Molecular structure of potassium [*N,N'*-bis(salicylideneamino)nitroguanidonate-*N,N',O,O'*]nickel(II), the product of condensation of (salicylideneamino)nitroguanidine with salicylaldehyde on a Ni^{2+} ion template

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The crystal structure of the product of the condensation of (salicylideneamino)nitroguanidine with salicylaldehyde on a Ni^{2+} ion template, $K[Ni(C_{15}H_{10}N_5O_4)] \cdot DMF$, has been studied. It was established that a planar Ni complex, consisting of isolated $[NiL]^-$ anions and solvated $[K^+ \cdot DMF]$ cations, is formed. The negative charge of the anion is localized mainly on the O atoms of the nitro group. The nitroguanidine fragment of the ligand occurs in the tautomeric form, which was not reported previously.

Key words: nitroguanidine, salicylidene nickel(II) complex; template condensation; potassium [*N,N'*-bis(salicylideneamino)nitroguanidonate-*N,N',O,O'*]nickel(II), molecular structure, X-ray structural study.

Derivatives of guanidine containing amino groups are very promising reagents for the synthesis of metallocomplexes with Schiff bases, including macrocyclic complexes.[1] The template effect of the ions of some metals (Ni^{2+}, Cu^{2+}, and VO^{2+}) is known. When coordinated to ligands, these ions change the reactivities of the ligands, which makes it possible to prepare complexes with rather complex structures based on these ligands.[2]

From this standpoint, the reaction between (salicylideneamino)nitroguanidine and salicylaldehyde in the presence of Ni^{2+} ions, which are used as template centers, is of interest. It is remarkable that condensation with aldehyde at the free amino group of the ligand does not occur in the absence of Ni^{2+} ions. The reaction produced a complex that was identified as potassium [*N,N'*-bis(salicylideneamino)nitroguanidonate-*N,N',O,O'*]nickel(II) (**1**) from the data of magnetochemistry, conductivity, mass spectrometry, and ^1H and ^{13}C NMR spectroscopy. However, many of the structural features of complex **1**, specifically, the nature of bonding of potassium to the complex anion, still remained unknown. Therefore, we carried out an X-ray structural study of complex **1**. In the present work, we discuss the results of this study.

Experimental

Aminonitroguanidine was prepared according to the known procedure.[3] (Salicylideneamino)nitroguanidine was synthesized by a method reported previously.[4]

Potassium [*N,N'*-bis(salicylideneamino)nitroguanidonate-*N,N',O,O'*]nickel(II) (1). Salicylaldehyde (1 mL, 0.01 mol) was added to a hot (80—90 °C) solution of (salicylideneamino)nitroguanidine (2.23 g, 0.01 mol) and KOH (3 g) in water (70 mL), and then a solution of $NiCl_2 \cdot 6H_2O$ (2.37 g, 0.01 mol) in hot water (10 mL) was added dropwise with stirring. Complex **1**, which crystallized after cooling, was filtered off, washed with water, and dried in a vacuum desiccator. The yield was 2.1 g (49%). Complex **1** is soluble in water, DMF, and DMSO and slightly soluble in alcohol, acetone, and ether.

Crystals suitable for X-ray structural study were grown by isothermal evaporation of a solution of **1** in DMF.

The red platelet-like crystals of **1** ($C_{18}H_{17}KN_6NiO_5$, M = 459.19) are triclinic. At 25°C, a = 10.100(2) Å, b = 10.130(2) Å, c = 10.848(2) Å, α = 92.31(3)°, β = 91.35(3)°, γ = 116.16(3)°, V = 994.3(3) $Å^3$, d_{calc} = 1654 g cm^{-3}, Z = 2, space group $P\bar{1}$. The unit cell parameters and intensities of 3290 independent reflections were measured on an automated four-circle CAD-4 diffractometer (25°C, Mo-Kα radiation, graphite monochromator, $\theta/\frac{5}{3}\theta$ scan mode, θ < 24°).

The structure was solved by the direct method and refined anisotropically by the full-matrix least-squares method. All H atoms were located from the difference electron density synthesis and refined anisotropically. The final values of the *R*

[†] Deceased in 1995.

