Synthesis and X-ray Crystal Structure of the Bis-ligand Complex [Cr (PLSC-H)2].2H2O (Bis[4-(2-carbamoylhydrazin-1-ylidene k2,N1,O)-5-hydroxymethyl-2-methylpyridinium-3-olato-k O3] Chromium (II) dihydrate)

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ABSTRACT :

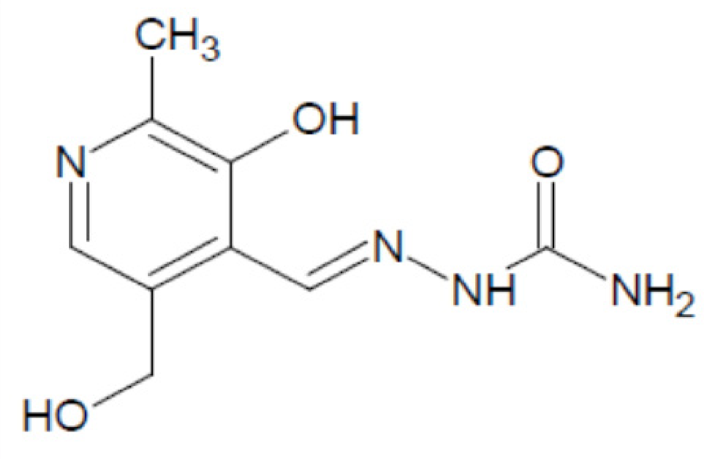
The title complex $[Cr(PLSC\text{-}H)_2].2H_2O$, is reported. The complex with molecular formula, C18 H26 Cr1 O8 N8 is an interesting metal(bis-ligand) complex, octahedral geometry, with a Schiff base derived from semicarbazide and pyridoxal (3-hydroxy-5-hydroxymethyl-2-methylpyridine-4-carboxaldehyde, form of vitamin B6). Ligand pyridoxal-semicarbazone (PLSC; H_2L) is mixed, tridentate ONO ligand. The PLSC ligand in the title complex is coordinated in monoanionic form (HL) by protonation of pyridine nitrogen, not hydrazine nitrogen, which has been a common practice so far. The Cr II environment is a distorted octahedral coordination, the equatorial plane of which is formed by two the tridentate ONO-coordinated pyridoxal-semicarbazone rings. Complex $[Cr(PLSC\text{-}H)_2].2H_2O$ crystallizes in monoclinic symmetry, in space group I 2/a, with lattice constants: a = 12.4645(14) Å, b =13.4321(15) Å c =13.7759(14) Å, α =90°, β =102.143(8) °, γ = 90°, V = 2254.8(4) Å3, Z = 4, F(000) = 1112.0, R1=0.0462(1849), wR2 = 0.1586 (2317). The reported complex was characterized by elemental analysis, IR spectra, conductometric and magnetochemically measurements, and X-ray diffraction.

KEYWORDS :

Pyridoxal-semicarbazone, Cr(II) complex, Synthesis, Characterization, Crystal structure.

1. INTRODUCTION :

Semicarbazones (SC) by West et al. (1991) can be considered as one potential chelating ligands, which possess different denticity and according it have significant biological activity as antifungal, anti-viral, anti-malarial and anti-tumor agents. Many published papers described the synthetic, structural and the biological activity of SC-based ligands. But, particular attention should be paid to the complex ligand, the combination of semi-carbazone (SC) and pyridoxal (PL) which is one form of vitamin B6.



Scheme 1. Structural formula of PLSC ligand

As was said before, complexes with SC-based ligand expressed a lot of biologically important properties (antiviral, antitumor and anti-inflammatory activities) Jakupec et al. (2008). Dehydration of SC with pyridoxal moiety leads to in the formation of Schiff base ligands PLSC (Scheme 1). The choice of the coordinated ligand(s) is most important because, sometimes the biological activity depends of it. Anyway, ligand PLSC can exist in neutral, mono- and dianionic forms depending on pH solution. Basically, PLSC ligand mostly is responsible for created biological activity of complex compound.

Actually,it has been published many papers about the syntheses and X-ray structure of complexes incorporating 3–hydroxy–5–hydroxymethyl–2–methyl– pyridine–4–carbaldehyde semi-carbazone (PLSC) ligand Knezevic et al. (2003) and Leovac et al.(2007). More recently, a plethora of papers have been published on the subject of coordination of the ligand PLSC with transition metals Poleti et al.(2003), Leovac et al. (2005), Jevtovic et al. (2010, 2011, 2014), Vidovic et al. (2011), A'Shidani et al. (2016) . Finally, a monograph (Jevtovic, 2010) on the complexation of the PLSC ligand and the biological activity of its complexes was published.

