Table 12:
Pascalôs constants for calculating diamagnetism.

Ion / molecule	*Pascal's constant x 10^{-6} (cm^3/mol)*
[a] Ni^{2+}	-12.0
[a] Cu^{2+}	-11.0
[b] C	-6.00
[b] C (in benzene ring)	0.24
[b] N (ring)	-4.61
[b] N (imide)	-2.11
[b] C = O	-1.73
[b] C = N	8.20
[b] C = C	5.50
[b] H	-2.93
[b] Cl	-20.1
[b] Br	-30.6

[a] *(Selwood, 1956)*
[b] *(Carlin, 1986)*

Table 13:
Crystallographic data for L1.

Empirical formula	$C_{19}H_{16}N_4O$
Formula weight	316.36
T (K)	173 (2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Monoclinic, *Cc*
Crystal size (mm)	0.60 x 0.15 x 0.10
a (Ù)	14.0350(2)
b (Ù)	14.4517(2)
c (Ù)	8.1372(1)
Ŭ(º)	90.00
ɓ (º)	102.193(1)
ɔ (º)	90.00
V ($Ù^3$)	1613.23(4)
Z	4
D_x (mgm^{-3})	1.303
ε (mm^{-1})	0.08
F (000)	664
ɗ range for data collection (º)	2.8 to 30.2
Limiting indices	-19 Ò*h* Ò19, -20 Ò*k* Ò20, -11 Ò*l* Ò11
Reflection collected / unique	0.023
Data / restraints / parameters	2340 / 17 / 281
Completeness to ɗ =	-
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	-
Final R indices [I > 2ů(I)]	R_1 = 0.034, wR_2 = 0.100
R indices (all data)	-
Largest difference peak and hole (e $Ù^{-3}$)	0.38 and -0.26

Table 14:
Selected bond lengths (Ù) and angles (º) for L1.

Bond lengths (Å)			
O1 - C10	1.2414(19)	C8 - C9	1.436(2)
N2 - C9	1.2952(18)	C10 - C11	1.5220(19)
N2 - N3	1.3985(16)	C11 - C13	1.501(2)
N3 - C10	1.3405(18)	C5 - H5	0.998(10)
Bond angles (º)			
O1 - C10 - N3	123.58(13)	N1 - C7 - C8	110.21(14)
O1 - C10 - C11	122.08(13)	C13 - C12 - N4	109.95(14)
N3 - C10 - C11	114.31(13)	N2 - C9 - C8	122.73(13)
C13 - C11 - C10	111.28(12)		

Hydrogen-bond (Å)				
D - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N1 - H1N------O1	0.88(1)	1.98(1)	2.848(2)	174(3)
N3 - H3N------O1	0.87(1)	1.97(1)	2.808(2)	163(2)

Table 15:
Crystallographic data for L2.

Empirical formula	$C_{19}H_{15}ClN_4O)_2$
Formula weight	350.80
T (K)	173 (2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Monoclinic, *Cc*
Crystal size (mm)	0.47 x 0.32 x 0.30
a (Ù)	13.7442 (2)
b (Ù)	15.0424 (2)
c (Ù)	8.3120 (1)
Ŭ(º)	90.00
ɓ (º)	103.095 (1)
ɔ (º)	90.00
V ($Ù^3$)	1673.78 (4)
Z	4
D_x (mgm^{-3})	1.392
ε (mm^{-1})	0.24
F (000)	728
ɗ range for data collection (º)	2.7 to 33.9
Limiting indices	-17 Ò*h* Ò17, -19 Ò*k* Ò19, -10 Ò*l* Ò10
Reflection collected / unique	0.108
Data / restraints / parameters	4683 / 2 / 278
Completeness to ɗ =	-
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	1.15
Final R indices [I > 2ů(I)]	R_1 = 0.026, wR_2 = 0.070
R indices (all data)	R_1 = 0.027, wR_2 = 0.084
Largest difference peak and hole (e $Ù^{-3}$)	0.24 and -0.22

Table 16:
Selected bond lengths (Ù) and angles (º) for L2.