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Table 1. Atomic coordinates ($\times 10^4$; $\times 10^3$ for H atoms) and equivalent isotropic (isotropic for H atoms) temperature factors ($\times 10^3$)

Atom	*x*	*y*	*z*	$U_{eq/iso}/Å^2$
Ni(1)	2282(1)	1799(1)	4501(1)	29(1)
K(1)	424(1)	1512(1)	1612(1)	38(1)
O(1)	2722(3)	3236(3)	3373(2)	36(1)
O(2)	1983(3)	532(3)	3141(2)	35(1)
O(3)	2255(4)	3486(3)	9980(3)	53(1)
O(4)	1497(4)	1235(3)	9261(3)	50(1)
O(5)	−1512(4)	2642(4)	1552(4)	75(1)
N(1)	2602(3)	3021(3)	5883(3)	29(1)
N(2)	1834(4)	835(4)	6891(3)	33(1)
N(3)	1785(3)	389(4)	5640(3)	30(1)
N(4)	2495(4)	3159(4)	8013(3)	35(1)
N(5)	2075(4)	2588(4)	9096(3)	36(1)
N(6)	−3882(4)	2281(4)	1354(4)	48(1)
C(1)	2269(4)	2250(5)	6992(4)	32(1)
C(2)	3098(4)	4450(5)	5901(4)	32(1)
C(3)	3450(4)	5300(4)	4857(4)	31(1)
C(4)	3263(4)	4655(4)	3633(4)	31(1)
C(5)	3692(5)	5604(5)	2642(4)	37(1)
C(6)	4246(5)	7094(5)	2851(4)	39(1)
C(7)	4410(5)	7723(5)	4048(5)	42(1)
C(8)	4037(5)	6861(5)	5019(5)	39(1)
C(9)	1361(4)	−1016(5)	5410(4)	36(1)
C(10)	1203(4)	−1699(4)	4218(4)	32(1)
C(15)	688(5)	−3248(5)	4125(5)	40(1)
C(14)	495(5)	−3984(5)	3013(5)	49(1)
C(13)	832(5)	−3203(5)	1945(5)	44(1)
C(12)	1336(5)	−1696(5)	2005(4)	41(1)
C(11)	1528(4)	−904(4)	3136(4)	32(1)
C(16)	−2440(6)	3064(6)	1296(5)	56(1)
C(17)	−4936(7)	2837(8)	1015(7)	66(2)
C(18)	−4460(9)	790(7)	1781(8)	76(2)
H(2)	323(5)	496(5)	671(4)	53(13)
H(5)	356(4)	516(4)	188(4)	29(11)
H(6)	458(4)	768(4)	209(3)	31(10)
H(7)	478(4)	861(5)	415(4)	37(13)
H(8)	422(5)	720(5)	587(4)	52(14)
H(9)	119(4)	−153(4)	607(4)	30(11)
H(12)	159(4)	−116(4)	129(4)	34(11)
H(13)	70(5)	−365(5)	113(4)	49(13)
H(14)	15(5)	−498(5)	290(4)	57(14)
H(15)	47(5)	−370(5)	486(5)	62(16)
H(16)	−225(5)	402(5)	106(4)	63(15)
H(171)	−443(6)	383(6)	81(5)	69(17)
H(172)	−547(7)	221(7)	27(6)	100(22)
H(173)	−555(7)	275(6)	176(6)	95(21)
H(181)	−540(8)	69(7)	232(7)	126(27)
H(182)	−503(9)	27(9)	120(7)	132(34)
H(183)	−365(9)	69(8)	205(7)	129(30)

