

LEAD

ANNUAL SURVEY COVERING THE YEAR 1983

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I. Books and Reviews

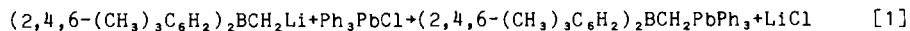
A number of reviews dealing with organolead chemistry have been published: annual survey 1982 (1); group IV organometallic chemistry 1981 (2); data on alkynyllead (IV) and related derivatives of alkynyltin (IV) compounds (3); the behaviour of the lone pair of Pb in Pb(II) oxides (4); gas chromatography (5); analysis of alkyllead compounds in air (6); leadfree gasolines (7); alkylation of heavy metals and the environment (8); environmental pollution with lead and the relevance of experimental animal studies to marginal lead toxicity in children (9).

The highly important field of low level lead exposure was covered by a range of specialists in a book edited by Rutter and Jones: sources of lead exposure, toxicity of lead and neuropsychological effects of lead (10). The varying contributions are garnished by vivid discussions.

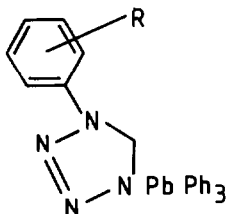
The "Fourth International Conference on the Organometallic and Coordination Chemistry of Germanium, Tin and Lead" was held in Montreal (11).

II. Syntheses

The industrial organic electrosynthesis of e.g. Me_4Pb has been discussed (12). The preparation of Et_4Pb from Pb-Na alloy and EtCl has been investigated microgravimetrically (13). Me_3Pb (3-furyl) has been obtained as the reaction product of 3-furyllithium and Me_3PbCl . The compound has been studied by ^1H -, ^{13}C - and ^{207}Pb -NMR (14). (Dimesitylboryl) (triphenylplumbyl) methane has been synthesized according to [1] in a yield of at least 60% (15).



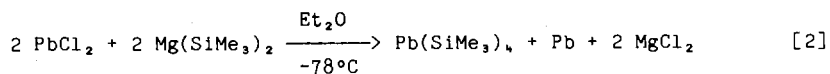
Organolead tetrazoles (I) have been prepared starting from Ph_3PbN_3 , CS_2 and $\text{RC}_6\text{H}_4\text{NCS}$ ($\text{R}=\text{H}$, $p\text{-Br}$, $o\text{-Me}$, $p\text{-Me}$) (16).



I

The compounds $(\text{Ph}_3\overset{\oplus}{\text{P}}\text{CHR} - \text{MPh}_3) \text{Cl}^-$ ($\text{R}=\text{H}$, Me , Me_2CH ; $\text{M}=\text{Ge}$, Sn , Pb) and $(\text{Ph}_3\overset{\oplus}{\text{P}}\text{CHR}) \text{MCl}_n^-$ ($\text{M}=\text{Ge}$, $n=4$; $\text{M}=\text{Pb}$, $n=2$) have been isolated (17). The ylide in $(\text{Ph}_3\overset{\oplus}{\text{P}}\text{CH}_2^- \text{PbPh}_3) \text{Cl}^-$ is bonded to the Pb atom through the carbanionic donor atom.

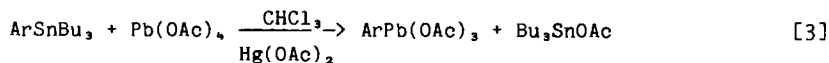
The first organosilicon-lead compound has been synthesized (18). The IR spectrum showed PbSi_4 valence vibrations at 305 and 302 cm^{-1} . $\text{Pb}_2(\text{SiMe}_3)_6$ was detected mass spectroscopically to be an intermediate in the reaction [2].



The reduction of Me Et ketone at Pb-Sn alloy cathodes leads to the formation

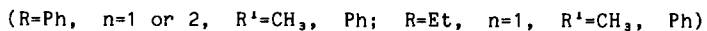
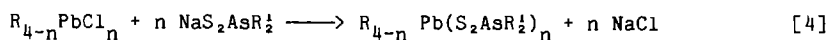
of polyalkylated metallic chains comprising Sn-Pb intermetallic bonds (19).

Aryllead (IV) triacetates can be prepared by Hg(II) catalyzed arylation of $\text{Pb}(\text{OAc})_4$ by ArSnBu_3 in 69-90% yields (20). Starting from 6-methoxy-2-naphthyllead triacetate obtained according to [3], the anti-inflammatory drug naproxen was synthesized.



The compounds $(\text{CH}_3)_3\text{PbCF}_3$, $(\text{CH}_3)_2\text{Pb}(\text{CF}_3)_2$ and $\text{CH}_3\text{Pb}(\text{CF}_3)_3$ have been prepared in 0.2-1.9% yields from $(\text{CH}_3)_3\text{Pb}^\cdot$ and $\cdot\text{CF}_3$ radicals produced in a radio-frequency discharge of hexafluoroethane. NMR data were given: $^2\text{J } ^{207}\text{Pb}-^1\text{H}$ 72.0, 88.5 and 94.0 Hz; $^2\text{J } ^{207}\text{Pb}-^{19}\text{F}$ 263, 403 and 601 Hz (21).

Organolead dithioarsinic acids have been prepared according to [4] and characterized by IR and $^1\text{H-NMR}$ (22).



