MNDO STUDIES OF THE $SiCl_4$ COMPLEXES WITH SOME ORGANIC LIGANDS

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Abstract

MNDO calculations of the $SiCl_4$ complexes with pyridine, trimethylamine and tetramethyl urea have been carried out when the coordinate of the reaction of complex formation is varied. These calculations conform to the experimentally established structure of these complexes. The changes of atomic charges and p-orbital populations of atoms at decrease of the distance between the complex components have been analysed. The peculiarities and mechanism of formation of complexes of different structures have been examined.

Introduction

Tetrachlorides of Si, Ge and Sn (MCl_4) with organic ligands L form complexes both of octahedral and trigonal-bipyramidal structure. Their structure depends on the M atom and ligand character, on the component ratio and peculiarities of complex formation. The structure of such complexes is established reliably by ^{35}Cl NQR. By this way the change of the electron density distribution in donor and acceptor molecules at complex formation, the peculiarities of this distribution in complexes of different structure have been studied; some conclusions about a mechanism of complex formation have been made, etc. (see, e.g., [1,2]). For more detail discussion of these problems the quantum mechanical calculations of the $SiCl_4$ complexes with some organic ligands (pyridine, trimethylamine and tetramethyl urea) have been carried out by us. The selection of these ligands is caused by a different structure of the complexes, which are formed with their participation. The selection of the $SiCl_4$ complex with tetramethyl urea is also caused by the fact, that this complex possesses unusual structure [1-4]. The ligand in this trigonal-bipyramidal complex occupies an equatorial position of the bipyramid. At the same time the organic ligands in practically all trigonal-bipyramidal complexes of tetrachlorides of metals of group IVA, which are studied till now, occupy their axial position (see, e.g., [1-3]).

The calculations were carried out using the MNDO approximation, valence sp-basis and molecule geometry optimization. Every system $SiCl_4$ - nL (n=1 and 2) was calculated at the changed coordinate of the reaction of complex formation (from practically endless distance d between the complex components to d=1.5 Å). Herewith the calculations were carried out not only for the real structures of the complexes but also for some another

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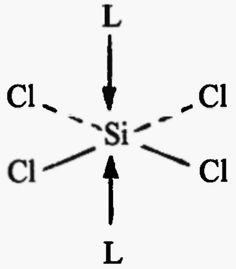
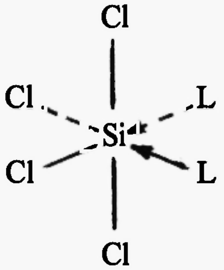
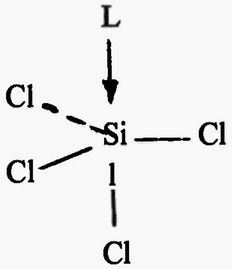
probable structures. For the $SiCl_4$ complex with pyridine the trans and cis octahedral structure (I and II accordingly) as well the two trigonal-bipyramidal ones were calculated:



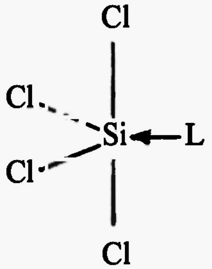
Structure I



Structure II



Structure III



Structure IV

in one of them the ligand occupies the axial position of the bipyramid (structure III) and in the other one it occupies the equatorial position (structure IV). For the $SiCl_4$ complexes with trimethylamine and tetramethyl urea only two later structures were calculated. Configuration of the complex acceptor fragment with all d values was set approximately the same as that must be in a complex. Then the structure of this fragment was optimized as of the all system $SiCl_4$ - nL as a whole. The results of the fulfilled calculations were compared to the mentioned above experimental data [1-4].

Results and Discussion

It was established by ^{35}Cl NQR that the $SiCl_4$ complex with pyridine is trans octahedral (structure I) [3,4]. The MNDO calculations conform to this structure. The minimum heat of this complex formation (at d=2.0 Å) is by 4.3 kcal/mole smaller than for the complex of the structure IV (at d=1.9 Å), by 8.9 kcal/mole smaller, than for the complex of the structure III (at d=2.0 Å) and by 15.4 kcal/mole smaller, than for the complex of the structure II (at d=2.0 Å too). The calculations show that the planes of the pyridine cycles in the trans octahedral complex $SiCl_4{\cdot}2NC_5H_5$ are perpendicular to each other and their projections on the plane, where are the Cl atoms, coincide with the angles ClSiCl bisectors. This provides the full equivalence of the electron distribution of the Cl atoms in this complex and conforms to its singlet ^{35}Cl NQR spectrum.