Table 2. Bond lengths (*d*) in the structure of **1**

Bond	*d*/Å	Bond	*d*/Å
Ni(1)—N(1)	1.832(3)	N(3)—C(9)	1.304(5)
Ni(1)—N(3)	1.832(3)	N(4)—N(5)	1.325(5)
Ni(1)—O(2)	1.844(3)	N(4)—C(1)	1.360(5)
Ni(1)—O(1)	1.847(3)	N(6)—C(16)	1.321(6)
K(1)—O(5)	2.667(3)	N(6)—C(17)	1.452(6)
K(1)—O(4B)	2.709(3)	N(6)—C(18)	1.459(7)
K(1)—O(2)	2.757(3)	C(2)—C(3)	1.406(6)
K(1)—O(3A)	2.780(3)	C(3)—C(8)	1.424(6)
K(1)—O(1)	2.835(3)	C(3)—C(4)	1.427(6)
K(1)—O(4A)	2.846(3)	C(4)—C(5)	1.416(6)
K(1)—N(2B)	3.033(4)	C(5)—C(6)	1.365(6)
O(1)—C(4)	1.308(4)	C(6)—C(7)	1.396(7)
O(2)—C(11)	1.318(4)	C(7)—C(8)	1.348(6)
O(3)—N(5)	1.248(4)	C(9)—C(10)	1.414(6)
O(4)—N(5)	1.253(4)	C(10)—C(11)	1.412(6)
O(5)—C(16)	1.221(6)	C(10)—C(15)	1.417(6)
N(1)—C(2)	1.306(5)	C(15)—C(14)	1.356(7)
N(1)—C(1)	1.426(5)	C(14)—C(13)	1.392(7)
N(2)—C(1)	1.299(5)	C(13)—C(12)	1.379(6)
N(2)—N(3)	1.406(4)	C(12)—C(11)	1.400(6)

factors were as follows: R_1 = 0.0343 for 2043 reflections with $I > 2\sigma(I)$, wR_2 = 0.0831. All calculations were carried out using the SHELXTL PLUS program package (Version 5). Atomic coordinates are given in Table 1; bond lengths and bond angles are listed in Tables 2 and 3, respectively.

Results and Discussion

The crystal structure of potassium [*N,N'*-bis(salicylideneamino)nitroguanidonate-*N,N',O,O'*]nickel(II) DMF solvate, $K[Ni(C_{15}H_{10}N_5O_4)] \cdot Me_2NCHO$ (or $K[NiL] \cdot DMF$), (**1**) consists of isolated $[NiL]^-$ anions and solvated $[K^+ \cdot DMF]$ cations, which form contact ion pairs. Figure 1 shows a fragment of the structure that presents the nature of the interactions between the three components (the anion, the cation, and the solvate DMF molecule).

All atoms of the $[NiL]^-$ anion, except for the N(4)N(5)O(3)O(4) fragment, are nearly in a single plane (the maximum deviation of the C(4) atom from the mean plane is 0.080 Å); the planar N(4)N(5)O(3)O(4) fragment is slightly bent away from this plane (the N(2)—C(1)—N(4)—N(5) torsion angle is 9.0°).

The coordination polyhedron about the Ni(1) atom is square planar; the Ni(1)—O(1) [1.847(3) Å] and Ni(1)—O(2) [1.844(3) Å] bond lengths are close to the corresponding values in *N,N'*-ethylenebis(salicylideneiminato)nickel[5] (**2**) and *N,N'*-ethylenebis(2-hydroxynaphthylideneiminato)nickel[6] (**3**) complexes (the average Ni—O distances are 1.853(2) and 1.849(2) Å in **2** and **3**, respectively), whereas the Ni(1)—N(1) [1.832(3) Å] and Ni(1)—N(3) [1.832(3) Å] bonds in complex **1** are slightly shortened (the average Ni—N distances are 1.848(2) and 1.840(2) Å in **2** and **3**, respectively).

