

## Silicon(IV) Derivatives of N-[*o*-Hydroxy Substituted (or H) Benzyl] Glycines

ROBINA AMAN, REETA SHARMA,  
HEMA PANDEY and MADHUP CHANDRA\*

*Department of Chemistry  
DSB Campus, Kumaun University  
Nainital-263 002, India*

Several silicon(IV) derivatives of N-[*o*-hydroxy substituted (or H) benzyl] glycines have been prepared by the interaction of silicon tetraisopropoxide with the latter in 1:1, 1:2, 1:3 and 1:4 molar ratios in benzene medium. The compounds thus prepared were generally obtained as coloured solids and amongst them those containing isopropoxy group(s) were found to be hygroscopic. All these compounds were characterized by azeotrope and elemental analyses, as well as by spectral measurements.

### INTRODUCTION

In continuation to the preparation of organometallic/metallo-organic derivatives of N-[*o*-hydroxy substituted (or H) benzyl] glycines (I)<sup>1-3</sup>, the work described here deals with the preparation of silicon(IV) derivatives of I, viz., (i) N-(2-hydroxybenzyl) glycine (H<sub>3</sub>hbg), (ii) N-(2-hydroxy-3-methyl benzyl) glycine (H<sub>3</sub>hmbg-3), (iii) N-(2-hydroxy-6-methyl benzyl) glycine (H<sub>3</sub>hmbg-6) and (iv) N-(2-hydroxy-5-methyl benzyl)glycine (H<sub>3</sub>hmbg-5) by alcoholysis reaction<sup>1-3</sup>. Involving the interaction of silicon tetraisopropoxide with I in 1:1, 1:2, 1:3 and 1:4 molar ratios in benzene medium. The various compounds thus prepared were obtained as coloured solids and out of them those containing isopropoxy group(s) were found to be hygroscopic. All these compounds were characterized by azeotrope and elemental analyses, as well as by IR and PMR measurements.

### EXPERIMENTAL

The details of the glass apparatus used have been given before<sup>1,2</sup>. Stringent precautions were taken to exclude moisture throughout the experiments, as earlier<sup>1,2</sup>.

Benzene (BDH, AR), isopropanol (BDH, Glaxo AnalaR) and solvent ether (E. Merck) were dried by standard procedures<sup>4</sup>. Silicon tetraisopropoxide was prepared by sodium method<sup>5</sup>, while N-[*o*-hydroxy substituted (or H) benzyl] glycines were prepared employing already reported methods<sup>1</sup>. Silicon(IV) was estimated by a known method<sup>6</sup>, whenever required.

The melting points were recorded on a CAT no IA 8103 digital melting point apparatus. The IR spectra were recorded on a Perkin-Elmer Model 983 spectrometer in KBr pellets, while the PMR spectra were taken in DMSO-d<sub>6</sub> solution and recorded on a Varian EM-390, 90 MHz spectrometer.

### Reaction Between $\text{Si}(\text{OPr}^i)_4$ and $\text{H}_3\text{hmbg-3}$ , 1 : 1 Molar Ratio

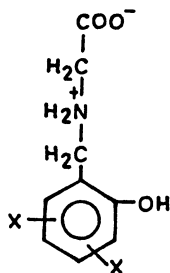
A mixture of  $\text{Si}(\text{OPr}^i)_4$  (2.7268 g, 10.3114 mmole) and  $\text{H}_3\text{hmbg-3}$  (2.0128 g, 10.3103 mmole) suspended in dry benzene (60 mL) taken in a R.B. flask was refluxed on a wax bath at 95–100°C, using a fractionating column. After *ca.* 24 h of reflux, isopropanol liberated was fractionated out azeotropically and estimated by an oxidimetric method<sup>7, 8</sup>. On completion of the reaction, the excess of solvent from the reaction mixture was removed *in vacuo*, when the product,  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-3})$  isolated as a light cream solid. It was then filtered under suction, washed with dry benzene (3–4 times) and then finally washed with dry ether (2–3 times).