Under calculations the heat of formation of the trigonal-bipyramidal complex $SiCl_4{\cdot}N(CH_3)_3$ of the structure (III) is by 5.8 kcal/mole smaller (at d=2.25 Å) than of the structure (IV) (at d=2.0 Å). It conforms to the ^{35}Cl NQR spectrum, which gives evidence of the formation of the $SiCl_4{\cdot}N(CH_3)_3$ complex of the structure (III) [3].

In accordance with experimental data (see above) the MNDO calculations show that the energetically most advantageous structure of the $SiCl_4{\cdot}OC[N(CH_3)_2]_2$ complex is the trigonal-bipyramidal one with the ligand in unusual for such complexes equatorial position of the bipyramid. The minimum heat of formation of the complex of this structure (at d=1.7 Å) is by 1.5 kcal/mole smaller, than of the structure III (at d=1.8 Å).

Thus, the structure of the complexes under consideration, which follows from the MNDO calculations, completely corresponds to the experimental data. For studying the peculiarities of the electron density redistribution in complex components, when the complex is formed, we have analysed the changes of atomic charges at the decrease of the distance d between the complex components from 200 Å, when complex is absent, to the d value when the heat of complex formation is minimum.

When the components of the $SiCl_4 \cdot 2NC_5H_5$ system draw together owing to electrostatic interaction of the complex-forming centers, in the main, the positively charged Si atom polarizes the pyridine cycles so, that their electron density displaces on the N atoms, and at the sufficiently small distance d it partially gets across on the electron acceptor. Under the action of the partially negatively charged N atom the electron density from the Si atom displaces on the Cl atoms. This gives rise to the increase of the positive charge at the Si atom and negative charge at the Cl atoms, when the complex is formed (Table 1). As a result of this polarization the electron density, not only transferred from the ligand, but also the part of the Si atom electron density is localized on the Cl atoms. The complex-forming Si and N centers in this complex are only initiators of the charge transfer and conductors for this charge. The total charges at the atoms of the electron acceptor gets negative (-0.523e) and of the donor - positive (0.260e in each pyridine cycle) at this complex formation. Analogous decrease of the Sn atom electron density, when a complex is formed, follows from quantum mechanical calculations, Mössbauer and X-ray emission SnK_α-spectra of octahedral $SnCl_4$ complexes [5-8].

When the components of the system $SiCl_4$ - $N(CH_3)_3$ (structure III) draw together, the positive charge at the Si atom increases and the negative charge at the N atom decreases. Herewith the negative charge at the axial Cl atom insignificantly increases whereas at the equatorial Cl atoms it increases essentially (Table 1). It is caused by the considerably greater polarization of the equatorial Si-Cl bonds, than the axial one, under the action of the partially negatively charged N atom. In this complex 0.2e get across from the donor to the electron acceptor. The polarization of the Si-Cl bonds gives rise to the localization on the Cl atoms of the acceptor the essentially greater electron density, than that transferred from the electron donor.

The mechanism of formation of the trigonal-bipyramidal $SiCl_4 \cdot N(CH_3)_3$ complex is the same, as that for the trans-octahedral complex $SiCl_4 \cdot 2NC_5H_5$. However in the system $SiCl_4$-$N(CH_3)_3$ the N atom not only initiates the complex formation and serves as a conductor of the electron density from the donor to the acceptor, but itself gives a part of its electron density.