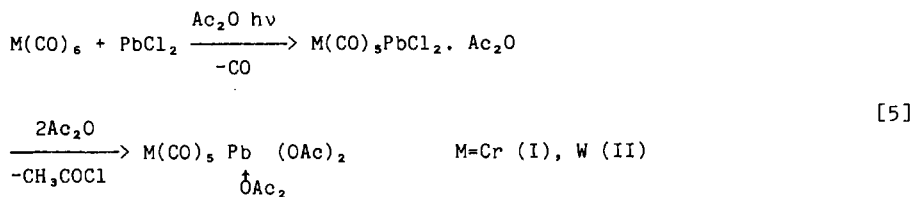
While Si and Ge compounds contain monodentate dithio groups, bidentate ligands occur in the organolead compounds. The preparation and properties of Ph_3PbSeCN have been reinvestigated (23). Adducts (1:1) with several ligands were reported e.g. a selenocyanate with TPPO but an isoselenocyanate with HMPA. N-bonded as well as Se-bonded isomers were found, whereas with $\text{Ph}_3\text{Pb NCS}$ only N-bonded adducts occurred. R_nN ($\text{R}=\text{Me}, \text{Et}$), Ph_nP and Ph_nSb salts of diaryllead (IV) mixed halogeno anions of the type $[\text{Ph}_2\text{PbXYZ}]$, $[\text{Ph}_2\text{PbXY}(\text{Z})_2]^{2-}$ and $[\text{Ph}_2\text{PbPbX}_3\text{Y}]^{2-}$ ($\text{XY}=\text{ClBr}, \text{ClI}, \text{BrI}, \text{Z}=\text{Cl}, \text{Br}, \text{I}, \text{N}_3, \text{NCS}$) have been synthesized (24). Conductance measurements have been performed. Complexes of lead with benzeneseleninato ligands have been described (25). Organolead halides form complexes with 1,3-dimethyl-2-imidazolidinone (DMEU) and 1,3-dimethyl-3,4,5,6-tetrahydro-2(1H)-pyrimidinone (DMPU) (26): $\text{Ph}_3\text{PbBr} \cdot \text{DMEU}$, $\text{Ph}_3\text{PbI} \cdot \text{DMEU}$, $\text{Ph}_3\text{PbBr} \cdot \text{DMPU}$, $\text{Ph}_3\text{PbI} \cdot \text{DMPU}$, $3\text{Ph}_3\text{PbCl} \cdot 2 \text{DMEU}$ and $2 \text{Ph}_3\text{PbCl} \cdot \text{DMPU}$. The adducts were highly dissociated in CH_2Cl_2 solution as was indicated by IR, ^1H - and $^{207}\text{Pb-NMR}$ data. Ph_2PbI_2 formed complexes with substituted 2-aminothiazoles (27).

Compounds containing the chain Pb-Pb-Pb have been isolated from reaction mixtures by a freezing out procedure in order to prevent decomposition: the red solids $\text{Pb}_3(\text{o-Hex})_6$ and $\text{Pb}_3(\text{o-Tol})_6$, stable up to 140°C , show thermochromism. Mixed compounds such as $\text{Ph}_3\text{Pb-SiPh}_3$, $\text{Ph}_3\text{Pb-GePh}_3$ and

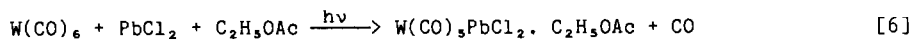
$\text{Ph}_3\text{Pb-SnPh}_3$ have been described. X-ray structural data have been obtained (28).

The anion Pb_5^{2-} with D_{3h} symmetry has been isolated through strongly complexing the Na or K ions with the polycyclic ligand 2,2,2-crypt (29).

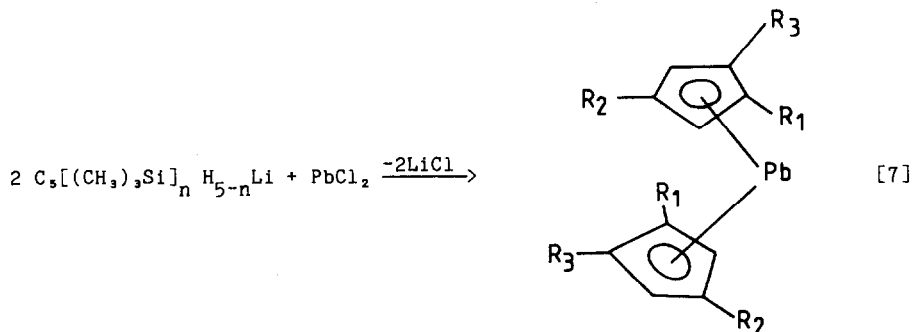
Cage compounds containing Pb(II) have been synthesized (30). The structures of the cage molecules are derived from a distorted cube, the corners of which are alternatively occupied by nitrogen or group IV elements. Group VI metal carbonyl complexes with plumbylenes have been prepared photochemically (31).



In ethyl acetate another complex was obtained:



The dark yellow to brown complexes were isolated in 18-21% yields; they decomposed at 105-120°C. The yellow dicyclopentadienyllead (II) compounds containing 2,4 or 6 trimethylsilyl groups have been synthesized according to [7] in hexane/THF at 0°C in yields of 7, 9.8 and 7.8% respectively (32).

