



## Thermodynamic Stability of the $[M(\text{Pyridine})_4X_2] \cdot 2G$ Clathrates as a Function of the Host Components (M, X) and Included Guest (G)

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### Abstract

This study compares thermodynamic stability of clathrate compounds belonging to three isomorphous series:  $[MPy_4(\text{NCO})_2] \cdot 2Py$  ( $M = M(\text{II}) = \text{Mn, Fe, Co, Ni}$ ),  $[MPy_4(\text{NO}_3)_2] \cdot 2Py$  ( $M = \text{Mn, Co, Ni, Cu, Zn}$ ), and  $[\text{CuPy}_4(\text{NO}_3)_2] \cdot 2G$  ( $G = \text{pyridine, benzene, THF, chloroform}$ ). Thermodynamic parameters ( $\Delta H_{\text{av}}$ ,  $\Delta S_{\text{av}}^0$  and  $\Delta G_{298}^0$ ) of the dissociation of the clathrates were determined from the dependences of the guest equilibrium pressure over the clathrates versus temperature (tensimetric method). Clathrate phases, when different only in the host formula, demonstrated the same order of thermodynamic stability as one expected for the host complexes in solution:  $\text{Mn} < \text{Fe} < \text{Co} < \text{Ni} < \text{Cu} > \text{Zn}$  for M and  $\text{NCO} > \text{NO}_3$  for X. The influence of the host complex formulation was comparable to the effect of guest template, the effect observed in the third series with the variation of the guest component. This study illustrates a dramatic impact of the stability of the host molecule on the overall stability of the clathrate phases, the impact being comparable to a contribution arising from the host–guest complementarity.

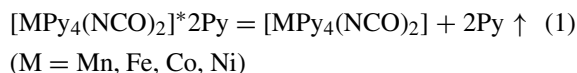
### Introduction

The conventional approach considers the complementarity between host and guest species as the major contributor to the stability of inclusion compounds [1, 2]. This approach is very useful when participating species are stable enough to remain intact upon guest exchange and removal. It is evident however that the overall stability of inclusion compounds is made up not only of intermolecular interactions but also of stability of the species themselves. Metal complexes form a special class of host species which can undergo a dramatic change during the inclusion process [3]. Change is a fundamental element in the design of functional and “smart” materials [3–9]. Metal complexes may undergo reversible conformational changes, isomerization, oligo- and polymerization, as well as other transformations affecting a range of physical and chemical properties of bulk materials [3]. Some complex molecules can exist only in the clathrate matrix while decomposing upon removal of the guest template (“contact stabilization” phenomenon) [10, 11]. This study was undertaken to compare two factors contributing to overall thermodynamic stability of the title clathrates: the intramolecular stability of the host molecule and the efficiency of the intermolecular host–guest interactions.

Clathrates studied in this work were characterized earlier (Table 1). All the compounds have similar stoichiometry

described by a general formula  $[MPy_4X_2] \cdot 2G$ . Most compounds are isostructural and crystallize in orthorhombic system, space group Ccca. The remaining compounds, with higher or lower symmetry of the crystal framework, have the same structural motif and may be considered as isomorphous with the main Ccca type [19, 21, 24]. Structure of one of the studied clathrates is illustrated in Figure 1. The host molecule (Figure 1a) has the nickel(II) cation in an octahedral environment, surrounded by six nitrogen atoms, from four pyridines in the equatorial plane and two isothiocyanato-groups coordinated axially. Guest pyridine species are located in parallel channels stretching along the a-axis direction (Figure 1b), with their dipole moments alternating along the channel. Compounds with nitrate as X ligand have two nitrate groups coordinated axially to the metal cation through the oxygen atom, to give the ‘ $\text{MN}_4\text{O}_2$ ’ coordination polyhedron.

The stoichiometry of the thermal dissociation of the studied compounds was reviewed in [25]. In all these cases the dissociation occurs as a reversible process yielding the same product in a wide range of pressures of the released guest. The compounds of the first series dissociate in one step to produce a guest-free form of the host complex:



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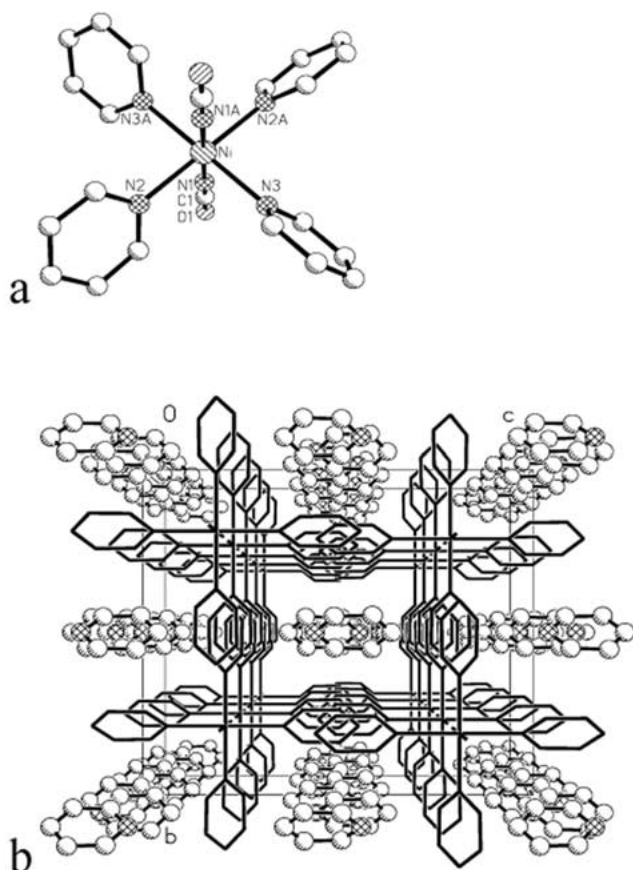
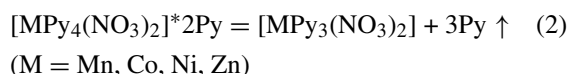
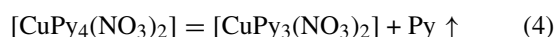


Figure 1. Structure of the host molecule (a) and crystal packing (b) as observed in the  $[\text{NiPy}_4(\text{NCO})_2]_2 \cdot 2\text{Py}$  compound [13].

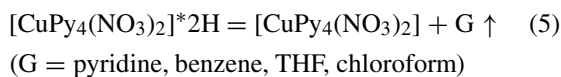
The compounds of the second series dissociate in one step releasing not only all guest pyridine but also one mole of pyridine initially bonded in the host molecule:



The only exception is the copper complex which dissociates to a tripyridine complex in two steps:



Reaction (3) has the same stoichiometry as (1) while the total (3) + (4) is equivalent to (2). All the tripyridine ( $[\text{MPy}_3(\text{NO}_3)_2]$ ) complexes are isostructural solids [14, 26, 27] and, therefore, the contribution of the factor of the crystal structure to the dissociation thermodynamic parameters of the reaction (2) (or the total of (3) + (4)) should be very similar for all the compounds of the second series. The compounds of the third series dissociate similar to (3):



In this case the final product is always the same and the thermodynamic parameters mostly should depend on the nature of releasing guest.

## Experimental

Clathrate compounds studied in this work were prepared as described elsewhere, according to the references listed in Table 1. Pressure of the volatile component over the compounds was measured by the static tensimetric method, using glass membrane spoon-type null-manometers [28–30]. A scheme of the utilized experimental set-up and other details of the technique were described in one of our previous papers [31]. Samples of 0.1 to 3.6 g of the studied compounds were sealed in evacuated reaction vessels equipped with glass membranes, with the curvature of the membranes being sensitive to the pressure inside the vessel. The sensitivity of the membranes varied from 0.03 to 0.1 torr (1 torr = 1 mm Hg = 133.322 Pa). The outer compensating pressure was measured with a mercury manometer. The temperature of the sample was controlled by placing the reactive vessel in a water or oil thermostat. The thermostating and temperature measurement accuracy was 0.05 K.

Typically, two experimental runs were performed for each compound. The runs differed in the ratio of the sample quantity to the reaction vessel volume. The independence of the P-T relation on this ratio served as an evidence of that the guest pressure over the clathrates does not change as the clathrates dissociate or, in other words, the release of the guest from the clathrate matrix results in a collapse of the matrix into a guest-free modification of the host complex. In questionable cases more independent runs were performed, or an additional run was performed after some part of the volatile component was removed from the system (frozen through the gas phase into a special side vessel which was then sealed off).

All studied processes were reversible. The equilibrium pressure established both in the direct and the back course of the dissociation process. In most cases the system reached the equilibrium in hours but in some cases it required several days.