Bond lengths (Å)							
O1 ï C10	1.242(3)	N3 ï N2	1.395(2)	C11 ï C10	1.522(3)	C9 ï C7	1.441(3)
N3 ï C10	1.337(2)	N2 ï C9	1.295(2)	C11 ï C12	1.504(3)	C4 ï C5	1.385(3)
Bond angles (º)							
O1 ï C10 ï N3	124.39(18)	N3 ï N2 ï C9	112.24(17)	C10 - N3 - N2	121.27(17)	C12 - C11 - C10	111.04(16)
O1 - C10 - C11	121.30(17)	N3 - C10 - C11	114.27(17)	N2 ï C9 ï C7	123.41(18)	C11 - C12 - C13	127.24(19)

Hydrogen bond *D* - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N1 - H1N------O1	0.88(10)	2.02(10)	2.893(2)	174(3)
N3 ï H3N------O1	0.88(10)	1.96(10)	2.827(2)	170(4)

Table 17:
Crystallographic data for L3.

Empirical formula	$C_{19}H_{15}BrN_4O.C_4H_8O_2$
Formula weight	483.36
T (K)	173 (2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Monoclinic, $P2_1/c$
Crystal size (mm)	0.45 x 0.26 x 0.05
a (Ù)	11.9285(2)
b (Ù)	12.8010(2)
c (Ù)	15.5177(3)
Ŭ(º)	90.00
ɓ (º)	110.665(1)
ɔ (º)	90.00
V ($Ù^3$)	2217.05(7)
Z	4
D_x (mgm^{-3})	1.448
ε (mm^{-1})	1.89
F (000)	992
ɗ range for data collection (º)	2.8 to 28.3
Limiting indices	-15 Ò*h* Ò15, -16 Ò*k* Ò16, -20 Ò*l* Ò20
Reflection collected / unique	0.044
Data / restraints / parameters	5100 / 3 / 294
Completeness to ɗ =	-
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	-
Final R indices [I > 2ů(I)]	R_1 = 0.031, wR_2 = 0.097
R indices (all data)	-
Largest difference peak and hole (e $Ù^{-3}$)	0.40 and -0.35

Table 18:
Selected bond lengths (Ù) and angles (º) for L3.

Bond lengths (Å)			
O1 - C10	1.241(2)	N2 - N3	1.392(2)
C9 - C10	1.516(3)	N3 - C11	1.283(3)
C11 - C12	1.438(3)	C7 - C9	1.498(3)
N2 - C10	1.333(3)	Br1 - C16	1.903(2)
Bond angles (º)			
C10 - N2 - N3	120.51(16)	O1 - C10 - C9	121.24(18)
C11 - N3 - N2	113.85(17)	N2 - C10 - C9	114.78(17)
C7 - C9 - C10	111.52(16)	N3 - C11 - C12	122.86(19)
O1 - C10 - N2	123.97(18)		

Hydrogen bond (Å)

D - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N1 - H1N------O1	0.88(1)	2.02(1)	2.841(2)	156(3)
N2 - H2N------O2	0.88(1)	2.02(1)	2.893(2)	171(2)
N4 - H4N------O1	0.88(1)	2.00(1)	2.881(2)	177(3)

Table 19:
Crystallographic data for NiL1.

Empirical formula	$[Ni(C_{19}H_{15}N_4O)_2]$
Formula weight	691.43
T (K)	100(2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Triclinic, P1bar
Crystal size (mm)	0.36 x 0.15 x 0.15
a (Ù)	4.961(5)
b (Ù)	9.101(10)
c (Ù)	17.052(18)
Ŭ(º)	76.921(14)
ɓ (º)	87.841(14)
ɔ (º)	86.096(17)
V ($Ù^3$)	748.0(13)
Z	1
D_x (mgm^{-3})	1.535
ε (mm^{-1})	0.702
F (000)	360
ɗ range for data collection (º)	1.23 to 30.03
Limiting indices	-2 Ò*h* Ò6, -9 Ò*k* Ò10, -22 Ò*l* Ò23
Reflection collected / unique	0.0198
Data / restraints / parameters	1825 / 150 / 223
Completeness to ɗ = 25.00	50.6 %
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	2.250
Final R indices [I > 2ů(I)]	R_1 = 0.2594, wR_2 = 0.5737
R indices (all data)	R_1 = 0.2924, wR_2 = 0.5891
Largest difference peak and hole (e $Ù^{-3}$)	1.539 and -1.880

Table 20:
Selected bond lengths (Ù) and angles (º) for NiL1.