Table 3. Bond angles (ω) in the structure of **1**

Angle	ω/deg	Angle	ω/deg	Angle	ω/deg
N(1)—Ni(1)—N(3)	82.63(14)	O(1)—K(1)—N(2B)	73.54(9)	C(2)—C(3)—C(8)	119.3(4)
N(1)—Ni(1)—O(2)	178.07(12)	O(2)—K(1)—Ni(1)	103.94(9)	C(2)—C(3)—C(4)	122.3(4)
N(3)—Ni(1)—O(2)	95.79(13)	C(4)—O(1)—Ni(1)	30.87(6)	C(8)—C(3)—C(4)	118.4(4)
N(1)—Ni(1)—O(1)	96.30(13)	C(11)—O(2)—Ni(1)	126.1(2)	O(1)—C(4)—C(5)	118.1(4)
N(3)—Ni(1)—O(1)	177.71(13)	C(2)—N(1)—C(1)	126.8(3)	O(1)—C(4)—C(3)	123.7(3)
O(2)—Ni(1)—O(1)	85.32(11)	C(2)—N(1)—Ni(1)	121.4(3)	C(5)—C(4)—C(3)	118.2(4)
O(5)—K(1)—O(4B)	96.02(11)	C(1)—N(1)—Ni(1)	125.8(3)	C(6)—C(5)—C(4)	120.9(4)
O(5)—K(1)—O(2)	144.31(11)	C(1)—N(2)—N(3)	112.8(2)	C(5)—C(6)—C(7)	120.9(4)
O(4B)—K(1)—O(2)	94.02(9)	C(2)—N(2)—N(3)	109.3(3)	C(8)—C(7)—C(6)	120.1(4)
O(5)—K(1)—O(3A)	93.18(12)	C(9)—N(3)—N(2)	115.5(3)	C(7)—C(8)—C(3)	121.5(4)
O(4B)—K(1)—O(3A)	119.08(10)	C(9)—N(3)—Ni(1)	126.5(3)	N(3)—C(9)—C(10)	124.7(4)
O(2)—K(1)—O(3A)	111.42(10)	N(2)—N(3)—Ni(1)	117.9(2)	C(9)—C(10)—C(11)	122.7(4)
O(5)—K(1)—O(1)	107.86(11)	N(5)—N(4)—C(1)	119.6(3)	C(9)—C(10)—C(15)	117.6(4)
O(4B)—K(1)—O(1)	146.58(9)	O(3)—N(5)—O(4)	119.9(3)	C(11)—C(10)—C(15)	119.6(4)
O(2)—K(1)—O(1)	53.12(8)	O(3)—N(5)—N(4)	115.9(3)	C(14)—C(15)—C(10)	121.1(5)
O(3A)—K(1)—O(1)	83.30(9)	O(4)—N(5)—N(4)	124.1(3)	C(15)—C(14)—C(13)	119.5(5)
O(5)—K(1)—O(4A)	113.80(12)	C(16)—N(6)—C(17)	123.0(5)	C(12)—C(13)—C(14)	120.8(5)
O(4B)—K(1)—O(4A)	76.57(11)	C(16)—N(6)—C(18)	119.1(5)	C(13)—C(12)—C(11)	121.2(5)
O(2)—K(1)—O(4A)	101.83(9)	C(17)—N(6)—C(18)	117.8(5)	O(2)—C(11)—C(12)	118.8(4)
O(1)—K(1)—O(4A)	45.25(9)	N(2)—C(1)—N(4)	130.1(4)	O(2)—C(11)—C(10)	123.4(4)
O(5)—K(1)—N(2B)	112.62(9)	N(2)—C(1)—N(1)	117.3(3)	C(12)—C(11)—C(10)	117.8(4)
O(4B)—K(1)—N(2B)	84.87(11)	N(4)—C(1)—N(4)	112.6(3)	O(5)—C(16)—N(6)	125.6(5)
O(2)—K(1)—N(2B)	54.38(9)	N(1)—C(2)—C(3)	125.4(4)		