It may be mentioned here that since  $\text{Si}(\text{OPr}^i)_4$  is soluble in benzene, while  $\text{H}_3\text{hmbg-3}$  is insoluble, the latter was taken in slightly less than the required stoichiometric amount in order to avoid contamination of impurities likely to occur by the unreacted  $\text{H}_3\text{hmbg-3}$  taken.

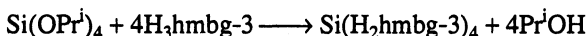
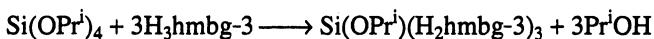
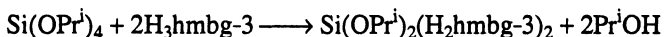
Similar procedures of preparation and purification were adopted in case of other derivatives. The relevant analytical details, characteristic IR frequencies and the PMR data are recorded in Tables 1–3 respectively.

### RESULTS AND DISCUSSION

It may be recalled here that N-[*o*-hydroxy substituted (or H) benzyl]glycines exist in zwitterionic form (Structure I). The various reactions between silicon tetraisopropoxide and  $\text{H}_3\text{hmbg-3}$  may be illustrated as:



(Where X = -H or -CH<sub>3</sub>)  
( I )



Identical reactions occurred in case of  $\text{H}_3\text{hbg}$ ,  $\text{H}_3\text{hmbg-6}$  and  $\text{H}_3\text{hmbg-5}$ .

TABLE-1  
ANALYTICAL DETAILS OF N-[*o*-HYDROXY SUBSTITUTED (OR H) BENZYL]  
GLYCINES AND THEIR SILICON(IV) DERIVATIVES

Compound (molar ratio)/(colour)	Reflux time (h)	m.p. (°C)	Elemental analysis % found (calcd.)			
			C	H	N	Si
H <sub>3</sub> hbg (off white)	—	50	59.60 (59.66)	6.09 (6.12)	7.68 (7.73)	—
H <sub>3</sub> hmbg-3 (off white)	—	190	61.46 (61.52)	6.68 (6.71)	7.08 (7.18)	—
H <sub>3</sub> hmbg-6 (off white)	—	88	61.48 (61.52)	6.67 (6.71)	7.10 (7.18)	—
H <sub>3</sub> hmbg-5 (off white)	—	148	61.47 (61.52)	6.67 (6.71)	7.12 (7.18)	—
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hbg) (1 : 1) (light cream)	12	>300	55.93 (56.10)	8.01 (8.05)	3.63 (3.64)	7.24 (7.27)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hbg) <sub>2</sub> (1 : 2) (light cream)	24	>300	56.84 (56.92)	6.69 (6.72)	5.51 (5.53)	5.50 (5.53)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hbg) <sub>3</sub> (1 : 3) (light cream)	12	>300	57.30 (57.42)	5.88 (5.90)	6.69 (6.70)	4.46 (4.47)
Si(H <sub>2</sub> hbg) <sub>4</sub> (1 : 4) (light cream)	18	>300	57.65 (57.75)	5.35 (5.35)	7.48 (7.49)	3.73 (3.74)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-3) (1 : 1) (light cream)	24	190	57.03 (57.14)	8.26 (8.27)	3.49 (3.51)	7.00 (7.02)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-3) <sub>2</sub> (1 : 2) (brownish cream)	26	205	58.39 (58.43)	7.10 (7.12)	5.22 (5.24)	5.24 (5.24)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-3) <sub>3</sub> (1 : 3) (brownish cream)	18	170	59.08 (59.19)	6.41 (6.43)	6.26 (6.28)	4.08 (4.19)
Si(H <sub>2</sub> hmbg-3) <sub>4</sub> (1 : 4) (brownish cream)	26	180	59.42 (59.70)	5.92 (5.97)	6.98 (6.97)	3.46 (3.48)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-6) (1 : 1) (light yellow)	12	160	57.08 (57.14)	8.25 (8.27)	3.50 (3.51)	7.00 (7.02)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-6) <sub>2</sub> (1 : 2) (light cream)	24	180	58.19 (58.43)	7.08 (7.12)	5.24 (5.24)	5.22 (5.24)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-6) <sub>3</sub> (1 : 3) (light cream)	24	175	59.04 (59.19)	6.41 (6.43)	6.26 (6.28)	4.18 (4.19)
Si(H <sub>2</sub> hmbg-6) <sub>4</sub> (1 : 4) (light cream)	24	175	59.64 (59.70)	5.93 (5.97)	6.96 (6.97)	3.47 (3.48)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-5) (1 : 1) (light cream)	12	165	57.09 (57.14)	8.25 (8.27)	3.48 (3.51)	6.99 (7.02)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-5) <sub>2</sub> (1 : 2) (light cream)	12	145	58.23 (58.43)	7.10 (7.12)	5.23 (5.24)	5.23 (5.24)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-5) <sub>3</sub> (1 : 3) (light cream)	18	140	59.01 (59.19)	6.40 (6.43)	6.25 (6.28)	4.18 (4.19)
Si(H <sub>2</sub> hmbg-5) <sub>4</sub> (1 : 4) (light cream)	20	160	59.62 (59.70)	5.95 (5.97)	6.96 (6.97)	3.47 (3.48)