It is important to note that very similar compounds with the title's Chromium compound were published with Vojinovic-Jesic research group Vojinovic-Jesic et al. (2015) Those compound are $[Cr(PLSC)(PLSC–H)](NO_3)_2·H_2O$, $K[Cr(PLSC–H)(NCS)_3]·EtOH$.

It is important to note that PLSC ligand have three different forms in the coordination sphere, namely neutral (but zwitterionic) H_2L, monoanionic HL- (hydrazine deprotonation) and dianionic L^{2-} (both pyridinium and hydrazine deprotonation) forms (see Fig. 1). In this report we present PLSC-based complex $[Cr(PLSC\text{-}H)_2].2H_2O$ (Figure 2), which contains two PLSC ligands as HL- form, but in an unexpected way how was made that coordination form.

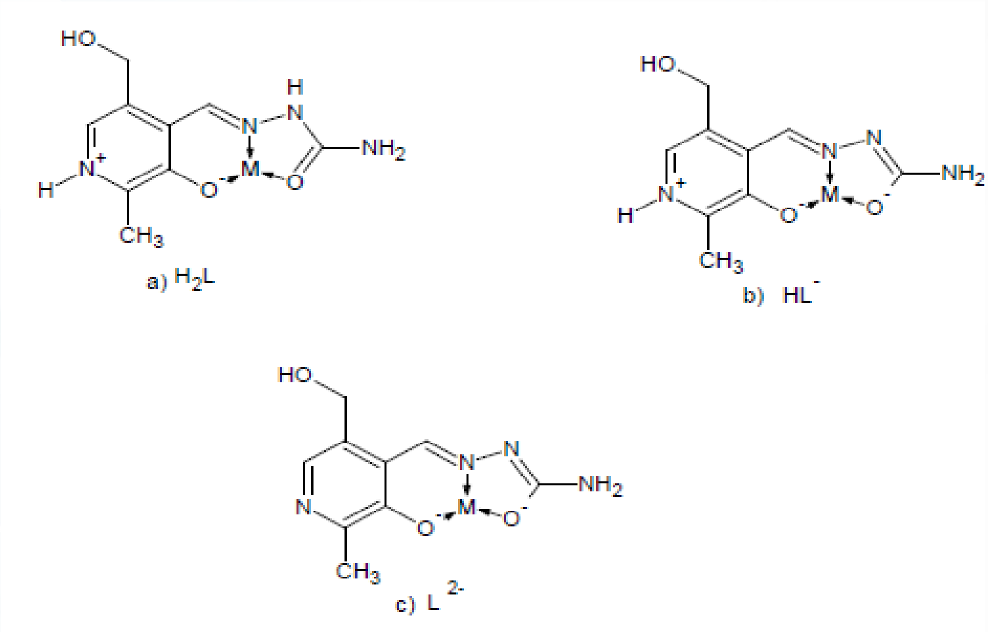


Fig. 1: Coordination models and ligand forms: a) neutral (H_2L), b) mono- (HL^-) and c) dianionic (L^{2-})

Namely, in only two cases so far, in complex $[Ni[PLSC\text{-}H)_2]$ $2H_2O$ Leovac et al. (2007) and in complexes $[Co[PLSC\text{-}H)_2]NCS$ H_2O and $[Co[PLSC\text{-}H)_2]$ H_2O Jevtovic et al. (2015) coordination participate the HL form. In these cases, the HL form was formed by de protonation of hydrazine hydrogen. Based on these crystalline structures, it has been concluded that the mono-ionic form of the PLSC ligand is always formed by deprotonation of the first hydrazine hydrogen. However, unexpectedly, in the complex we are reporting, the HL- form was formed by de protonation of pyridine hydrogen. So far, it has been established that de protonation of hydrogen from the pyridine ring always results in the dianionic form of the ligand in coordination. Accordingly, newly synthesized complex, may be considered a candidate for moving the boundaries in the understanding of the forms of the ligand PLSC.

So clear proof about is the IR of our Cr complex. Actually, here there is not a visible pick at 2800cm^{-1}, who is characteristic of the pyridine nitrogen (who is the protonated) ν (NH^+) vibrations from PL part of ligand. In our title compound the pyridine nitrogen (Vojinovic-Jesic et al. (2015)) is deprotonated, as we can see in X-ray crystal structure of Cr title compound (Figure 2). In other hand, IR spectrum of ligand showed clear bands in 2800cm^{-1}.