The character of electron density redistribution at the formation of the $SiCl_4 \cdot OC[N(CH_3)_2]_2$ complex of the structure IV is caused, first of all, as it is at another complexes formation, by the electrostatic interaction of the electron donor and acceptor atoms, especially of the O and Si atoms. This interaction causes the approach of these atoms. Herewith the polarization of the Si-Cl bonds, especially axial ones, is intensified under the action of the negatively charged O atom and the polarization of the ligand is intensified under the action of the positively charged Si atom. The former causes the increase of the negative charge at the Cl atoms, especially axial ones, and positive charge at the Si atom, and the last causes the increase of the negative charge at the O atom and the positive charge at the carbonyl C atom and the H atoms of the methyl groups (Table 1). At the sufficiently small distances d the transfer of the electron density from the electron donor to the acceptor is realized too. Herewith the Si and O atoms serve only as the conductors of electron density. As a result of this transfer the total charge at the electron acceptor in this complex gets negative (-0.315e), and at the donor - positive. The polarization of the Si-Cl bonds under the O atom charge action carries out to the significantly greater total change of the electron density at the Cl atoms (0.396e), when the complex is formed, than the value of the transferred charge (see above).

In going from the $SiCl_4$ molecule of tetrahedral structure to the plane one, which it possesses in the trans octahedral structure I (at d=200 Å, when the complex has not formed yet), the partial positive charge at the Si atom increases by 0.114e, and in going to such structure which this molecule possesses in cis isomer (structure II) (at d=200 Å too) this charge decreases by 0.104e. In going from the tetrahedral $SiCl_4$ molecule to such its structure, which it has in the trigonal-bipyramidal structure III (at d=200 Å) the positive charge at the Si atom decreases too (by 0.116e), and in going to such structure which it possesses in the structure IV this charge increases by 0.095e.

One may suppose that the complex formation is initiated, first of all, by the electrostatic interaction of the oppositively charged complex-forming centers (see, e.g., [2]). In this case the increase of the charges at these centers when the geometry of the components of the complex at its formation is changed, must promote the complex-formation. From this point of view taking into account the change of the Si atom charge, the trans octahedral structure (I) of the $SiCl_4 \cdot 2L$ complex is more preferable, than cis octahedral one (II), and the trigonal-bipyramidal structure IV of the $SiCl_4 \cdot L$ complex is more preferable, than structure III. The trans octahedral structure of the $SiCl_4 \cdot 2NC_5H_5$ complex and the trigonal-bipyramidal structure (IV) of the $SiCl_4 \cdot OC[N(CH_3)_2]_2$ complex conform to this supposition. However, the structure III of the $SiCl_4 \cdot N(CH_3)_3$ complex does not conform to it. This is caused by the fact, that the preferability of the formation of a complex of either structure depends not only on the charges at the complex-forming centers, but on many other reasons too. One of them is the decrease not only positive charge at the Si atom, but also negative charges at the equatorial Cl atoms in going from tetrahedral $SiCl_4$ molecule to such its structure which it must possess in the system of the structure III. This facilitates the approach of the donor and acceptor molecules. In going from the tetrahedral $SiCl_4$ molecule to such its structure, which it possesses in a complex of the structure III, not only positive charge at the Si atom increases, but the negative charges at the all Cl atoms too. The former must hamper the approach of the complex components. Evidently, depending on the ligand character, the influence on it either positively charged Si atom or negatively charged Cl atoms may turn out predominant.

For more deep analysis of the electron density redistribution in the complex components at the complex formation we have examined the atomic p-orbital populations and their changes when the distance d between the components decreases from 200 Å to the d value which provides the minimum heat of the complex formation (Table 2). The X axes of atoms, which are in equatorial plane of a polyhedron, are perpendicular to this plane. The Z axes of all Cl atoms coincide with the corresponding Si-Cl bond directions, as this is usually accepted in NQR spectroscopy.