The experimental data on the guest equilibrium pressure over the clathrates versus temperature were approximated by the linear equation  $\log P = A - B/T$  ( $P$ , torr;  $T$ , K), using the least-squares technique in the  $\log P - 1000/T$  coordinates. Average enthalpy change ( $\Delta H_{av}$ ) for a given temperature range, as well as standard entropy ( $\Delta S_{av}^\circ$ ) and free energy ( $\Delta G_{298}^\circ$ ) changes were calculated from these data.

## Results and discussion

Calculated thermodynamic parameters of the dissociation of the studied compounds are listed in Tables 2–4. The P-T dependence for the dissociation of the clathrates of the first series is illustrated in Figure 2. In the  $\log P - 1000/T$  coordinates the experimental data are well approximated by straight lines. However, for several clathrates the dependences experience a bend suggesting phase transformations in the solid samples. They are of two types. The bends observed for  $[\text{MnPy}_4(\text{NCO})_2] \cdot 2\text{Py}$  at 356 K, for  $[\text{MnPy}_4(\text{NO}_3)_2] \cdot 2\text{Py}$  at 353 K, for

Table 1. Composition and structural type of studied compounds

Series	Formula	Ref.	Crystal system, space group	Ref.
[MPy <sub>4</sub> (NCO) <sub>2</sub> ]*2Py	[MnPy <sub>4</sub> (NCO) <sub>2</sub> ]*2Py	[12, 13]	Orthorhombic, Ccca	[12, 13]
	[FePy <sub>4</sub> (NCO) <sub>2</sub> ]*2Py	[12, 13]	Orthorhombic, Ccca	[12, 13]
	[CoPy <sub>4</sub> (NCO) <sub>2</sub> ]*2Py	[12, 13]	Orthorhombic, Ccca	[12, 13]
	[NiPy <sub>4</sub> (NCO) <sub>2</sub> ]*2Py	[12, 13]	Orthorhombic, Ccca	[12, 13]
[MPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[MnPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[14, 15]	Orthorhombic, Ccca	[14]
	[CoPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[15, 16]	Orthorhombic, Ccca	[17]
	[NiPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[15, 16]	Orthorhombic, Ccca	[17, 18]
	[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[15, 19]	Orthorhombic, Pnna <sup>a</sup>	[20, 21]
	[ZnPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[11, 22]	Orthorhombic, Ccca	[11, 17, 23]
[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2G	[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2Py	[15, 19]	Orthorhombic, Pnna <sup>a</sup>	[20, 21]
	[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2C <sub>6</sub> H <sub>6</sub>	[19]	Tetragonal, I422 <sup>b</sup>	[19]
	[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2C <sub>4</sub> H <sub>8</sub> O	[19]	Orthorhombic, Ccca	[24]
	[CuPy <sub>4</sub> (NO <sub>3</sub> ) <sub>2</sub> ]*2CHCl <sub>3</sub>	[19]	Monoclinic, C2/c <sup>b</sup>	[24]

<sup>a</sup> Above 46 °C transforms into orthorhombic (Ccca) type [21].

<sup>b</sup> Isomorphous with the orthorhombic (Ccca) type [19, 24].

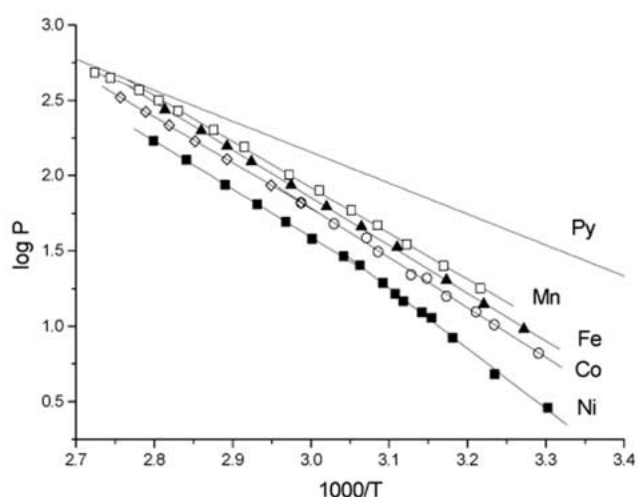


Figure 2. Pyridine vapour pressure over the [MPy<sub>4</sub>(NCO)<sub>2</sub>]\*2Py compounds versus temperature. Experimental data are presented in the log *P* – 1000/*T* coordinates (*P*, torr; *T*, K). M=Mn (clear squares), Fe (triangles), Co (circles), Ni (black squares). The dependence for pyridine vapour pressure over the liquid pyridine is shown for comparison.

[CoPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py at 361 K, and for [ZnPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py at 335 K are expected and correspond to incongruent melting of the clathrates at indicated temperatures [11–16, 22]. The bends for [CoPy<sub>4</sub>(NCO)<sub>2</sub>]\*2Py (~335 K), [NiPy<sub>4</sub>(NCO)<sub>2</sub>]\*2Py (~328 K), [NiPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py (~344 K), and [CuPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py (~333 K) should correspond to polymorphous transformations of the clathrates. As may be seen from Tables 2 and 3, the transitions into high-temperature modifications occur with a significant entropy profit while the resulting change in free energy is not so big. Some rise in symmetry of the clathrate crystal structure may be expected in these cases. Indeed, X-ray structural studies recently performed for [CdPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py [32] and [CuPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py [21] demonstrated the temperature-induced transformation of the primitive unit cell into C-centered, presumably followed by the changes in the dynamics of the nitrate group.

Table 2. Thermodynamic parameters of the process 1/2[MPy<sub>4</sub>(NCO)<sub>2</sub>]\*2Py = 1/2[MPy<sub>4</sub>(NCO)<sub>2</sub>] + Py↑

M	<i>T</i> -range (K)	$\Delta H_{av}$ (kJ/mol)	$\Delta S_{av}^{\circ}$ (J/(mol K))	$\Delta G_{298}^{\circ}$ (kJ/mol)
Mn	311–356	58.4(4)	154(1)	12.4(5)
Fe	305–355	60.7(4)	160(1)	12.9(5)
Co	303–335	63(1)	166(4)	14(1)
	335–363	58.1(3)	151(1)	13.2(3)
Ni	302–328	76(1)	203(6)	16(2)
	328–357	61.0(6)	155(2)	14.8(8)

Table 3. Thermodynamic parameters of the process 1/3[MPy<sub>4</sub>(NO<sub>3</sub>)<sub>2</sub>]\*2Py = 1/3[MPy<sub>3</sub>(NO<sub>3</sub>)<sub>2</sub>] + Py↑

M	<i>T</i> -range (K)	$\Delta H_{av}$ (kJ/mol)	$\Delta S_{av}^{\circ}$ (J/(mol K))	$\Delta G_{298}^{\circ}$ (kJ/mol)
Mn	292–353	53.8(3)	142(1)	11.4(5)
Co	293–360	59.5(2)	157(1)	12.8(2)
Ni	293–347	64.6(4)	168(1)	14.4(6)
	347–374	57.2(3)	147(1)	13.4(1)
Cu <sup>a</sup>	304–333	73(1)	185(3)	17.9(6)
	333–378	61.7(3)	150(1)	16.7(6)
Zn	300–335	58.3(7)	158(2)	11.1(9)

<sup>a</sup>This compound dissociates in two steps (Equations (3) and (4)). Here values for total (3) + (4) per one mole of releasing pyridine are given for the purpose of comparison. The thermodynamic parameters for the step (3) are given in Table 4. For step (4)  $\Delta H_{av} = 70.1(7)$  kJ/mol,  $\Delta S_{av}^{\circ} = 162(2)$  J/(mol K), and  $\Delta G_{298}^{\circ} = 21.9(9)$  kJ/mol.

Experimental data for the [MPy<sub>4</sub>(NCO)<sub>2</sub>]\*2Py series (Figure 2) indicate that vapour pressure of the pyridine guest essentially differs from compound to compound, in spite of structural similarity of the host matrices in the clathrates. For example, at room temperature (298 K) the equilibrium pyridine pressure over the clathrates is 5.1, 4.1, 3.0 and 1.3 torr for M=Mn, Fe, Co and Ni, respectively (20.5 torr over li-

Table 5. Comparison of  $\Delta G_{298}^{\circ}$  of dissociation of the  $[\text{CuPy}_4(\text{NO}_3)_2] \cdot 2\text{G}$  clathrates (see Table 4) and selected factors which can contribute to the observed stability changes

$G$	$\Delta G_{298}^{\circ}$ (kJ/mol)	B.p. of neat $G$ (K)	Polarity of $G$ (D)	Cu—O <sub>nitrate</sub> (Å)	Crystal system, lattice type
Pyridine	15.9(5)	413	2.22	2.42	Orthorhombic, P
Benzene	7.7(5)	378	0	2.47	Tetragonal, I
THF	7(2)	364	1.75	2.46	Orthorhombic, C
Chloroform	4.8(3)	359	1.04	2.57	Monoclinic, C