2. MATERIAL AND METODS :

Ligands were prepared according to the previously described procedures (Jevtovic et al.(2010)) and other chemicals were commercially obtained. Elemental (C, H, N) analysis by micro methods, was obtained in the Center for Instrumental Analysis, Faculty of Chemistry, Belgrade. Magnetic susceptibilities were measured by using a magnetic susceptibility balance MSB-MKL (Sherwood Scientific Ltd. Cambridge, England). A Jenway 4010 conductivity meter was used for measure molar conductivities of the freshly prepared 1×10^{-3} M solution. IR spectra (KBr disk) were recorded on a Thermo Nicolet (NEXUS 670 FT-IR) instrument. Crystallography Center Service from Research Lab of University Nizwa, was responsible for the X-ray analysis.

2.1. SYNTHESES OF PLSC $2H_2O$:

Ligand synthesis is performed by a very simple method in an air atmosphere, according to the procedure given below.

Semi-carbazide hydrogen chloride (1.07 g, 10 mmol) and H_2O (10 mL) were mixing, to which a warm solution of PLHCl (2.00 g, 10 mmol) in H_2O (10 mL) was added. To this solution, $Na_2CO_3 . 10H_2O$ (3.00 g, 15 mmol in 10 ml of water) was added in portions, and the heating of mixture was continuing in the next few minutes. After, solution need to cold in room temperature and after 20 hours are becoming the ligand crystals. The form of the synthesized PLSC ligand is always neutral. During complexation with a metal, in our case Cr, the form of the complexing ligand can also be in the form of a mono- or dianionic form. The obtained crystals are pale yellow in color, crystalline in structure, stable in air and very soluble in polar solvents. If not obtained satisfactory quality of the crystals, it is desirable to do recrystallization and thus obtain a more desirable and better quality of the ligand. Yield: 2.30 g (88%).

2.2. PREPARATION OF THE TITLE COMPOUND [CR (PLSC-H)$_2$]. $2H_2O$:

In the synthesis of the complex, it is always crucial to decide on the properly choice of a simple metal salt and in that way, the choice of central metal in the complex. In this work, we have a very specific situation that chromium metal oxide was used, not salt of chromium. This fact may be the reason for some unexpected results of the ligand form in coordination, which will be discussed in the discussion. The procedure for the synthesis of the chromium complex is also very simple, as is the synthesis of the ligand, and it is performed under very similar conditions.

The pyridoxal-semicarbazone ligand, H_2L, was prepared with standard procedure, in way that semi-carbazide hydrogen chloride was dissolved in warm H_2O and was added to a warm solution of PLSC with subsequent add HCl solution in drops for hold PH less than 5. It is very important to adjust the pH of solution because it has a direct effect on the ligand, in terms, of whether a protonated or deprotonated form of PLSC ligand will be created. White single crystals of title compound were obtained by the reaction of a hot water solution of the ligand with Cr_2O_3 (molar ratio 1:1).

The crystals are stable in air, well soluble in water as a solvent. They are not hygroscopic, but it is better to keep them away from potential sources of moisture, even from the air. Yield: 74%.

2.3. X-RAY CRYSTALLOGRAPHY :

A single crystal of 0.12 x 0.09 x 0.17 dimension was examined. The temperature was 296 K on a computing data collection For the X-ray measurements, single crystal of the complex were mounted on a glass fiber and examined at 296 K on a Bruker D8 Venture APEX diffractometer equipped with Photon 100 CCD area detector using graphite-monochromated Mo-Kα radiation [λ = 0.71073 Å]. In general, in the difference map the hydrogen atoms were 109 visible. Hydrogen atoms bound to carbon were initially positioned geometrically while the hydrogen atoms for the coordinated water molecules were located in the difference map. All hydrogen positions and isotropic displacement parameters were then refined in a separate cycle. Hydrogen positions were checked for feasibility by examination of the hydrogen-bonding network. Crystallographic data in the Cambridge Crystallographic Data Centre (CCDC, 12 Union Road, Cambridge CB2 IEZ, UK; e-mail: deposit@ccdc.cam.ac.uk) were deposit, CCDC deposition number is 1840928. Crystal data collection and structure refinement are given in Table 1.

