.9	63	
		100.0	6/6	5.6/7.9	70	
		50.0	6/6	7.3 7.9	92	
		25.0	6/6	6.0/7.9	75	
2,7-Br_2	W256	400.0	6/6	2.3 6.0	38	
2-NH_2-3-Br	W256	400.0	6/6	4.5 6.2	72	
2-NH_2-7-Br	W256	400.0	6/6	2.7 5.3	50	
		400.0	5/6	2.3 5.6	41	
		200.0	6/6	4.1 5.6	73	
		100.0	6/6	4.4 5.6	78	
2-NH_2-3-Cl	W256	400.0	6/6	4.2 6.2	67	
2-Cl-7-NO_2	W256	500.0	6/6	3.0 5.8	51	
		400.0	6/6	2.9 5.8	50	
		200.0	6/6	5.1 5.8	87	
		100.0	6/6	4.5 5.8	77	
						95.0
2-Br-7-NO_2	W256	400.0	6/6	2.2 5.4	40	
		400.0	6/6	2.6/6.2	41	
		400.0	6/6	3.7/6.6	56	
2,7-Cl_2-4-NH_2	W256	400.0	6/6	4.0 5.3	75	
2,7-Cl_2-4-NO_2[f]	W256	600.0	6/6	1.6 5.8	27	
		400.0	6/6	3.6 5.8	62	
		200.0	6/6	4.2 5.8	72	
		100.0	6/6	5.0 5.8	86	
						95.0
2-$NHCOCF_3$-3-Br	W256	400.0	6/6	4.8 6.5	73	
2-$NHCOCF_3$-7-NO_2	W256	400.0	6/6	4.7 6.5	72	
2-NH_2-1,3,4,7-Cl_4	W256	200.0	6/6	0.7/6.0	11	
		100.0	6/6	2.2/6.0	36	
		50.0	6/6	4.3 6.0	71	
2,4,7-Cl_3[g]	W256	500.0	6/6	2.3 5.8	39	
		400.0	6/6	2.0 5.8	34	
		200.0	5/6	2.5 5.8	96	
		600.0	6/6	5.6 8.4	66	
		400.0	6/6	4.7 8.4	55	
		200.0	6/6	4.7 8.4	55	
		100.0	6/6	5.9 8.4	70	
						99.7
		6(5H)-Phenanthridinone				
3-NH_2	W256	400.0	6/6	4.0/6.2	64	
3,8-Cl_2	W256	400.0	6/6	3.2/5.7	56	
3,8-Br_2	W256	400.0	6/6	2.6 5.7	45	
		400.0	6/6	3.6 6.0	60	

[a] The screening data were kindly supplied by Dr. Harry B. Wood, Jr., of the Cancer Chemotherapy National Service Center, National Institutes of Health, Bethesda, Md. Assays were performed according to CCNSC specifications as published in *Cancer Chemotherapy Rept.*, **25**, 1 (1962). [b] A compound having confidence at the 99.7% level is considered truly specific. For details see H. E. Skipper, W. S. Wilcox, F. M. Schabel Jr., W. R. Laster, Jr., and L. Mattill, *Cancer Chemotherapy Rept.*, **29**, 1 (1963). [c] Lymphoid leukemia was tested in BDF_1 mice. [d] Walker carcinosarcoma (intramuscular) was tested in random-bred albino rats. [e] Reference 3. [f] Sarcoma 180 was tested in Swiss mice.

B.—The oxofluorene was dissolved in hot DMSO (minimum amount). To the stirred hot mixture saturated aqueous $HONH_2 \cdot HCl$ (1.1 equiv) was added in one portion. This was heated at 90–95° for a short period and diluted with H_2O. The product was isolated in the usual manner.

Substituted 6(5H)-Phenanthridinones (1a–e).—The 9-oxofluorene oxime was mixed with 40–50 times its weight of PPA. The mixture was heated with constant stirring for 0.1–0.25 hr, cooled, and triturated in H_2O and the product was isolated and purified by recrystallization from a suitable solvent, *e.g.*, AcOH.