Abbreviations: OPr<sup>i</sup> = OC<sub>3</sub>H<sub>7</sub>, H<sub>3</sub>hbg = OHC<sub>6</sub>H<sub>4</sub>CH<sub>2</sub>NH<sub>2</sub>CH<sub>2</sub>COO<sup>-</sup> and H<sub>3</sub>hmbg-3 (or -6 or -5) = OHC<sub>6</sub>H<sub>3</sub>(CH<sub>3</sub>)CH<sub>2</sub>NH<sub>2</sub>CH<sub>2</sub>COO<sup>-</sup>

TABLE-2  
 CHARACTERISTIC INFRARED FREQUENCIES ( $\text{cm}^{-1}$ ) OF N-[ $\beta$ -HYDROXY SUBSTITUTED (OR H) BENZYL] GLYCINES  
 AND THEIR SILICON(IV) DERIVATIVES

Compound	v(OH) and aromatic v(C—H)	v(N—H) and aromatic v(C—H)	v(C—H) of —CH <sub>2</sub> — and —CH <sub>3</sub> groups	$\text{>NH}_2$	$\nu_{\text{asym}}(\text{COO})$	$\nu_{\text{sym}}(\text{COO})$	$\Delta\nu(\text{COO})$	v(C—N)	v(Si—O)	v(Si—N)
H <sub>3</sub> hbg	3450–3000 (vb)	—	2940 (w) 2850 (w)	2600 (wb)	1630 (vsb)	1400 (s)	—	1230 (m)	—	—
H <sub>3</sub> hmbg-3	3600–3000 (vb)	—	2950 (vb) 2855 (wb)	2390 (wb)	1635 (vsb)	1405 (m)	—	1230 (s)	—	—
H <sub>3</sub> hmbg-6	3500–3000 (vb)	—	2910 (mb) 2860 (wb)	2375 (wb)	1640 (vsb)	1405 (m)	—	1225 (m)	—	—
H <sub>3</sub> hmbg-5	3500–3000 (vb)	—	2910 (mb) 2870 (w)	2380 (w)	1635 (vsb)	1400 (s)	—	1220 (m)	—	—
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hbg)	3450–3350* (wb)	3200–3000 (b)	2980 (vw)	—	1640 (vsb)	1395 (mb)	245	1260 (m)	760 (m)	690 (mb)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hbg) <sub>2</sub>	3450–3300* (b)	3200–3000 (mb)	2980 (w)	—	1645 (vsb)	1395 (mb)	250	1265 (m)	760 (s)	710 (s)
Si(OPr <sup>i</sup> ) <sub>1</sub> (H <sub>2</sub> hbg) <sub>3</sub>	3450–3300* (b)	3200–3000 (vb)	2980 (w)	—	1645 (vsb)	1395 (vsb)	250	1260 (wb)	760 (s)	705 (m)
Si(H <sub>2</sub> hbg) <sub>4</sub>	3450–3300* (mb)	3150–3000 (mb)	2980 (w)	—	1640 (vsb)	1395 (mb)	245	1260 (mb)	760 (s)	705 (m)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-3)	3450–3300* (b)	3200–3000 (b)	2980 (w)	—	1640 (vsb)	1390 (mb)	250	1260 (w)	765 (w)	690 (m)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-3) <sub>2</sub>	3450–3300* (b)	3150–3000 (wb)	2960 (m)	—	1640 (vsb)	1395 (mb)	245	1260 (s)	740 (s)	680 (m)