Empirical formula	$C_{18}H_{26}CrN_8O_8$
Formula weight	534.47
Temperature	296 K
Wavelength	0.71073 Å
Crystal system	monoclinic
Space group	a/2 I

Unit cell dimensions	a= 12.4645(14) Å α= 90° b= 13.4321(15) Å β= 102.143(8)° c=13.7759(14) Å □ = 90°
Volume	2254.8(4) Å^3
Z	4

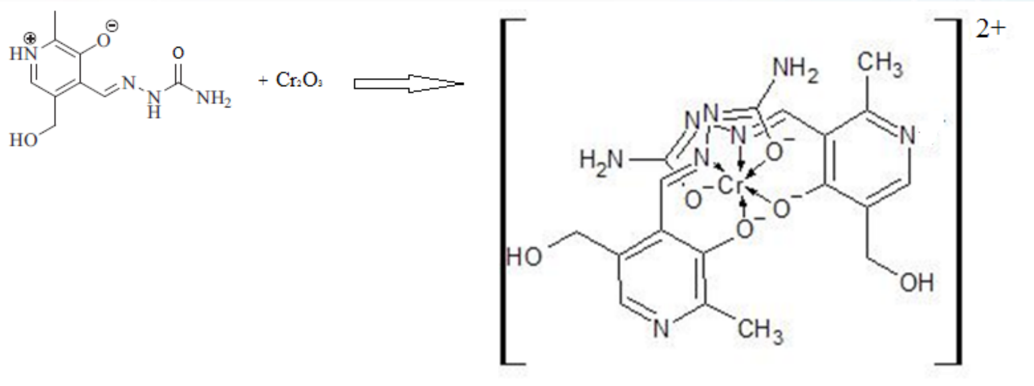
Table 1. Crystal data and structure refinement details of the complex

3. RESULTS :

The molecular structure for new complex is [Cr (PLSC-H)$_2$] 2H$_2$O shown in Figure 2. It crystallizes in I 2/a space group with one discrete neutral unit [Cr(PLSC-H)$_2$] and two H$_2$O molecule in inner sphere. The environment around the central chromium atom can be best described as a distorted octahedral geometry.

Scheme 2 shows the synthetic routes to the Cr (II) complex. The reactions of Cr_2O_3 with warm EtOH solution of PLSC 2H$_2$O, produced the title complex compound. In the reaction of Cr_2O_3 with ligand the molar ratio was 1:2. The complex is not stable in the air.

Very unexpected is oxidation state of Cr metal into compound, Cr^{2+}. Especially, because the starting compound for synthesis was oxide Cr(III) and during the oxidation is not happened oxidation into Cr(III). Measurement of the physical properties of Chromium (III) complexes is plagued by the ease which with the Cr(II) oxidized by air. But, Cr(II) by dilute aqueous acid non oxidize into in Cr(III), as happens with less pure metal (II) compound (Facker at al. (1965)) what probably happened with our title compound.



Scheme 2. Synthesis of complex compound

Chromium is one central metal in metabolism of living organisms, especially of carbohydrates and lipids (Praveen et al. (2015)), and one of the most controversial transition metals regarding his activity in living cell (Cindric et al. (2012)). The controversy is reflected in the fact that the Cr (VI), is toxic, especially of Cr(VI) which exist in the form of oxyanion and in that form can easier enter the cells (Vaidyanathan et al.(2005)). In other hand, Cr (III) is more stable and have less cytotoxic behavior for human cells (Beiderman et al. (1990)), However,insufficiency of Cr (III) in organism leads to diabetes and some cardiovascular disease (Cindric et al. (2012)).

The first Cr complexes with PLSC/PLTSC were presented in paper by L.S. Vojinovic´-Ješic´ et al (2015). Ethanolic solution of $Cr(NO_3)_3$ and $K_3[Cr(NCS)_6]$ (ligands in mole ratio 1:1 or 1:2) were formed complexes: $[Cr(PLSC)(PLSC–H)](NO_3)2H_2O$, $K[Cr(PLSC–H)(NCS)_3]$ EtOH. Those complexes have mer-octahedral structure. The coordination of Schiff-base is the usual tridentate ONO manner. The pyridoxalic fragment is zwitter ion, as the coordinated ligands: neutral (keto/thion) and monoanionic (enolic/thiolic) form. Our special attention will be focused in complex 2 ($K[Cr(PLSC–H)(NCS)_3]$ EtOH), because our new synthetized compound is with same ligand PLSC. In that complex PLSC is monoanion in coordination, with the deprotonation of hydrazinic nitrogen into ligand PLSC. This fact is consistent with the previous behavior of the ligand when it builds a mono-ionic form, described in all papers before our own. In the case of our complex $[Cr (PLSC\text{-}H)_2]$. $2H_2O$, it seems that the anomaly exist.

Namely, as can be seen from the attached crystal structure in Figure 2, a mono-ionic form is formed by the deprotonation of hydrogen on the pyridine ring, which never happened before. Deprotonation of hydrogen atom from pyridine ring is means that will forming dianionic form, if we follow previous cases of coordination (Jevtovic, (2010)) .