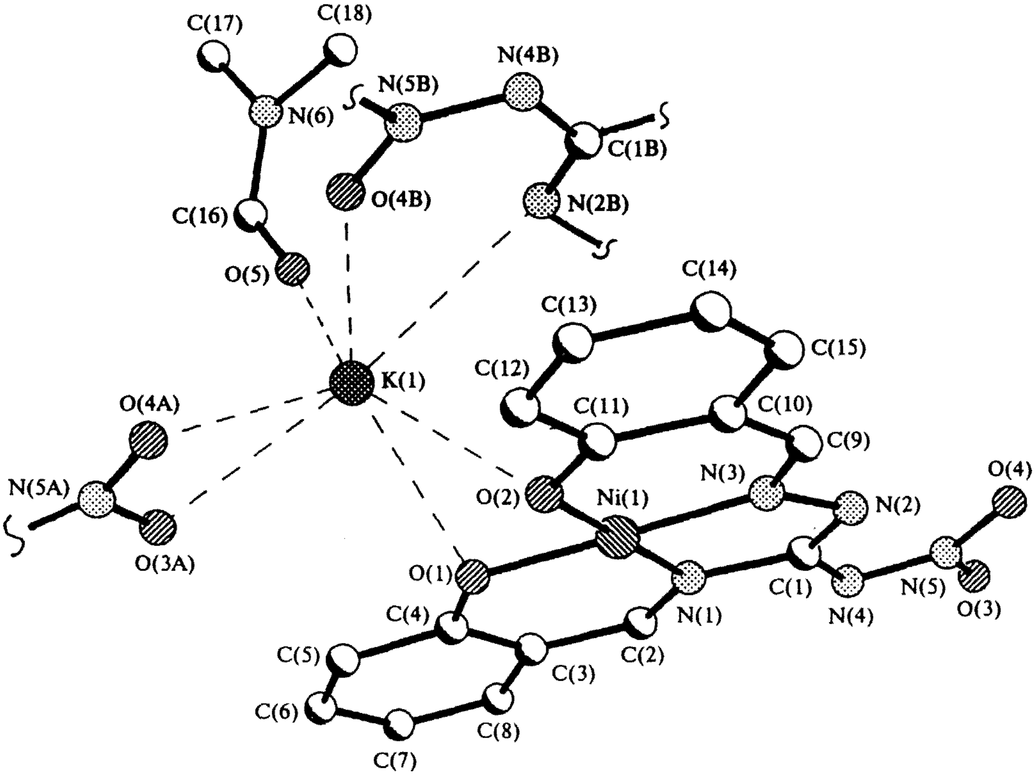
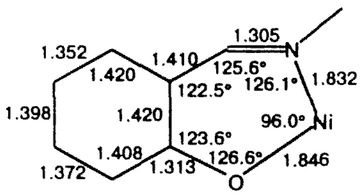


Fig. 1. Fragment of the structure of **1**. The N(5A), O(3A), and O(4A) atoms belong to cation A, which is related to the reference cation by the transformation *x*, *y*, *z* − 1; the N(2B) and O(4B) atoms belong to anion B, which is derived by the transformation *x*, −*y*, 1 − *z*.

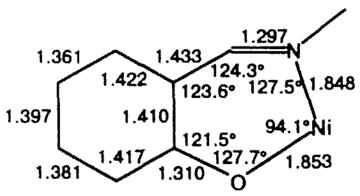
It is appropriate to consider the salicylideneiminate and aminonitroguanidine fragments of ligand L of molecule **1** separately, because these fragments have peculiar features.

The geometric parameters of the two salicylideneiminate fragments in the structure of **1** are identical to within twice the error (see Tables 2 and 3). For complex **1**, the bond lengths (Å) and bond angles averaged over both these fragments are given in Scheme 1; the corresponding values for complex **2** are given in Scheme 2.

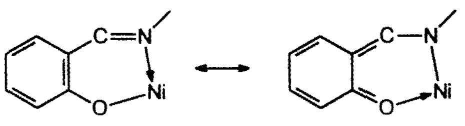
Scheme 1



Scheme 2



These data are indicative of partial localization of double bonds, which was noted previously for these complexes (see, for example, Ref. 7), and a substantial contribution of the quinoneamine canonical form (**B**) along with the ordinary aldiminate form (**A**) to the resonance, which describes the bonding in salicylaldiminate complexes.