2-α,α,α-Trifluoroacetamido-6(5H)-phenanthridinone (3).—2-Aminophenanthridinone[8] (21 g, 0.1 mole) was trifluoroacetylated in CH_2Cl_2 (1.5 l.) with trifluoroacetic anhydride (60 ml) giving 27.6 g (90%), mp 329–330°. Recrystallization from Me_2CO gave an analytical sample, mp 330–331°. *Anal.* ($C_{15}H_9F_3N_2O_2$) C, H, N.

2-α,α,α-Trifluoroacetamido-4-nitro-6(5H)-phenanthridinone (4).—To a stirred suspension of **3** (9.2 g, 0.03 mole) in AcOH

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(120 ml), HNO_3 (*d* 1.42) (8 ml) was slowly run in at 45 ± 2°. After stirring at this temperature for a few minutes, H_2SO_4 (2 ml) was added to the reaction mixture in several portions. Stirring was continued at 55–60° for 15 min and cooled, and the product was filtered off giving 10.2 g (97%). Recrystallization from EtOH gave yellow needles, mp 308–309°. *Anal.* ($C_{15}H_8F_3N_3O_4$) C, H, N.

2-Amino-4-nitro-6(5H)-phenanthridinone (5).—A solution of KOH (1.1 g) in H_2O (5 ml) was added in one portion to a boiling suspension of **4** (3.1 g, 9 mmoles) in 95% EtOH (400 ml). The solution was boiled until crystallization of the product took place. The rest of the solvent was then driven off without heat by an air stream. The solid was triturated in H_2O and collected by filtration, 2 g (87%). Recrystallization from PhMe gave an analytical sample, mp 308–309° dec. *Anal.* ($C_{13}H_9N_3O_3$) C, H, N.

N-Acetyl derivative melted at 284–285° (AcOH). *Anal.* ($C_{15}H_{11}N_3O_4$) C, H, N.

4-Nitro-6(5H)-phenanthridinone (2b).—Deamination of **5** with H_3PO_2 (50%) gave yellow needles (C_6H_6–EtOH), mp 259–260° (lit.[9] mp 264–265°). *Anal.* ($C_{13}H_8N_2O_3$) C, H, N.

4-Amino-6(5H)-phenanthridinone (6). By Rearrangement of 9-Oxofluoren-1-amine.—Saturated aqueous NaN_3 (20 g) was added dropwise to a stirred and ice-cooled mixture of 9-oxofluoren-1-amine[10] (30 g) and H_2SO_4 (200 ml) over a period of 2.5 hr. After 22 hr of stirring at ambient temperature the reaction mixture was diluted with ice–water (200 ml). The amine sulfate was collected, treated with excess 5% NaOH, and the product, 28 g (87%), was recrystallized from EtOH giving lustrous crystals, mp 311.5–312.5°. *Anal.* ($C_{13}H_{10}N_2O$) C, H, N.

By Reduction of 2b.—A suspension of **2b** (1.4 g), 85% $N_2H_4 \cdot H_2O$ (3 ml), and 5% Pd–C (50 mg) in EtOH (100 ml) was gently refluxed for 5 hr and filtered, and the filtrate was concentrated giving 0.9 g, melting point and mixture melting point with the above compound showed no depression.

2,4-Diamino-6(5H)-phenanthridinone.—Reduction of **5** the same way as described above gave the diamine (70%), mp 310–311°. *Anal.* ($C_{13}H_{11}N_3O$) C, H, N.

Conversion of 1-Iodo-9-oxofluorene Oxime to 1-Iodo-9-oxofluorene in PPA.—1-Iodo-9-oxofluorene oxime[3] (0.5 g) was mixed with PPA (25 g). The mixture was heated at 125–130° for 15 min, cooled, and diluted (H_2O). The yellow solid was recrystallized from EtOH and then chromatographed in C_6H_6 through an alumina column giving 0.3 g of 1-iodo-9-oxofluorene[10,11] (melting point and mixture melting point).