One of the reasons for this unusual coordination may be an accidental mistake in maintaining the middle of the solution, which should have been slightly acidic (pH ~5). The change in the pH of solutions environment may have been due to that chromium metal oxide was used, not salt of Cr, which is common. It is possible that unplanned hydrolysis occurred, because water was used as a solvent during the chromium complex synthesis reaction. In any case, an interesting situation is provided, which can be repeated in the following complexation reaction, using of similar synthesis conditions (use of metal oxide and water as solvents).

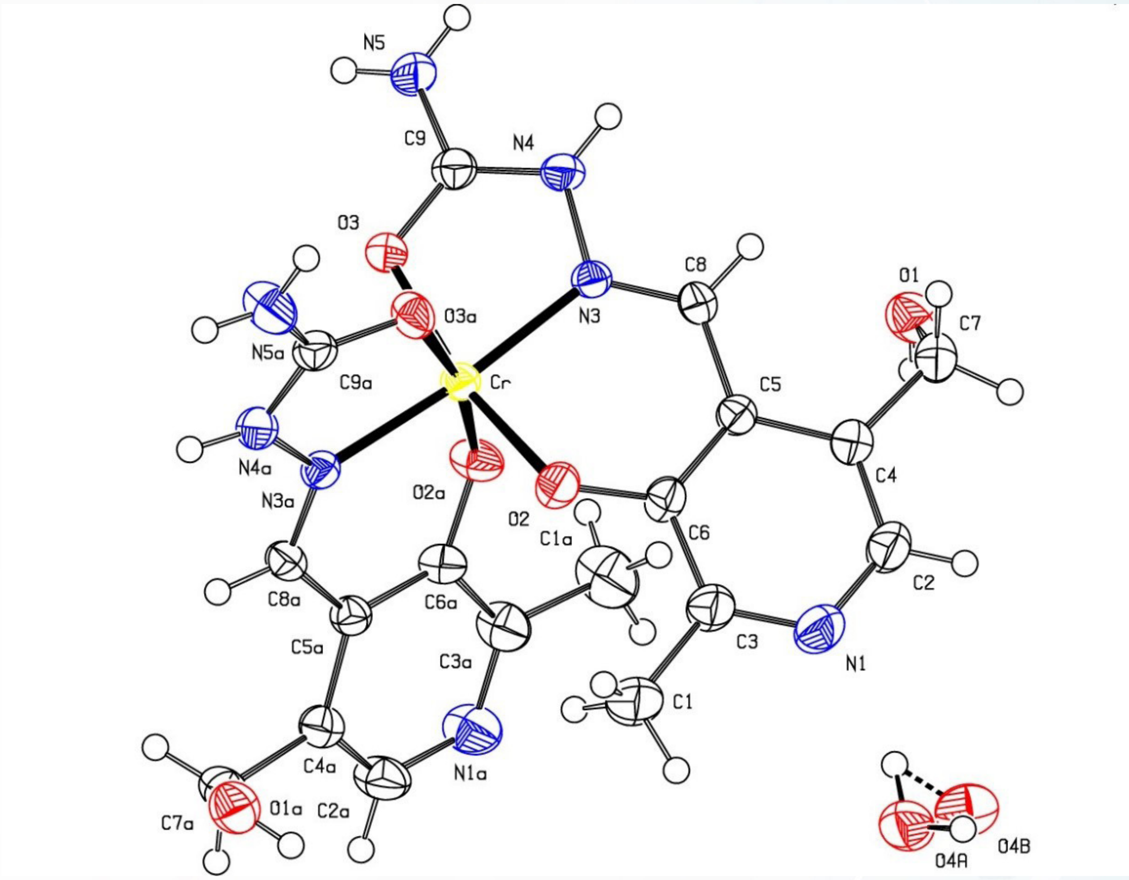


Fig. 2: The molecular structure for Cr (II) complex

Ligands PLSC is coordinated as tridentate, via oxygen atom of phenolic group, carbonyl oxygen and nitrogen of azomethine and carbonyl oxygen (PLSC), in that way forming six-membered pyridoxylidene and five membered semicarbazide metallocycles (Figure 1).

The dipolar zwitterion formed pyridoxalic fragment, and that is not surprise because ligands in their non-coordinated forms are zwitter-ions. According the zwitterionic structure, we can consider the value of the angle between the pyridine nitrogen atom and the neighboring carbon atoms, same as in the complexes (ca 125^{0}) (Jevtovic, (2010)). But, in title complex, the PLSC ligand, as monoanionic form was coordinated, with deprotonation of pyridine nitrogen (N1). According that, the value of angle C2-N1-C3 is 119.8^{0} (Table 3 and Figure 3), actually, different than 125^{0}, which is unusual for protonated pyridine N atom as zwitterion. Note that the same conclusion is comes from understanding IR spectra for non-coordinated ligand and complex, which was discussed before.