At d=200 Å, when the complex $SiCl_4 \cdot 2NC_5H_5$ of the structure I has not formed yet, the p_x-orbital population of the Si atom (0.177e) is equal to the sum of the electron deficit on the p_x-orbitals of the equatorial Cl atoms. Evidently, the small lateral overlap of the Cl and Si atom p_x-orbitals possessing practically equal sizes takes place in the plane $SiCl_4$ fragment. At the decrease of the d distance in the $SiCl_4$ - $2NC_5H_5$ system the p_x-orbital population of the Si atom decreases just as the electron deficit on the p_x-orbitals of the Cl atoms. Herewith the sum of their deficit is equal to the Si atom p_x-orbital population at the decrease of the d value to 3.5 Å. At the further decrease of d the Si atom p_x-orbital population exceeds the sum of the electron deficit on the p_x-orbitals of the Cl atoms. At this d the transfer of electron density from the electron donor on the Si atom p_x-orbital is started. Herewith the multicenter bond, including four Cl atoms, Si atom and two N atoms is formed. Analogous multicenter bond we suggested in molecules of pentacoordinated Si and P atoms taking into account the ^{35}Cl NQR data and X-ray emission SiKβ- and PKβ-spectra of such compounds [9-11]. Judqing by the small deficit of p_x-electrons of the equatorial Cl atoms in the $SiCl_4 \cdot 2NC_5H_5$ complex (only 0.012e), the participation of the Cl atoms in this multicenter bond is insignificant. At the same time, for the equatorial Cl atoms in complexes of tetrachlorides of metals of group IVA, possessing a trigonal-bipyramidal or octahedral structure, the great value of asymmetry parameter (η) of the electric field gradient (EFG) at the ^{35}Cl nuclei is typical [1,2,4,11-13]. This value may give evidence of noticeable deviation of the electron density distribution of these Cl atoms from axially symmetrical, which is caused by the interaction of one of their unshared electron pair with the central atom. The other unshared electron pair of the equatorial Cl atoms does not participate in any interaction with another atoms. The population of its orbital is close to 2e (Table 2). Unfortunately, the η value for the Cl atoms in the $SiCl_4 \cdot 2NC_5H_5$ complex has not established. However, for another $SiCl_4$ complexes it average 0.4 [4]. Using this value as well as the experimental ^{35}Cl NQR frequency for the $SiCl_4 \cdot 2NC_5H_5$ complex [3] and Townes' and Dailey's approximation (see, e.g., [14]) one may estimate the p_x-orbital populations of the Cl atoms in the $SiCl_4 \cdot 2NC_5H_5$ complex (at Np_y=2). This value (1.909e) is slightly less than that obtained from MNDO calculations (Table 2). This is caused by the lesser sensitivity of the latter to the change of population of the unshared electron pair orbitals of the Cl atoms than the experimental NQR data. When the distance d decreases the p_z-orbital populations of the Cl atoms increase essentially (by 0.123e). The analysis of the p-orbital populations of these atoms shows that the negative charges at the Cl atoms increase at the complex formation, in the main, as a result of their p_σ-electron density increase. This is in accord with the substantial decrease of the ^{35}Cl NQR frequency of the $SiCl_4 \cdot 2NC_5H_5$ complex in comparison with $SiCl_4$ (approximately by 1.2 MHz) [3].

The p_y- and p_z-orbital populations of the Si atom are decreased by the same value (by 0.089e) at the complex formation. This is quite clear if to take into consideration the geometry of the $SiCl_4$ - $2NC_5H_5$ system. The p_x-orbital population of the Si atom increases by the substantial value (by 0.186e) when d is decreased. Nevertheless herewith the positive charge at the Si atom increases. This is caused by so that still greater electron density displaces from the Si atom along the Si-Cl bonds.

When the $SiCl_4 \cdot 2NC_5H_5$ complex of the structure I is formed, the p_x-orbital population of the N atom increases by 0.061e, but the $p_z(p_\pi)$-orbital by 0.224e. Only the population of its p_y-orbital somewhat decreases (by 0.049e), and the population of 2s-orbital decreases noticeably (by 0.104e). The total electron density on the N atom increases at the complex formation. This atom is no electron donor in the complex, but, on the contrary, it is electron acceptor.