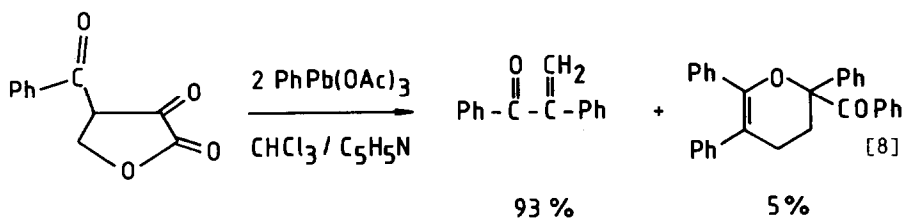


Organolead derivatives of biologically relevant compounds have been synthesized. Preparative procedures of organolead derivatives of amino acids and amino acid derivatives as well as structural data, biocidal and antitumour activity have been reported (33). Triorganolead derivatives of N-acetyl-

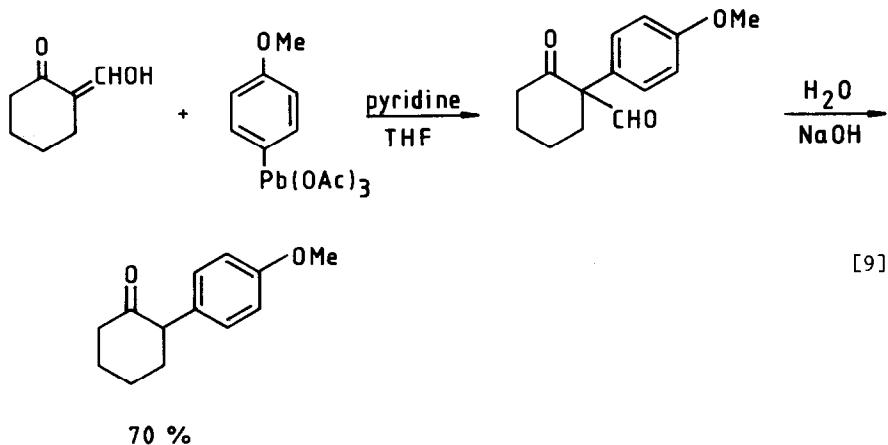
glycine, N-acetyl- α -alanine and N-acetylmethionine $\text{PbR}_2(\text{AcGlyO})$ ($\text{R}=\text{Me}$ or Ph), $\text{PbPh}_2(\text{AcAlaO})$, $\text{PbPh}_2(\text{AcMeO})$ have been obtained by reacting PbR_2OH ($\text{R}=\text{Me}$ or Ph) with the appropriate N-acetylamino acid in yields of 41-63%. Vibrational data indicated coordination of Pb by monodentate carboxylic groups and the oxygen of CO amide; in DMSO a five coordinated organolead species was formed (34).

III. Reactions

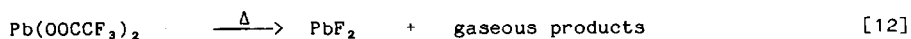
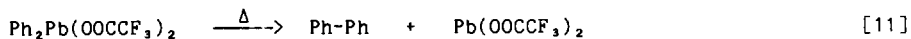
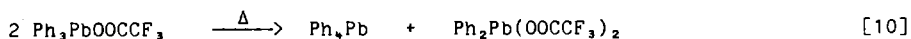
Aryllead (IV) tricarboxylates may be used in the synthesis of some α -aryl α,β -unsaturated carbonyl compounds (35). Treatment of 4-benzoyl-tetrahydrofuran-2,3-dione with 2 equivalents of $\text{PhPb}(\text{OAc})_3$ gave 1,2-diphenylpropenone in excellent yield.



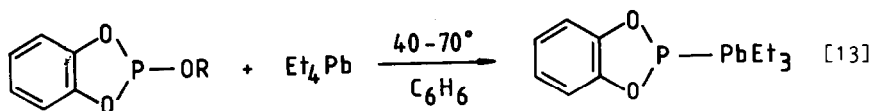
α -Hydroxymethylene ketone could be arylated with aryllead (IV) triacetates in pyridine/THF yielding α -arylketones after hydrolysis (36). NMR spectroscopic evidence supported the intermediacy of α -aryl α -formyl ketones.



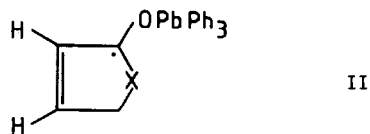
The thermolysis of phenyllead trifluoroacetates has been studied (37).



Attempts were made to prepare $\text{PhPb(OCCF}_3)_3$. The reaction of phenyllead plumbonic acid with trifluoroacetic acid led to $\text{PhPb(OCCF}_3)_2\text{OH}$ which thermally decomposed to PbF_2 . The thermolysis of several compounds i.a. Et_4Pb has been studied (38); a dissociation energy ΔH_{298} of $54 (\pm 2)$ kcal/mol was found. Et_4Pb has been applied to introduce the Et_3Pb group into organo-phosphorus compounds (39).