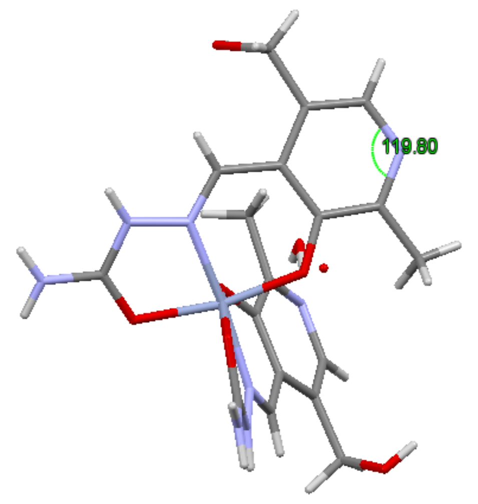


Fig. 3: Value of deprotonated Pyridine Nitrogen N1

In Table 2 are given Bond lengths [Å] of the [Cr (PLSC-H)$_2$].2H_2O complex. The CN of Cr central metal is 6, octahedral structure. Atom ligators are Ophenoic, Nazomethine and Oketo. The most important distances are between Cr and two Oxygen, O2 (phenolic) and O3 (carbonyl). That distances are very similar, 1.910 Å and 1.994 Å, respectively. A little longer is distance Cr-N3, 2.007 Å. Very interesting that completely match the same distance in other coordinated PLSC ligand in Cr bis-ligand compound.

The angles (Table 3) O2-Cr- O3 (168.27°) and N3-Cr-N3 (174.07°), are very close to 180°, but, anyway the complex is deformed octahedral.

Cr-O2	1.910
Cr-O3	1.994
Cr-N3	2.007
Cr-O2(a)	1.910
Cr-O3(a)	1.994
Cr-N3(a)	2.007
O-1C7	(3)1.417
O-2C6	(3)1.319
O-3C9	(3)1.292

N-1C2	(5)1.352
N-1C3	(4)1.329
N-3N4	(3)1.381
N-3C8	(3)1.297

Table 2. Bond lengths [Å] of the [Cr (PLSC-H)$_2$].2H$_2$O complex

Cr-O3-O2	168.27
Cr-N3-O2	89.97
O-2Cr-O2(a)	91.71
O-2Cr-O3(a)	91.06
O-2Cr-N3	94.17
Cr-N3-O3	78.46
Cr-O2-O3	91.06
O-3Cr-O3	88.53
O-3Cr-N3	97.23
N-3Cr-O2	94.17
N-3Cr-O3	97.23
N-3Cr-N3(a)	174.07
O2(a)-Cr-O3(a)	168.27
O-2Cr-N3	89.97
O-3Cr-N3	78.46
C-2N-1C3	119.8
N-3N-4N5	110.16

Table 3. Angles [°] of the [Cr (PLSC-H)$_2$].2H$_2$O complex

4. DISCUSSION :

In this research was synthesized Cr bis-ligand complex compound with two PLSC ligands in coordination with Cr (II). In coordination is monoanionic forms HL-, of the ligand PLSC, is obtained through deprotonation of enol forms. PLSC ligand is tridentate ligand, in coordination trough, phenolic oxygen, pyridine and hydrazine N atom. The physicochemical properties of Cr complex are according to structure given by X-ray crystallography analysis. The metal bis-ligand complex has octahedral geometry, this is a Schiff base produced from semicarbazide and pyridoxal (3-hydroxy-5-hydroxymethyl-2-methylpyridine-4-carboxaldehyde, form of vitamin B6) is reported. Ligand pyridoxal-semicarbazone (PLSC; H2L) is mixing, tridentate ONO ligand. The Cr II environment is a distorted octahedral coordination, the equatorial plane of which is formed by two the tridentate ONO-coordinated pyridoxal-semicarbazone rings. Cr complex crystallizes in monoclinic symmetry, in space group I 2/a.

Based on the results obtained by microanalysis (Elemental Analysis calcd. (%) for complex C18 H26 Cr1 O8 N8, C 33.37, H 5.04, N 19.46; S 8.91; Found: C 33.38, H 5.09, N19.96; S 8.53) , we have an unequivocal confirmation of the composition of the new Cr(II) complex. Oxidation number of Chromium is unexpected, but proven by magnetochemical measurement, μ=4.9BM, which value is related for Cr^{2+}, paramagnetic compound.