Conversion of 1-Nitro-9-oxofluorene Oxime to 1-Nitro-9-oxofluorene in PPA.—Similarly 1-nitro-9-oxofluorene oxime (0.5 g) and PPA (25 g) were heated at 120–125° for 15 min and treated with H_2O. After chromatography on alumina (C_6H_6), 1-nitro-9-oxofluorene[12] (melting point and mixture melting point) was obtained.

4-Iodo-6(5H)-phenanthridinone (2a).—Saturated aqueous $NaNO_2$ (3.5 g, 0.05 mole) was added portionwise to a stirred mixture of **6** (6.3 g, 0.03 mole), H_2SO_4 (60 ml), and H_2O (120 ml) at 5–10° (15 min). After stirring at 0–5° for 1.5 hr, excess HNO_2 was destroyed by means of urea (1.2 g). A cold (5°) solution of KI (48 g), I_2 (24 g), and H_2O (50 ml) was then added all at once to the diazotization mixture, which was allowed to stand overnight, heated for 15 min on a steam bath, and diluted with H_2O. The product was filtered off and treated with dilute $Na_2S_2O_3$ giving 7.6 g (83.5%). Chromatography on alumina with C_6H_6 as eluent gave lustrous platelets, mp 243–244°. *Anal.* ($C_{13}H_8INO$) C, H, I, N.

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(12) R. H. Chase and D. H. Hey, *J. Chem. Soc.*, 553 (1952).

Potential Carcinolytic Agents. VII. Substituted Bis(2-methanesulfonoxyethyl)anilines[1a]

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New nuclear-substituted (3-acetamido, -amino, -carbethoxy, -chloro, -fluoro, -methyl, and -trifluoromethyl and 4-amino, -nitro, and -nitroso) bis(2-methanesulfonoxyethyl)anilines have been prepared by (1) N-hydroxyethylation of an appropriately ring-substituted aniline with ethylene oxide, (2) esterification of the hydroxyl groups with methanesulfonyl chloride, and (3) further ring substitution (nitrosation or nitration). The compounds were evaluated for antitumor activity and the pertinent results are reported. N,N-Bis(2-methanesulfonoxyethyl)-*p*-nitrosoaniline reported previously is still the most active compound in the series.

Earlier we reported[2,3] the high antitumor activity of N,N-bis(2-methanesulfonoxyethyl)-*p*-nitrosoaniline (**20**) against a variety of animal tumors. The most significant activity of **20** was shown against Walker carcinosarcoma 256 (intramuscular), Dunning leukemia (ascites), and against the cytoxan- and thiopurine-resistant strains of Dunning leukemia (ascites). It was also effective against intracerebral Dunning leukemia and had an ED_{50} in the order of 10^{-4} μg/ml in KB and L1210 cell cultures. In a mitotic index study using L1210 cell culture, the compound was found to be a potent inhibitor of cell division.[4] Preclinical toxicology studies of **20** unfortunately showed that dogs and monkeys developed leukopenia and congestive heart failure at doses of about 0.25 mg/kg.[5]

Chemistry.—Because of the interesting biological properties of **20** and in the hope of finding a compound of even higher activity, we undertook a program to synthesize a series of related compounds. These were prepared *via* the straightforward route illustrated in Scheme I. The substituted anilines I were hydroxyethylated with ethylene oxide[6,7] to the N,N-bis(2-

(1) (a) Presented in part at the First Northeast Regional Meeting of the American Chemical Society, Boston, Mass., Oct 13–15, 1968. The work was sponsored by the Cancer Chemotherapy National Service Center, National Cancer Institute, National Institutes of Health, Contract No. PH-43-66-502. Paper VI: Z. B. Papanastassiou, R. J. Bruni, and E. White, V, *Experientia*, **24**, 325 (1968). (b) Deceased. (c) To whom inquiries should be addressed.

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