When the distance d in the system $SiCl_4$ - $N(CH_3)_3$ decreases from 200 Å to 2.25 Å the populations of both unshared electron pair orbitals (p_x- and p_y-orbitals) of the axial Cl atom remain constant (1.987e). Only the population of its p_z-orbital slightly increases (by 0.031e). The same population of the p_x- and p_y-orbitals of this Cl atom in the $SiCl_4 \cdot N(CH_3)_3$ complex conforms with the practically axial symmetry of its electron distribution, which is established experimentally [4]. The p_z-orbital population of the equatorial Cl atoms at such decrease of d increases more substantially (by 0.095 e). The populations of their p_x- and p_y-orbitals at d=200 Å are the same, and at d=2.25 Å they differ unsignificantly. At the same time, the large η value is observed for the equatorial Cl atoms in this complex [4]. Therefore the essential distinction between the populations of their p_x- and p_y-orbitals one may expect. The p_x-orbital populations of these atoms, calculated using the experimental ^{35}Cl NQR frequencies and asymmetry parameters (at Np_y=2e) is essentially smaller (1.983e for one and 1.907e for the two another equatorial Cl atoms), than that obtained from MNDO calculations. This is caused by the lesser sensitivity of the latter to the change of the p_x- and p_y-orbital populations of the Cl atom, than the experimental NQR data. The greater p_z-orbital population of the axial Cl atom (1.529e), than that of the equatorial ones (1.488e), at d=2.25e corresponds to the lower ^{35}Cl NQR frequency of the former in comparison with the latter [3].

Analogously to the system $SiCl_4$ - $2NC_5H_5$ of the structure I one may suppose that the p_x-orbitals of the equatorial Cl atoms in the $SiCl_4$ - $N(CH_3)_3$ system of the structure III participate in lateral overlapp with the Si atom p_x-orbital. Herewith the multicenter bond is formed (see above). However, in the latter system the sum of electron deficit on the p_x-orbitals of the equatorial Cl atoms at d=200 Å is essentially less than the p_x-orbital population of the Si atom. This is caused by the participation of the latter orbital in the axial Si-Cl bond formation as well. At the distance d decrease from 200 Å to 2.25 Å the electron deficit on the p_x-orbitals of the equatorial Cl atoms slightly decreases as in the $SiCl_4$ - $2NC_5H_5$ system.

At d decrease from 200 Å to 2.25 Å the population of the unshared electron pair orbital (p_x-orbital) of the N atom decreases by 0.264e. The electron density from this orbital passes, in the main, on the Si atom p_x-orbital. However, it does not stay on the latter. It follows from that, that at such decrease of d the population of this Si atom orbital increases only by 0.011e. The population of the axial Cl atom p_σ-orbital, which participates in formation of the axial $Cl\text{-}Si{\leftarrow}N$ fragment of the complex, increases insignificantly too (see above). The electron density from the ligand passes, mainly, on the p_σ-orbitals of the equatorial Cl atoms.

At the decrease of the distance d between the components of the system $SiCl_4$ - $OC[N(CH_3)_2]_2$ from 200 Å to 1.7 Å, when the complex of the structure IV is formed, the $p_x(p_\pi)$-orbital population of the O atom increases (by 0.275e), but that of the C atom in carbonyl group decreases (by 0.105e). Herewith, the population of their $p_z(p_\sigma)$-orbitals decreases and of p_z-orbital of the Si atom, the symmetry axis of which coincides with the σ-bond of this group, increases. Evidently, the electron density from the carbonyl C atom and dimethylaminogroups passes across the O atom σ– and π-orbitals to the electron acceptor,

Table 1. The changes of atomic charges on the atoms in the $SiCl_4$ - nL system ($\Delta q=|q(d_{min})|-|q(d=200\ Å)|$) at the distance d between its components decreasing from d=200 Å to d_{min} value, when the heat of complex formation is minimum. In the brackets at the nL the structure of a complex is shown

nL	Δq, e					
	Si	Cl_{ax}	Cl_{eq}	N	C	O
$2NC_5H_5$ (I)	0.109	-	0.158	0.131	0.096	-
$2NC_5H_5$ (II)	0.315	0.220	0.152	0.140	-	-
$N(CH_3)_3$ (III)	0.134	0.031	0.101	-0.052	-0.093	-
$N(CH_3)_3$ (IV)	-0.035	0.115	0.028	-0.055	-0.101	-
$OC[N(CH_3)_2]_2$ (IV)	0.081	0.139	0.059	-0.086 0.011	0.185	0.207
$OC[N(CH_3)_2]_2$ (III)	0.272	0.054	0.155	-0.074 0.024	0.176	0.209