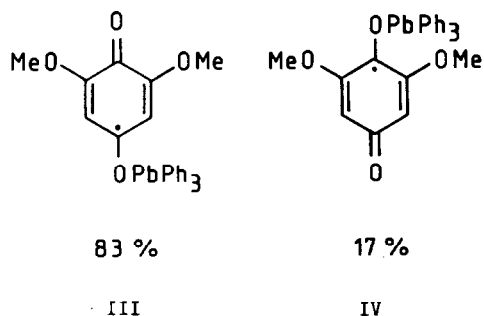


ESR data showed that even at low temperatures ($\leq 25^\circ\text{C}$) Ph_3Pb^+ gave spin adducts of the type (II) with maleic anhydride, maleic thioanhydride and maleimide (40).

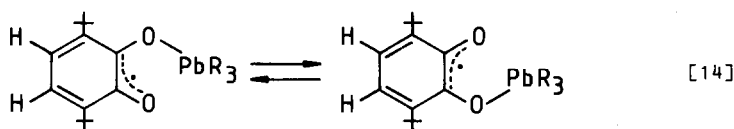


$\text{X} = \text{O, S, NH}$

ESR spectroscopy was also used in the study of the formation of adducts (III) and (IV) in the reaction of hexaphenyldilead with 2,6-dimethoxybenzoquinone in tert. butylbenzene/HMPTA (41). The formation of (IV) was explained by coordination of one of the adjacent oxygen atoms. No adducts with pyrazines could be obtained.



In an investigation on the fluxional behaviour of organotin derivatives of 3,6-di-*t*-butyl-1,2-benzoquinone some attention was paid to a few organo-lead compounds (42).



PbR_3 ($\text{R}=\text{Me}, \text{Ph}$) complexes carry monodentate semidione ligands that are fluxional down to low temperatures. The introduction of one chloro or one cyclopentadienyl ligand ($\text{R}_3=\text{Ph}_2\text{Cp}, \text{Me}_2\text{Cl}, \text{PhCl}_2$) reduces the fluxionality.

The reaction of alkali metal trialkylplumbanides with acetylene compounds was reported (43). The reaction of the OH radical with compounds R_4Pb has been studied (44,45). The decay of the OH absorption at 3087 Å in the reference system was simple 2nd order with a half life of 124 μs. Upon addition of a R_4Pb compound pseudo first order kinetics with much shorter half lives were found: $k(\text{OH} + \text{Me}_4\text{Pb}) = 3.8 \cdot 10^9/\text{mol s}$ and $k(\text{OH} + \text{Et}_4\text{Pb}) = 7.0 \cdot 10^9/\text{mol s}$. The Et_4Pb and Me_4Pb half lives under typical summertime conditions were 2 respectively 9 hrs. Et_3PbCl and Et_4Pb underwent successive dealkylation reactions under the influence of sunlight and UV irradiation in laboratory tests (46). Compounds such as 2PbCO_2 , $\text{Pb}(\text{OH})_2$, $\text{Pb}(\text{OH})_2 \cdot \text{PbCl}_2$ and BuEt_3Pb were formed. The photolysis of $[\text{CpFe}(\text{CO})_2\text{PbPh}_3]$ in solution has been described (47).

The electrochemical behaviour of Me_3Pb^+ at the mercury electrode in aqueous solution has been investigated by polarography, cyclic linear-sweep voltammetry, and differential pulse polarography (48). In the first 1-electron step the $\text{Me}_3\text{Pb}^\cdot$ radical is formed which irreversibly decomposes to the final product $\text{Pb}(\text{o})$.

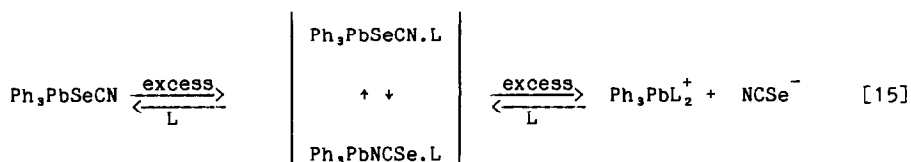
Methylcobalamine reacted with PbO_2 via a fast reaction with a short-lived methyllead intermediate that dissolved in water and subsequently decomposed (49).

The Diels-Alder cycloaddition was compared with the alkylmetal insertion under similar conditions (50). Both systems show transient charge-transfer absorption bands. The charge-transfer formulation of the activation process provides a unifying basis for comparing Diels-Alder cycloadditions and organo-metal cleavages when using a common acceptor.