Table 4. Thermodynamic parameters of the process  $1/2[\text{CuPy}_4(\text{NO}_3)_2] \cdot 2\text{G} = 1/2[\text{CuPy}_4(\text{NO}_3)_2] + \text{G}\uparrow$

$G$	$T$ -range (K)	$\Delta H_{\text{av}}$ (kJ/mol)	$\Delta S_{\text{av}}^{\circ}$ (J/(mol K))	$\Delta G_{298}^{\circ}$ (kJ/mol)
Pyridine	304–333	74.6(9)	197(3)	15.9(5)
	333–378	57.2(3)	145(9)	14.1(4)
Benzene	290–346	45.3(3)	126(1)	7.7(5)
THF	290–320	51(1)	146(6)	7(2)
Chloroform	300–335	42.9(2)	128(1)	4.8(3)

quid pyridine). The difference shows over the whole studied temperature range. Calculated thermodynamic parameters for dissociation of the clathrates of the  $[\text{MPy}_4(\text{NCO})_2] \cdot 2\text{Py}$  series are listed in Table 2. Thermodynamic stability of the compounds changes in the following sequence:  $\text{Mn} < \text{Fe} < \text{Co} < \text{Ni}$ .

One of possible explanations of the difference in the clathrate stability is a distortion of coordination polyhedra of the host complexes upon collapse of the high-symmetry clathrate phases into guest-free modifications, as was observed for  $[\text{NiPy}_4(\text{NCO})_2] \cdot 2\text{Py}$  [13]. Weakening of coordination bonds caused by such a phase change is expected to be more significant for more stable complexes and *vice versa*.

Similar sequence was observed in the  $[\text{MPy}_4(\text{NO}_3)_2] \cdot 2\text{Py}$  series (Table 3):  $\text{Mn} < \text{Co} < \text{Ni} < \text{Cu} > \text{Zn}$ . However, in this case the host complex is directly involved in the process releasing additional pyridine and transforming to a tripyridine complex while a guest-free host complex does not appear as a thermodynamically stable product.

The order of stability of the clathrate phases in two above series follows the general stability sequence for complexes in solution (Irwing–Williams sequence [33]):  $\text{Mn} < \text{Fe} < \text{Co} < \text{Ni} < \text{Cu} > \text{Zn}$ . This observation demonstrates how the strengthening of the coordination bonds of the host complex results in a progressive rise in the clathrate phase stability. Therefore, the observed difference in thermodynamic stability of the clathrates of two above series is determined by difference in stability of the respective host complex molecules.

Experimental data for the  $[\text{CuPy}_4(\text{NO}_3)_2] \cdot 2\text{G}$  series (Table 4) make it possible to estimate contribution of the guest to stability of the clathrate phases. The observed stability sequence is pyridine  $<$  benzene  $\sim$  THF  $<$  chloroform. The contribution of the guest is a balance of several factors;

the most important are volatility of the guest, polarity of the guest molecule (dipole moment), distortion of the host complex caused by the guest (in the series studied the most sensitive parameter is the length of the  $\text{M—O}_{\text{nitrate}}$  coordination bond [24]), and distortion of the host framework (which may be judged from deviations of the crystal lattice symmetry upon inclusion of different guests). Apparently, there is a correlation between boiling points of the neat guest liquids and the observed stability of the clathrate phases. The other three parameters show different degree of host–guest complementarity in the studied series. Although the total impact of these factors on thermodynamics of the dissociation appears to be significant, the problem of how the guest molecule geometry and charge distribution on the molecule reflect on the observed differences could hardly be solved quantitatively in this study. An important conclusion resulting from the reported results is that the whole range of variations in  $\Delta G_{298}^{\circ}$  observed upon isostructural replacement of the host metal complexes ( $[\text{MPy}_4(\text{NCO})_2] \cdot 2\text{Py}$  and  $[\text{MPy}_4(\text{NO}_3)_2] \cdot 2\text{Py}$  series) and upon replacement for the guest component ( $[\text{CuPy}_4(\text{NO}_3)_2] \cdot 2\text{G}$  series) are comparable. In other words, this study illustrates a dramatic impact of the stability of the host complex on the overall stability of the clathrate phases it forms, the impact being comparable to a contribution arising from the host–guest complementarity.

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