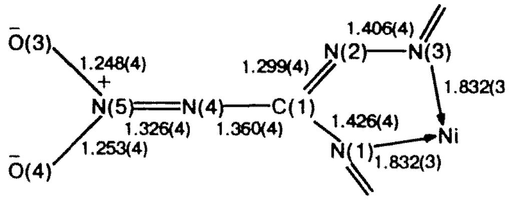
A **B**

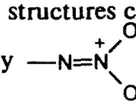
Elongation of the C—C bonds "nearest" to the chelate cycle and shortening of the "remote" C—C bonds in the benzene ring were noted even in early works on chelate salicylaldiminate complexes of transition metals.[8,9] It was pointed out that this effect is not accidental, *i.e.*, this effect does not result from errors in solving the structure but reflects the electron delocalization throughout the bicyclic system and is independent of the nature of the metal atom. The average bond lengths in the salicylideneiminate fragment of complex **2**, which are given in Scheme 2, as well as the literature data[8,9] are close to those found in complex **1**, with one exception: in all molecules studied previously, the C(Ar)—CH(=N—) bond is somewhat longer and averages 1.43 Å (1.406(6) and 1.414(6) Å for two fragments of complex **1**).

It is noteworthy that localization of the double bonds in one of the benzene cycles of molecule **1** is most pronounced: the C(5)—C(6) bond length [1.365(6) Å] is smaller than that typically observed in analogous molecules (1.38—1.39 Å), and the C(7)—C(8) bond [1.348(6) Å] is also shortened. In the second benzene cycle, the corresponding bond lengths [C(12)—C(13), 1.379(6) Å; and C(14)—C(15), 1.356(6) Å] agree with those found previously in complexes **2** and **3**.

The aminonitroguanidine fragment has a pronounced zwitterionic form (Scheme 3, bond lengths (Å) are indicated).

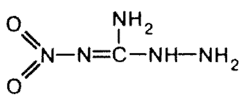
Scheme 3



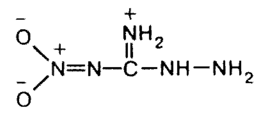
The N(2)—N(3) bond length is nearly identical to that typical of the N—N single bond (1.401 Å)[10] between nitrogen atoms in planar configurations. The data on the N(4)=N(5) bond lengths are unavailable in the literature because, to our knowledge, structures containing a sufficiently well defined moiety  $—N{=}\overset{+}{N}\begin{smallmatrix}O^-\\ O^-\end{smallmatrix}$ have not been reported previously.

The N(4)—N(5) bond [1.326(4) Å] is slightly longer than the aromatic N---N bond in pyridazine (1.304 Å) but shorter than the N---N bonds in pyridazinium salts (1.350 Å).[10] As expected, the N(5)—O(3) [1.248(4) Å] and N(5)—O(4) [1.253(4) Å] bonds are longer than the standard distance (1.239 Å) calculated previously[10] from a large body of structural data (105 entries) on the NO_2 group. Note the clear-cut distinction between the different types of C—N bonds: the "purely" double C(1)=N(2) bond [1.299(5) Å], the "purely" single C(1)—N(1) bond [1.426(5) Å], and the intermediate C(1)---N(4) bond [1.360(5) Å].

The molecular structure of the uncoordinated $O_2N—N=C(NH_2)NHNH_2$ (L^1) precursor of the aminonitroguanidine fragment in complex **1** has not been studied previously, and therefore, which form of the molecule (neutral nitroimine (**C**) or zwitterionic guanidinium (**D**)) is preferred is still a question.

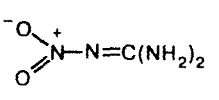

$O_2N—N{=}\overset{NH_2}{C}—NH—NH_2$

C


$\begin{smallmatrix}O^-\\ O^-\end{smallmatrix}\overset{+}{N}{=}N—\overset{\overset{+}{NH_2}}{C}—NH—NH_2$

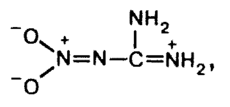
D

The possibility of the existence of nitroguanidinium analog, $(H_2N)_2C{=}NNO_2$, in the zwitterionic form has been discussed previously.[11,12] The preferred forms are



E

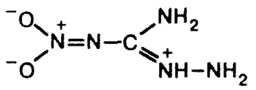
and



F

according to the literature data,[11] the concentrations of these forms in solutions are 30 and 20%, respectively. In the crystal,[13] form **E** predominates.