IV. Physical Chemistry

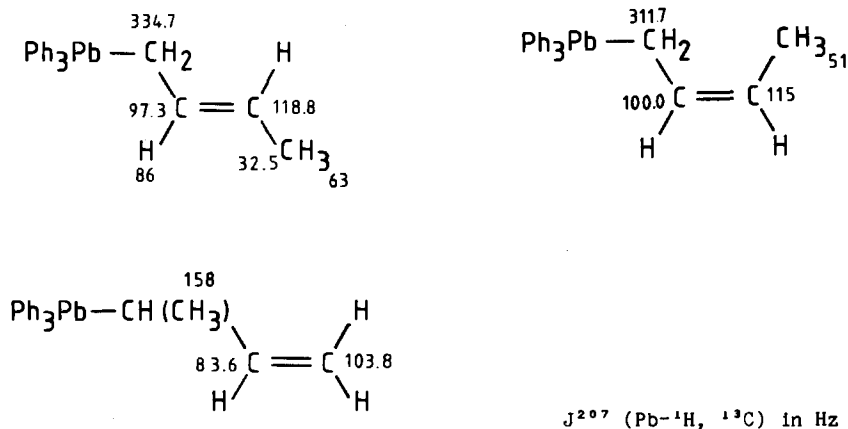
The structural chemistry of lead in particular of bivalent compounds has been described in relation to toxicity (51). The molecular structure of $\text{Pb}[\text{N}(\text{SiMe}_3)_2]_2$ has been determined in the crystal and the vapour: Pb-N bond length 2.24 Å (crystal), 2.20 Å (gas); N-Pb-N angle 103.6° (crystal), 91° (gas) (52). The compound consists of V-shaped monomers in both the vapour and the crystal. The crystal structure of $[\text{Pb}(\text{SC}_6\text{H}_3\text{Pr}_2^1 - 2,6)_2]_3$ has been studied (53). The terminal Pb atoms may be regarded as having tetrahedral geometry with one lone pair, while the central Pb has distorted trigonal bipyramidal geometry with an equatorial lone pair.

All three forms of selenocyanate bonding viz. ionic, N-bonded and Se-bonded, occur in adducts of Ph_3PbSeCN and $\text{Ph}_2\text{Pb}(\text{SeCN})_2$ with a variety of O- and N-donor ligands (54,55).



IR spectroscopy showed that dilution of a strong donor solvent, as DMF by an inert solvent e.g. CH_2Cl_2 , subsequently leads to decreased ionization and adduct dissociation. IR and Raman spectra of organometallic derivatives of phenols and thiophenols have been examined (56,57). Sulphur-metal stretch vibrations have been discussed in relation to the coordination of the metal in compounds such as Ph_3PbSPh ν (IR, solid state) 318 cm^{-1} . IR data of $\text{P}_7(\text{PbMe}_3)_3$ have been reported: ν (PbMe_3) 472 (vs), 455 (vs) and $\nu(\text{P-Pb})$ 310 (s) and 115 (vs) cm^{-1} . The crystal structural data have been compared with group IV analogues (58).

^1H - and ^{13}C -NMR data of allyl group IV compounds have been presented (59). Electronic mesomeric donating effects by $\sigma - \pi$ hyperconjugation could explain the trends in the data.



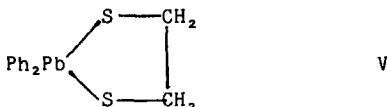
^{207}Pb -NMR data have been reported for Pb(II) polydentate phosphines at 295 K in nitromethane e.g. $\text{Pb}[\{\text{Ph}_2\text{P}(\text{CH}_2)_2\}_3\text{P}]_2^{2+}$ $\delta^{207}\text{Pb} - 269$ ppm relative to external Me_4Pb , $^1J^{207}\text{Pb} - ^3\text{PPh}_2$ 487 Hz, $^1J^{207}\text{Pb} - ^3\text{IP}$ 2148 Hz (60). Regression analyses of representative chemical shift data show a very good $^{119}\text{Sn} - ^{207}\text{Pb}$ correlation (61). The observed shift ratios correspond closely to the $\langle r^{-3} \rangle$ np ratio for the element pair. ^{207}Pb chemical shift data have been given for a range of bivalent lead compounds (62). There is a strong correlation between the ^{207}Pb chemical shift and the electronic structure of the species present in solution. Efficient nuclear shielding is achieved in species in which the lone pair of electrons is confined to the lead 6s orbital. The shielding is weaker in species with more covalently bound ligands, where the lone pair is stereochemically active in a hybrid sp^3d^4 orbital. Structures of lead dithiophosphoridates have been determined (63). Solution ^{207}Pb -NMR data have been presented. The sensitivity of coupling constants in i.a. organoleads towards changes in the metal-carbon bond has been discussed on the basis of changes in s-character and mutual polarizability (64). Spin-spin coupling constants have been calculated for R_4Pb (65). Synthesis and properties of cyclic organolead compounds have been described (66). ^1H -, ^{13}C - and ^{207}Pb -NMR data of these compounds and of a variety of acyclic organoleads have been compared.

Phase equilibrium in the system PbMe_4 -toluene resp. PbMe_4 -benzene has been studied (67). Crystallization can be put to use in the preparation of

PbMe₄ for the preparation of high-purity Pb and Pb oxide. The synthesis of Et₄Pb from Pb-Na alloy and EtCl was the subject of a microgravimetric study (13).

The usually transient cation-radicals of alk₄Pb were trapped in a CFC1₃ matrix after γ -irradiation at 90 K (68). The ESR spectrum of Me₄Pb revealed the presence of one unique Me ligand with ¹H hyperfine splitting of 14.7 G. The structure was envisaged as approximating a trigonal-pyramidal configuration at the Pb center, in which the singly occupied MO is largely localized in one methyl-metal bond.