According IR spectra of ligand and complex ((ligand= (KBr) n/cm^{-1} Selected IR bands [~m/cm^{-1}]: 3466, 3381, m(OH), m(NH2);3202, m(NH);2850, m(NH+);1697,1678, m(C=O);1583, m(CN);1280, m(CO phenolic);IR (complex) (KBr) n/cm-1 (KBr pellet): 1624.60, 1599, 1501.) is evident, that characteristic pyridine angle of 125^0 exist in ligand but in complex that angle is changed under dehydratation of pyridine hydrogen. Actually, IR vibration of 125^0 pyridine angle is characteristic 2850cm^{-1}, visible for ligand, but for complex not. However, this anomaly can be note as one curiosity of this research, because first time happened that monoanionic form of ligand PLSC was made through dehydrogenation pyridine hydrogen, but not hydrazine hydrogen. Maybe, the reason for reduction of central metal charge, should be in that evidence, because is also normal that oxidation number during complexation should be increasing or stay same.

Anyway, that evidence fact deserves special attention.

The value of the pyridine angle has so far been a good orientin that indicates the form of a coordinated ligand. Earlier results (125^0) of pyridine’s angle measurements of uncoordinated ligand indicate a deprotonated ligand form, zwitterionic form (H2L) (Jevtovic, (2010)).

Coordination ligand PLSC is zwitterions in the neutral form, with deprotonated phenolic OH-group and protonated “pyridine” N atom (Figure 1 (a)). Monoanionic form of the ligands PLSC is obtained through deprotonation of enol-thiol forms (Figure 1 (b)). Finaly, dianionic form of PLSC ligand is obtained through further deprotonation of pyridine N atom (Figure 1 (c)). These postulates have been valid until now, more precisely, to the results obtained in this paper.

As we can see from Figure 4, a comparison was made between the two chromium complexes, previously published K[Cr(PLSC–H) $(NCS)_3$]EtOH (a) (L.S. Vojinovic´-Ješic´ et al. (2015)) and title compound [Cr(PLSC-H)$_2$].2H_2O (b). The complexes are very suitable for comparison because both chromium complexes, were obtained by a very similar synthesis procedure.

The complex K[Cr(PLSC–H) $(NCS)_3$]EtOH is an anionic, mono ligand complex, of octahedral structure, with a mono anionic (HL) form of ligand in coordination. It can be seen very clearly in the Figure 4 (a), that the position of the hydrogen atom is visible in the N 4 (Pyridine atom of nitrogen) position, while there is no hydrogen in the N2 (hydrizine atom of nitrogen) position. This is all in line with the literature on this issue so far.

The value of the Pyridine angle (C5-N4-C7) in the complex K[Cr(PLSC–H) (NCS)3]EtOH is 123.8^o., which corresponds to deprotonated pyridine nitrogen (125^o). In complex K[Cr(PLSC–H) $(NCS)_3$] EtOH, the coordinated HL form was obtained by protonation of the hydrazine N 2 atom Figure 4 (a).

On the other hand, the examined complex [Cr(PLSC-H)$_2$].2H_2O possesses a ligand PLSC coordinated in HL as well, but this form is derived in a different way. The pyridine N 1 atom is protonated here, which is shown in Figure 4. Accordingly, the value of this angle (C2-N1-C3 119.8) is given in Table 4.

Complex	Pyridine Angle	Hydrizine Angle
K[Cr(PLSC–H) $(NCS)_3$]EtOH	C5-N4-C7 123.8(3)	Not availabe
[Cr(PLSC-H)$_2$].2H_2O	Diesel electricity Future cost	DEFC

Table 4. Select angles of complexes K[Cr(PLSC–H) $(NCS)_3$]EtOH and [Cr(PLSC-H)$_2$].2H_2