Compound	v(OH) and aromatic v(C—H) v(C—H)	v(N—H) and aromatic v(C—H)	v(C—H) of —CH <sub>2</sub> — and —CH <sub>3</sub> groups	>N <sup>+</sup> H <sub>2</sub>	v <sub>asym</sub> (COO)	v <sub>sym</sub> (COO)	Δv(COO)	v(C—N)	v(Si—O)	v(Si—N)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-3) <sub>3</sub>	3400–3250* (mb)	3150–3000 (b)	2980 (w) 2940 (w)	—	1645 (vsb)	1395 (vsb)	250	1260 (m)	740 (mb)	700 (m)
Si(H <sub>2</sub> hmbg-3) <sub>4</sub>	3400–3300* (b)	3150–3000 (mb)	2980 (w)	—	1645 (vsb)	1395 (mb)	250	1270 (m)	750 (vs)	700 (s)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-6)	3450–3300* (b)	3150–3000 (mb)	2985 (vs)	—	1635 (vsb)	1395 (mb)	240	1265 (s)	740 (m)	670 (m)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-6) <sub>2</sub>	3400–3300* (b)	3150–3000 (mb)	2980 (m)	—	1640 (vsb)	1395 (mb)	245	1270 (s)	750 (m)	710 (m)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-6) <sub>3</sub>	3450–3300* (b)	3150–3000 (wb)	2990 (m) 2960 (w)	—	1645 (vsb)	1395 (mb)	250	1270 (s)	740 (w)	710 (s)
Si(H <sub>2</sub> hmbg-6) <sub>4</sub>	3400–3300* (vb)	3180–3000 (mb)	2980 (m) 2960 (w)	—	1640 (vsb)	1395 (mb)	245	1265 (m)	740 (w)	705 (m)
Si(OPr <sup>i</sup> ) <sub>3</sub> (H <sub>2</sub> hmbg-5)	3400–3300* (b)	3190–3000 (vb)	2970 (w) 2940 (w)	—	1640 (vsb)	1390 (mb)	250	1270 (s)	750 (w)	710 (w)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-5) <sub>2</sub>	3400–3300* (b)	3250–3000 (b)	2980 (m)	—	1640 (vsb)	1390 (mb)	250	1270 (s)	750 (m)	690 (m)
Si(OPr <sup>i</sup> )(H <sub>2</sub> hmbg-5) <sub>3</sub>	3400–3300* (mb)	3180–3000 (wb)	2980 (w) 2940 (w)	—	1640 (vsb)	1390 (vsb)	250	1260 (m)	770 (m)	710 (s)
Si(H <sub>2</sub> hmbg-5) <sub>4</sub>	3450–3300* (b)	3190–3000 (mb)	2960 (w)	—	1630 (vsb)	1390 (vsb)	240	1270 (s)	770 (m)	700 (w)

\*v(OH) alone

Abbreviations: s = strong, vs = very strong, b = broad, vb = very broad, vsb = very strong broad, m = medium, mb = medium broad, w = weak, wb = weak broad.

TABLE-3  
 PROTON MAGNETIC RESONANCE SPECTRAL DATA ( $\delta$  VALUES) OF N-[*o*-HYDROXY SUBSTITUTED (OR H) BENZYL] GLYCINES  
 AND THEIR SILICON(IV) DERIVATIVES