The geometry of coordinated "neutral" aminonitroguanidine L^1 (form **C**) in the $[NiL^1{}_2(H_2O)_2](NO_3)_2$ complex (see Ref. 14) as well as in the $[NiL^1{}_2Cl_2] \cdot 2H_2O$ complex, which we have studied previously, can be virtually considered as a superposition of the forms analogous to zwitterions **E** and **F**. However, it is, apparently, the aminonitroguanidine fragment that mainly compensates for the charge of the cation in molecule **1**, and therefore, the third canonical form (**G**) with negative charges on each O atom of the nitro group actually occurs for this structure:



G

Stabilization of the negatively charged $^{-}O(O^{-})N^{+}{=}$ group is, apparently, favored by formation of C=N double bonds by both terminal NH_2 groups of aminonitroguanidine (when complex **1** is produced by condensation on a Ni^{2+} template from (salicylideneamino)nitroguanidine $Ar(OH)CH{=}N{-}NH{-}C({=}N{-}NO_2)NH_2$ and salicylaldehyde $Ar(OH){-}CHO$) and by deprotonation of the NH group of the ligand L, which may result from the effect of an alkaline medium in the synthesis of **1**.

Therefore, the qualitative analysis of the geometric parameters of the $[NiL]^{-}$ anion makes it possible to consider this anion to be a system with localized bonds and a negative charge on the O atoms of the nitro group. Nevertheless, some degree of delocalization of the π electron density and a total negative charge throughout the virtually planar anion must not be ruled out. The fact that the O(3) and O(4) atoms, which carry a formal negative charge, are involved in ionic interaction with the K^+ cation, is an additional argument in support of the scheme suggested, in which double bonds and a negative charge are predominantly localized and an essential contribution of the quinoneamine form is observed.

It was found that the K^+ ion forms short contacts with six oxygen atoms (K^+...O, 2.667—2.846 Å) and with one nitrogen atom [K^+...N(2B), 3.033(4) Å], but the latter contact, apparently, results from the fact that a very short K^+...O(4B) distance [2.710(4) Å] necessarily brings the cation and the N(2B) atom of the O(4B)N(5B)N(4B)C(1B)N(2B) fragment of the anion B into proximity. The sum of the ionic radii of O^{2-} and K^+ (when the coordination numbers are equal to 4, these radii are 1.40 and 1.38 Å, respectively[15]) is 2.78 Å. Three out of the six K^+...O contacts are shorter than this value. The shortest contact is formed by the solvate DMF molecule [K^+...O(5), 2.667(4) Å]; next in order of strength are the contacts with the O(4B), O(2), and O(3A) atoms [2.709(3), 2.757(3), and 2.780(3) Å, respectively]. Note that the ionic K^+...O interaction in the structure of **1** is rather strong. Therefore, based on the values of the shortest interatomic distances, it may be concluded that contact ion pairs $[(DMF)K]^+$...$[NiL]^-$ are formed in the structure of **1**.

In conclusion, one more interesting fact should be mentioned. Over a period of years, extensive studies of the character of the coordination of the salts of monovalent metals M'X (M' = NH_4^+, Li^+, or Na^+) with salicylaldiminates of transition metals $ML^2{}_2$ (M = Co, Cu, Ni, or Zn) or with macrocycles containing the bis(salicylaldiminate) fragment $ML^2{}_2$ (see, for example, Refs. 16 and 17) have been carried out. In all binary complexes studied, the M' cations (and Ba^{2+} ions) are coordinated by $O(L^2)$ atoms to form complex aggregates $[M_mL^2{}_2M'_n]X$ (with different M : M' ratios) through the $O(L^2){-}M'$ ionic bonds; these associates contain, on occasion, water of crystallization or solvent molecules. However, as far as we know, analogous complexes with K^+ ions have not yet been structurally studied. Moreover, an unsuccessful attempt to prepare a binary complex with M' = K was reported in one of the works devoted to this line of investigation.[18]

Therefore, compound **1** is the first example of a structurally studied bimetal salicylaldiminate complex in a series of guanidine derivatives, in which an anion occurs in a previously unknown form with a negatively charged aminonitroguanidine group, while a potassium ion acts as an alkaline component.

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