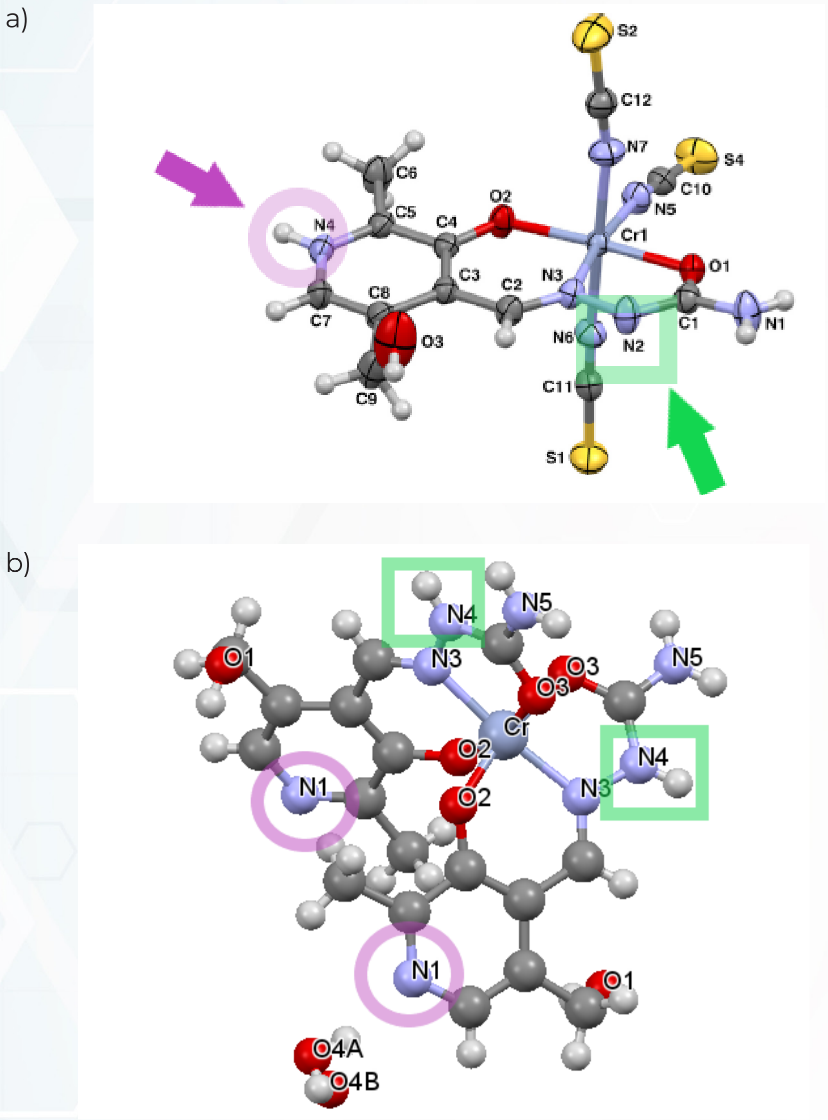


Fig. 4: Crystal structures of K[Cr(PLSC–H) (NCS)$_3$]EtOH complex (a) and title compound [Cr(PLSC-H)$_2$].2H$_2$O

The hydrazine atom N4 in the complex [Cr(PLSC-H)$_2$].2H$_2$O is protonated and the presence of hydrogen is clearly visible in the Figure 4 (b).

What is the cause of this different formation procedure of the HL form of the ligand cannot be clearly established only on the basis of the results of title complex. It is necessary to synthesize new complexes with a similar course of formation of the HL form and only then report the conclusion.

Anyway, that questions can received answer in further investigations according to Cr complexation with PLSC ligand system and other similar with them.

5. CONCLUSION :

This paper reporting the synthesis, crystal structure and physicochemical characteristics of the chromium (II) complex with the PLSC (Pyridoxal-semicarbazone) ligand.

First was synthesized the ligand PLSC according to the previously determined procedure, and then, using chromium oxide Cr_2O_3 in the reaction with the mentioned ligand in water solution, a complex $[Cr(PLSC\text{-}H)_2].2H_2O$ was synthesized. A direct method of synthesis in an air atmosphere was used, with special reference to the adjustment pH of the solution.

White colored monocrystals of the complex were obtained, stable in air, which were characterized by physico-chemical and X-ray analysis was performed. Performed elemental analysis, IR spectra, conductometric and magneto chemically measurements, showed the characteristics of the Cr (II) complex in coordination with the monoanionic form of the PLSC ligand. IR spectra show characteristic peaks for the monoanionic form of the PLSC ligand in coordination, microanalysis confirmed the assumed structure and molecular formula C18 H26 Cr1 O8 N8, magnetic measurements show that it is a chromium (II) complex.

However, final confirmation of the structure was given by X-ray diffraction analysis, and it was confirmed that was synthesized complex with octahedral geometry. The Cr II environment is a distorted octahedral coordination, the equatorial plane of which is formed by two the tridentate ONO-coordinated pyridoxal-semicarbazone rings. Complex [Cr(PLSC-H)2].2H2O crystallizes in monoclinic symmetry, in space group I 2/a, with lattice constants: a = 12.4645(14) Å, b =13.4321(15) Å c =13.7759(14) Å, α =90°, β =102.143(8) °, γ = 90°, V = 2254.8(4) Å3, Z = 4, F(000) = 1112.0, R1=0.0462(1849), wR2 = 0.1586 (2317).

This paper is interesting from the aspect that it was discovered for the first time that the coordination of the monoanionic form does not take place through the previously described method with hydrazine nitrogen, but, in title complex, the coordination of PLSC ligand, leads through monoanionic form, with deprotonation of pyridine nitrogen. This revealed another possibility of ligand PLSC coordination, which can also occur with other ligands from the pyridoxal charbazone ligand family.

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