Compound	Aromatic ring	Phenolic OH	$>NH_2$	$>NH$	—CH <sub>2</sub> —		Gem-dimethyl of isopropoxy group(s)
					Benzene ring	Glycine part	
H <sub>3</sub> hbg	6.50–7.30 (q)	4.50–5.90 (h)	3.60 (s)	—	3.25 (s)	2.45 (s)	—
H <sub>3</sub> hmbg-3	6.50–7.10 (t)	4.50–5.70 (h)	3.55 (s)	—	3.15 (s)	2.50 (s)	2.15 (s)
H <sub>3</sub> hmbg-6	6.40–7.10 (t)	4.40–5.60 (h)	3.65 (s)	—	3.20 (s)	2.45 (s)	2.15 (s)
H <sub>3</sub> hmbg-5	6.50–7.00 (t)	4.50–5.50 (h)	3.70 (s)	—	3.25 (s)	2.45 (s)	2.15 9s)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-3)	6.20–6.80 (m)	4.00–4.90 (h)	—	3.50 (s)	3.10 (s)	2.50 (s)	2.15 (s) 1.15 (d)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hbg) <sub>2</sub>	6.20–7.10 (m)	4.00–4.50 (m)	—	3.55 (s)	3.15 (s)	2.35 (s)	2.10 (s) 1.16 (d)
Si(OPr <sup>i</sup> ) <sub>2</sub> (H <sub>2</sub> hmbg-3) <sub>3</sub>	6.40–7.00 (m)	4.20–5.20 (h)	—	3.50 (s)	3.10 (s)	2.50 (s)	2.20 (s) 1.55 (d)
Si(H <sub>2</sub> hmbg-5) <sub>4</sub>	6.20–7.10 (m)	4.20–4.70 (h)	—	3.65 (s)	3.15 (s)	2.50 (s)	2.15 (s) —

Abbreviations: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, h = hump.

### Spectra Studies<sup>9-12</sup>

**Infrared spectra:** The derivative,  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-3})$  displays a broad band between  $3450\text{--}3300\text{ cm}^{-1}$  which may be assigned to the stretching of unbonded phenolic ( $\text{—OH}$ ) group. A broad band in the region  $3200\text{--}3000\text{ cm}^{-1}$  indicates possible overlapping of  $\nu(\text{N—H})$  and aromatic  $\nu(\text{C—H})$ . The appearance of  $\nu(\text{N—H})$  in the lower region suggests coordination of nitrogen to silicon. Further, a shift of  $30\text{ cm}^{-1}$  in  $\nu(\text{C—N})$  as compared to  $\text{H}_3\text{hmbg-3}$ , again shows the bonding of nitrogen to silicon. The absence of any characteristic band corresponding to the ( $\text{C=O}$ ) group in the region  $1750\text{--}1650\text{ cm}^{-1}$  rules out the possibility of a normal ester type of linkage between the carboxylate oxygen and silicon. A shift of  $15\text{ cm}^{-1}$  in  $\nu_s(\text{COO})$ , as compared to  $\text{H}_3\text{hmbg-3}$ , suggests the bonding of the carboxylate oxygen to silicon. The separation value,  $\Delta\nu(\text{COO})$  [ $\nu_{\text{asym}}(\text{COO}) - \nu_{\text{sym}}(\text{COO})$ ] of  $250\text{ cm}^{-1}$ , as observed here shows the absence of bridged or coordinated carboxylate group. The weak absorption at  $765\text{ cm}^{-1}$  occurs due to  $\nu(\text{Si—O})$ , while the medium band at  $690\text{ cm}^{-1}$  corresponds to  $\nu(\text{Si—N})$ .

It is thus evident that the silicon atom in  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-3})$  exhibits penta-coordination as a result of bonding with one of the oxygens from the carboxylate group and the nitrogen from the imino group, along with three isopropoxy groups (Structure II).

**Proton Magnetic Spectra:** The PMR spectrum of  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-3})$  shows a multiplet between  $\delta\ 6.20\text{--}6.80$  which corresponds to the aromatic ring protons. The hump between  $\delta\ (4.00\text{--}4.90)$  shows the presence of unbonded phenolic group proton. The singlet at  $\delta\ 3.55$  due to  $>\text{NH}_2$  protons, as observed in  $\text{H}_3\text{hmbg-3}$  is found to be absent here with the appearance of a new peak at  $\delta\ 3.50$  suggesting possible coordination of nitrogen to silicon. The singlets at  $\delta\ 3.10$  and  $\delta\ 2.50$  may be assigned to the protons associated with the  $\text{—CH}_2\text{—}$  group attached with benzene ring and the glycine part of  $\text{H}_3\text{hmbg-3}$ , respectively. The singlet at  $\delta\ 2.15$  and a doublet at  $\delta\ 1.15$  correspond to the protons associated with the  $\text{—CH}_3$  group attached with benzene ring and *gem*-dimethyl protons of the isopropoxy groups, respectively. Thus, the inferences drawn here are well in conformity to those derived from the IR measurements earlier.