and localizes on its Cl atoms. The p_z-orbital population of the axial Cl atoms increases by 0.109e and of the equatorial ones to a considerably lesser extent (by 0.031e). In the $SiCl_4 \cdot OC[N(CH_3)_2]_2$ complex the p_z-orbital population of the axial Cl atoms is by 0.068e larger, than of the equatorial ones. This corresponds to the essentially smaller NQR frequency of the axial Cl atoms in this and another trigonal-bipyramidal complexes of tetrachlorides of metals of group IVA, than of the equatorial ones (see, e.g., [1-3]). The p_x- and p_y-orbital populations of the axial Cl atoms are practically the same. They are close to 2e. This corresponds to the small η value for these atoms in $SiCl_4 \cdot OC[N(CH_3)_2]_2$ [4]. The difference of the populations of these orbitals for the equatorial Cl atoms somewhat greater, than for the axial ones, however it is not great too. At the same time, the great η value is observed for the equatorial Cl atoms in this complex [4]. The p_x-orbital populations of the equatorial Cl atoms, calculated using the experimental ν and η values for this complex and at $Np_y=2$, are noticeable smaller (1.897e), than those obtained from MNDO calculations (Table 2). This is caused by a low sensitivity of the latter to the change of the p_x- and p_y-orbital populations of the Cl atoms.

Conclusions

At the decrease of the distance d between the components of the system $SiCl_4$ - nL of octahedral or trigonal-bipyramidal structure the polarization of the ligand under the action of the positively charged Si atom, just as the polarization of the Si-Cl bonds under the action of the negatively charged heteroatom of the ligand becomes stronger. The former gives rise to the increase of negative charge at the ligand heteroatom and the latter to the increase of the p_σ-electron density of the Cl atoms. The nearest to the ligand Si-Cl bonds (cis-bonds) are polarized stronger than the more removed ones (trans-bonds). At the sufficiently small distances d in addition to this polarization the transfer of electron density takes place from an electron donor to an acceptor. Herewith it gets across often not from complex-forming center of a ligand, but from its peripheral atoms and groups. The complex-forming centers initiate the complex formation and serve as the conductors of electron density from the remote atoms of a ligand on the Cl atoms nearest to it, in the main.

The lateral overlapp of corresponding p-orbitals of the Si atom and equatorial Cl atoms produces some contribution to the electron density redistribution in the $SiCl_4$ molecule when it participates in complex formation with organic ligands. However the main contribution to this redistribution the p_σ-orbitals of the Si and Cl atoms produce.

As a matter of record it is impossible to say about participation of unshared electron pairs of a ligand heteroatom in complex formation.

Table 2. The population of atomic orbitals in the $SiCl_4$ - nL system (a structure type is shown) at the distance between its components d=200 Å and at the optimal one for a complex.

Atom	Orbital	$SiCl_4$ - $2NC_5H_5$ (I)		$SiCl_4$ - $N(CH_3)_3$ (III)		$SiCl_4$ - $OC[N(CH_3)_2]_2$ (IV)	
		Population at the distance					
		d=200 Å	d=2.0 Å	d=200 Å	d=2.25 Å	d=200 Å	d=1.7 Å
Si	p_x	0.177	0.363	0.456	0.445	0.529	0.464
	p_y	0.527	0.438	0.508	0.465	0.525	0.469
	p_z	0.527	0.438	0.509	0.466	0.199	0.337
Cl_{ax}	p_x	-	-	1.987	1.987	1.991	1.992
	p_y	-	-	1.987	1.987	1.959	1.988
	p_z	-	-	1.498	1.529	1.482	1.591
Cl_{eq}	p_x	1.956	1.988	1.981	1.982	1.991	1.990
	p_y	1.991	1.992	1.981	1.986	1.957	1.983
	p_z	1.494	1.617	1.393	1.488	1.492	1.523
N	2s	1.715	1.611	1.564	1.629	-	-
	p_x	1.376	1.326	1.755	1.491	-	-
	p_y	0.987	1.047	1.060	1.136	-	-
	p_z	1.154	1.378	1.060	1.135	-	-
C	p_x	-	-	-	-	0.733	0.628
	p_y	-	-	-	-	0.790	0.815
	p_z	-	-	-	-	0.817	0.730
O	p_x	-	-	-	-	1.439	1.714
	p_y	-	-	-	-	1.876	1.911
	p_z	-	-	-	-	1.164	1.208

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Received: September 27, 1994 - Accepted: October 26, 1994 - Received in camera-ready format: January 3, 1995