He(I) and He(II) photoelectron spectra of (CH₃)₃PbCo(CO)₄ have been measured (69). Vertical ionization potentials were 8.11 (σ_{M-CO}), 8.34 (2e), 8.99 (1e), 9.43, 9.98 (σ_{M-C}), 13.8 (5 σ_{CO} , 1 π_{CO} , σ_{CH}) and 17.8 (4 σ_{CO}) eV. Relativistic effects have to be taken into account leading to contraction and stabilization of s orbitals on the Pb atom. The electronic structure in i.a. (V) has been investigated by UV photoelectron spectroscopy (70): 8.94 sh, 9.44, 10.16 sh, 11.6 eV and a band accounting for the ionization of the 5d orbitals of Pb: 28.06 eV.



Transfer parameters of keV electrons have been approximated for i.a. PbH₄ and PbMe₂ (71).

V. Applications

Polypropylene glycol has been prepared in the presence of catalysts obtained by treating Et₂Zn with Ph₃PbOH (72). Polypropylene glycol has also been synthesized in the presence of Ph₃PbSCH₂CH₂OH. This polymer has a higher molecular weight and crystallinity than the former, which is rubbery. Et₄Pb with SiCl₄ is an effective catalyst for homo- or copolymerization of Me methacrylate and styrene in air (73). Organic lead compounds have been used as catalysts in the preparation of dialkylcarbonates from monoalcohols and urea, or monoalcohols and urethane (74). The effect of poisoning of the Pt/Al₂O₃ catalyst by TEL has been investigated in the oxidation reaction of CO in a tubular reactor (75).

Metal containing plastic scintillators with effective absorption of soft X-rays and γ -rays and with minimal quenching of the luminescence of the plastic scintillators have been studied (76). The extent of quenching can be reduced by introducing e.g. diphenyl(α -naphthyl)lead in polystyrene. Ph_2PbOAc has been used in antifouling paints (77). A process for the purification of Et_4Pb has been published (78): Et_4Pb containing 0.05 - 1 vol.% water as an emulsion was cooled to -50 to -10° and filtered.

The amount of Et_4Pb to be added to a gasoline can be calculated from its hydrocarbon composition determined by GLC (79). The addition of organic sulphur compounds promotes the combustion of fuels, in particular in the presence of Pb compounds (80). Rapid carbonization has been observed with gasolines containing xylenes and PbEt_4 or PbMe_4 (81,82). Coke deposition can be minimized by Me_4Pb and dibromopropane. Experimental particulate traps in engines have been discussed (83). The addition of Et_4Pb to the diesel fuel reduces the ignition temperature of particulates on a trap by about 300°F .

VI. Analysis

A number of determinations of organolead compounds by Atomic Absorption Spectrometry has been published (84-94). Trialkyllead derivatives have been analyzed in water by fast vacuum distillation, saturation of the solution with NaCl , extraction in CHCl_3 , treatment with H_2SO_4 , and graphite-furnace atomic absorption. The detection limit was $0.02 \mu\text{g}$ with 1 L samples (88). Tetraalkylleads have been determined by AAS after extraction with CCl_4 , followed by oxidation with HNO_3 . The maximal standard deviations were 1.6 and 1.5% for Et_4Pb and Me_4Pb respectively (89). Et_4Pb or Me_4Pb emulsions in water have also been mineralized with iodine preceding AAS (90). Alkyllead vapours in outdoor air have been analyzed by adsorption enrichment of the alkyllead at -83° followed by AAS. The detection limit was 1 ng/m^3 and the reproducibility 6-9% (91). The detection limit for Pb is about $1 \mu\text{g/L}$ in the direct analysis of urine by AAS. Matrix effects can be minimized by adding $25 \mu\text{L}$ of 20% H_3PO_4 to $250 \mu\text{L}$ undiluted urine and by impregnating the graphite tube with 10% NH_4 -molybdate solution (92). A modified ASTM procedure for the determination of lead alkyls in fuels by AAS has been described. The method simplifies the assay and reduces the possibility of contamination, whilst retaining accuracy (93). A method for simultaneous analysis of the ionic species $\text{Me}_2\text{Pb}^{2+}$, and Me_3Pb^+ , $\text{Et}_2\text{Pb}^{2+}$, Et_3Pb^+ and also Me_4Pb , Me_3EtPb , $\text{Me}_2\text{Et}_2\text{Pb}$, MeEt_3Pb , Et_4Pb and inorganic Pb^{2+} has been presented (94). Chelation extraction with dithiocarbamate and benzene, followed by n-butylation to the tetraalkyllead form,

separation by gas chromatography and determination by AAS resulted in a detection limit of 0.1 ng.

Gas Chromatography (GC) has also been applied to the analysis of organolead compounds (95 - 100). GC with microwave plasma emission detection has been used for the determination of n-butylated trialkyllead compounds. After benzene extraction from a NaCl saturated solution trialkyllead compounds have been reacted with a n-Bu Grignard (95). Me_4Pb and Et_4Pb have been analyzed in solution by GC, BuOH being the internal standard (96,97). Volatile impurities in Et_4Pb have been detected by GC. The detection limit was 1×10^{-4} - $5 \times 10^{-5}\%$, with a column of 20% colloidal polyethylene glycol 400 - aerosil on chromaton N-AW-DMCS (98). GC with flame ionization detection was used for the assay of Me_3PbCl and Et_3PbCl . A carrier gas and a glass column packed with 10% OV-101 on Gaschrom were used. The detection limits were 25 and 5 ng respectively (99). After chelation with Na-diethyldithiocarbamate organoleads have been extracted into benzene. After butylation by Grignard reagent the tetraalkyllead formed has been analyzed by GC/MS. With 1L of water a detection limit of 0.1 $\mu\text{g/L}$ Pb can be achieved (100).