The IR and PMR (wherever available) data in respect of the other derivatives were interpreted similarly and the main findings relating to their structures are as under:

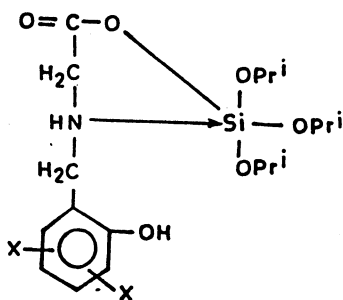
The derivatives,  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hbg})$ ,  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-6})$  and  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-5})$  contain a penta-coordinated silicon atom in each case displaying similar modes of bonding as those observed in  $\text{Si}(\text{OPr}^i)_3(\text{H}_2\text{hmbg-3})$  (Structure II).

The silicon atom in  $\text{Si}(\text{OPr}^i)_2(\text{H}_2\text{hbg})_2$ ,  $\text{Si}(\text{OPr}^i)_2(\text{H}_2\text{hmbg-3})_2$ ,  $\text{Si}(\text{OPr}^i)_2(\text{H}_2\text{hmbg-6})_2$  and  $\text{Si}(\text{OPr}^i)_2(\text{H}_2\text{hmbg-5})_2$  displays hexa-coordination in each case by way of bonding with one of the oxygens from each of the two carboxylate groups and the nitrogen from each of the two imino groups available from two moles of I, along with two isopropoxy groups (Structure III).

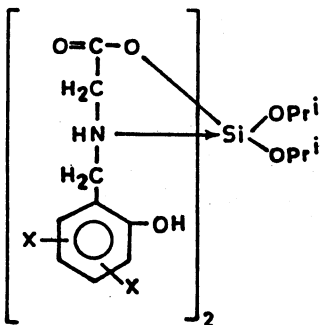
The derivatives  $\text{Si}(\text{OPr}^i)(\text{H}_2\text{hbg})_3$ ,  $\text{Si}(\text{OPr}^i)(\text{H}_2\text{hmbg-3})_3$ ,  $\text{Si}(\text{OPr}^i)(\text{H}_2\text{hmbg-6})_3$  and  $\text{Si}(\text{OPr}^i)(\text{H}_2\text{hmbg-5})_3$  possess a hepta-coordinated silicon atom in each case as a consequence of bonding with one of the oxygens from each of the three

carboxylate groups and the nitrogen from each of the three imino groups available from three moles of I, along with an isopropoxy group (Structure IV).

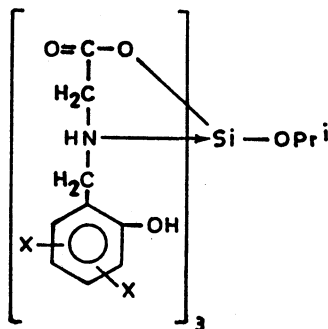
The silicon atom in  $\text{Si}(\text{H}_2\text{hbg})_4$ ,  $\text{Si}(\text{H}_2\text{hmbg-3})_4$ ,  $\text{Si}(\text{H}_2\text{hmbg-6})_4$  and  $\text{Si}(\text{H}_2\text{hmbg-5})_4$  displays octa-coordination in each case as a result of bonding with one of the oxygens from each of the four carboxylate groups and the nitrogen from each of the four imino groups available from four moles of I (Structure V).



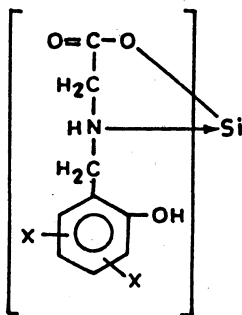
(Where X = -H or -CH<sub>3</sub>)  
(II)



(Where X = -H or -CH<sub>3</sub>)  
(III)



(Where X = -H or -CH<sub>3</sub>)  
(IV)



(Where X = -H or -CH<sub>3</sub>)  
(V)



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