Reversed-phase high-performance liquid chromatography coupled to a sensitive chemical detection has been developed for the separation and detection of inorganic lead and organolead compounds. The compounds separated undergo complexation with 4-(2-pyridylazo)resorcinol. The red-coloured complex has been detected spectrophotometrically at 546 nm. Linear calibration curves have been obtained in the range 0.3 - 1.5 nmole (101). γ -Hexachlorocyclohexane is a new reagent for PbEt_4 in the determination of Pb in leaded gasoline (102). The use of X-ray spectrometry for the measurement of low concentrations of environmental contaminants such as organoleads has been reported (103).

VII. Wastes, Pollution and Industrial Hygiene

An industrial hygiene survey has been conducted to estimate workers' exposure to Et_4Pb (104).

After catalytic combustion in a diesel engine the applied Pb catalyst is trapped on the soot (105).

The removal of Et_4Pb is promoted by filtering solid impurities prior to filtering the wastewater through a semipermeable cellulose acetate membrane (106). Pb emissions from gasolines have been reduced by up to 90% by an exhaust filter filled with high-surface area Al_2O_3 , impregnated with a phosphate compound (107).

The regeneration of poisoned catalysts for exhaust gas from combustion of leaded gasoline has been reported (108).

Alcohol-gasoline mixtures reduce CO and SO_x emissions and can be used to eliminate the use of Et₄Pb (109).

The process of bioconversion of Pb(II) to Me₄Pb appears to be slow, inconsistent and variable and is strongly in doubt. It has been concluded that there is no solid evidence for the biomethylation of Pb (110, 111).

A relationship between atmospheric Pb concentrations and lead in gasoline has been found in Rio de Janeiro, Brazil (112). Concentrations of airborne Pb have been determined at one urban roadside site and two rural sites in Northwest England. Pb originates mainly from local vehicular sources at the urban site and from regional transport at the rural sites (113). Concentrations of Pb were much higher in various sections of highway than in city streets and were dependent on traffic intensity and meteorologic conditions (114).

The discussion on the reduction or omission of lead in petrol is being continued (115-120). The efforts for an EEC directive banning lead in petrol have been discussed (116). The British Government has accepted a recommendation by the Royal Commission on Environmental Pollution that lead should be eliminated from petrol. The costs of such an operation have been estimated (117).

The effect on environmental Pb, e.g. Pb in gasoline or paint, on blood Pb level in children has been studied. Pb-based paint was a significant factor (121). Without judging the evidence on the effects of low lead concentration on childrens' intelligence and behaviour, the removal of lead from petrol has been recommended (122). The dispersion and deposition of lead from motor exhausts and its effect on vegetables grown along a highway have been studied (123). The tissue lead concentration in pigeons increased according to locality: rural < subrural < urban. The trialkyllead derivatives in liver and kidney tissue of urban pigeons probably originate from the exhaust of vehicles using leaded gasoline (124). The Turin isotopic lead experiment based on added lead with a ²⁰⁶Pb/²⁰⁷Pb ratio of 1.04 instead of the usual 1.18 has been discussed and criticized (125).

VIII. Toxicology

The alga *Poterioochromonas malhamensis* appeared to be a sensitive test system for lead toxicity and lead antidotes. EGTA and CaNa₂EDTA completely suppressed the growth inhibition caused by inorganic lead. The toxic effects

of Et₃PbCl could not be reduced by any antidote (126). Etiology and diagnostic value of laboratory tests in acute Et₃Pb poisoning have been discussed (127).

A considerable increase in bird mortality was observed in 1979 in the Mersey Estuary UK. Birds containing high amounts of alkyl Pb died after consuming food contaminated with industrial effluents. Apparently healthy birds contained lower amounts of alkyl Pb, but still enough to suffer some ill effects (128). Starlings fed high doses (2 mg/day) of Et₃PbCl or Me₃PbCl died on or before the 6th day of the experiments. Even low doses (200 µg/day) administered to birds altered the food consumption. High-dose trimethyllead birds exhibited the most severe behavioural symptoms i.a. uncoordination, tremors and flight inability. All birds had an enlarged gall bladder, activated bone marrow and discoloured small intestine. In both low-dose groups muscle weight was reduced (129). Renal effects of trialkyllead derivatives have been observed in urban pigeons at the ultrastructural level (124).

Et₃PbCl intraperitoneally administered to pregnant guinea pigs decreased the placental amino acid transport by either inhibition of placental Na/K ATP-ase or reduction in maternal placental blood flow (130). In rabbits the total amount of Pb excreted into bile during 24 h after injection of 12 mg/kg of Et₃Pb was about 8% of the injected Pb. About 97% of the lead in the bile consisted of diethyllead salts. The total amount of Pb in the cecal contents 24 h after the injection of Et₃Pb was about 12% of the injected Pb; about 90% was inorganic Pb (131).

The effect of several ionic organometallic compounds i.a. organoleads on monoamine uptake and release in brain synaptosomes and blood platelets has been studied (132). The neurotoxicity of Bu₃PbOAc has been investigated in cultures of neuroblastoma and glioma (133). Me₃PbCl blocked (100-200 µM) and completely inhibited (≥ 300 µM) microtubule assembly in vitro in tubulin of porcine brain (134). Nerve cells in the cerebrum, cerebellum and spinal cord treated with Et₃Pb displayed extensive degeneration. The most striking feature was selective necrosis of the nerve cells in the hippocampus treated with > 20 mg/kg Et₃Pb: increased number of swollen mitochondria, lysosomes and multivesicular bodies, enlargement of Golgi complexes and rough endoplasmatic reticula (135). The neurotoxicity of Et₃PbCl has been studied in rats (136-138). Animals treated with Et₃PbCl showed after one week decreased Met-enkephalin level in hypothalamus, septum and frontal cortex, while the substance P level was decreased in hippocampus and frontal cortex. Concentrations of dopamine (DA), dihydroxyphenylacetic acid (DOPAC), serotonin (5-HT) and

5-hydroxyindoleacetic acid. (5-HIAA) were not affected. One day after administering the concentrations of 5-HT and 5-HIAA had increased. Et₃PbCl induced analgesia which could be attenuated by chlordiazepoxide or naloxone. This analgesia may be due to altered emotionality or reactivity towards noxious stimuli, which may be associated with alteration in the delta opiate mechanism in the limbic system (136). The neurotoxic effects of Et₃PbCl have been characterized in adult male rats. The single dose LD₅₀ (subcutaneous) was 11 mg/kg; after repeated exposure during 5 days this value rose to 14 mg/kg. Hyperexcitability and hyperactivity occurred 1-2 weeks after dosing, hypoexcitability and hypoactivity 3-4 weeks post dosing. Hot plate and tail flick latences were increased during the first 2 weeks. Changes in reactivity or emotionality were thought to be possibly similar to those in animals with lesions in limbic forebrain areas (138). With rat pups subcutaneous injection of 3 or 6 mg/kg of Et₃PbCl led to early sensory deficits, persistent hypoactivity at the highest dose level in males and hyperactivity in low dose females. The single postnatal day 5 injections of Et₃PbCl thus produced transitory effects possibly reflecting direct Et₃PbCl activity as well as apparent long term effects suggesting potential alterations in behavioural functions (137).

Average blood lead level of the US population dropped by approximately 37% from 1976 through 1980. A strong correlation between the lead level in blood and in gasoline was found. This correlation does not prove cause and effect, but the most likely explanation for the fall in blood lead level is the reduction of the lead content of gasoline during this period (139). The claim of 70% decrease in blood lead level since the introduction of lead free petrol in Tokyo has been questioned however (140). Therefore the outcome of measurement of bone, blood and hair lead concentrations in aborigine petrol sniffers in Australia is of importance. Values for bone lead of 20 ppm or over have been found in 23 of 60 sniffers. No bone lead has been detected in non-sniffer controls. The mean blood lead level was significantly greater in sniffers (2.55 $\mu\text{mol/L}$) than in non-sniffers (0.6 $\mu\text{mol/L}$). Mean hair lead also differed significantly between sniffers (23.0 ppm) and non-sniffers (12.4 ppm) (141). Workers exposed to organic Pb compounds in gasoline had higher average urine level of Pb (85 $\mu\text{g/L}$) and lower blood lead level than workers exposed to inorganic Pb compounds (urine level 62 $\mu\text{g/L}$) (143). There is a continuing discussion on these investigations of the relationship between blood lead levels, neuropsychological data and the use of lead in petrol (142). A positive correlation between Pb levels in the umbilical cord blood and the use of leaded gasoline has been found in Boston (144). Nevertheless the British

Department of the Environment has announced that investigations have failed to detect a statistically significant effect of lead on behaviour and intelligence of children in British cities; social factors have been stressed (145).

During the last few years the toxicology of organolead compounds has been studied extensively. An assessment of the current knowledge has been presented recently by P. Grandjean and E.C. Grandjean (146). A range of topics has been discussed: properties of organoleads (K.A. Jensen), gasoline additives (A. Laveskog), biomethylation (K. Beijer and A. Jernelöv), aquatic environmental organolead (Y.K. Chau and P.T.S. Wong), airborne organic lead (S. Berg, A. Jonsson and T. Nielsen), toxicity in plants (G. Röderer), metabolism and kinetics (A.A. Jensen), genotoxicity (E. Niebuhr and H.C. Wulf), effect on membranes (P.J. Bjerrum), mitochondria and other enzyme systems (W.N. Aldridge), membranes in the CNS (J. Clausen), reproduction and hormone metabolism (A. Odenbro), experimental pathology (A.A. Seawright, A.W. Brown, Y.C. Ng and J. Hrdlicka), neurotoxicity (J. Cremer), gasoline sniffing syndrome (R.A. Keenlyside), exposure and intoxication (P. Grandjean), preventive measures (J. Gething and G.R. Oxley) and governmental regulations (A.A. Jensen and P. Grandjean). This illustrates the shift in organolead research from pure and applied chemistry to environmental aspects and toxicology.

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