Bond lengths (Å)							
Ni - O1	1.65(2)	Ni - N2ô	1.85(2)	O1 - C10	1.37(5)	N2 - C9	1.38(3)
Ni - N2	1.85(2)	N2 - N3	1.24(5)	C2 - C9	1.61(6)	C11 - C12	1.55(3)
Ni - O1ô	1.65(2)	N3 - C10	1.37(4)	C10 - C11	1.59(6)	N1 ï C1	1.52(4)
Bond angles (º)							
O1 - Ni - O1ô	180.000(4)	O1ô - Ni - N2	92.5(8)	C10 - N3 - N2	111(4)	C10 - O1 - Ni	110.9(16)
O1 - Ni - N2ô	92.5(8)	N2 - Ni - N2ô	180.000(3)	N3 - C10 - O1	117(3)	C9 - N2 - N3	123(3)
O1ô - Ni - N2	87.5(8)	C9 - N2 - Ni	124(3)	N3 - C10 - C11	115(4)	C12 - C11 - C10	107(3)
O1 - Ni ï N2	87.5(8)	N3 - N2 - Ni	113.4(18)	O1 - C10 - C11	127(2)	N2 - C9 ï C2	123(4)

Hydrogen bond *D* - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N4 - H4N----N3	0.88	2.51	3.16(4)	131.0

Table 21:
Crystallographic data for NiL2.

Empirical formula	$[Ni(C_{19}H_{15}ClN_4O)_2].2C_2H_6OS$
Formula weight	914.55
T (K)	139 (2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Monoclinic, $P2_1/c$
Crystal size (mm)	0.42 x 0.05 x 0.02
a (Ù)	11.659(1)
b (Ù)	10.219(1)
c (Ù)	17.585(2)
Ŭ(º)	-
ɓ (º)	102.272(3)
ɔ (º)	
V ($Ù^3$)	2047.2(3)
Z	2
D_x (mgm^{-3})	1.484
ε (mm^{-1})	0.76
F (000)	948
ɗ range for data collection (º)	4.4 to 22.5
Limiting indices	-15 Ò*h* Ò14, -13 Ò*k* Ò10, -22 Ò*l* Ò22
Reflection collected / unique	0.108
Data / restraints / parameters	4683 / 2 / 278
Completeness to ɗ =	-
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	-
Final R indices [I > 2ů(I)]	R_1 = 0.049, wR_2 = 0.198
R indices (all data)	-
Largest difference peak and hole (e $Ù^{-3}$)	0.50 and -0.89

Table 22:
Selected bond lengths (Ù) and angles (º) for NiL2.

Bond lengths (Å)							
Ni - O1	1.845(4)	Ni - N2ô	1.861(4)	O1 - C10	1.301(6)	N2 - C9	1.300(6)
Ni - N2	1.861(4)	N2 - N3	1.413(6)	C8 - C9	1.425(7)	C11 - C12	1.499(8)
Ni - O1ô	1.845(4)	N3 - C10	1.297(6)	C10 - C11	1.510(7)	Cl - C3	1.745(6)
Bond angles (º)							
O1 - Ni - O1ô	180.000(1)	O1ô - Ni - N2ô	96.52(17)	C10 - N3 - N2	107.2(4)	C10 - O1 - Ni	110.4(3)
O1 - Ni - N2ô	96.52(17)	N2 - Ni - N2ô	180.000(1)	N3 - C10 - O1	124.4(5)	C9 - N2 - N3	117.9(4)
O1ô - Ni - N2	83.48(17)	C9 - N2 - Ni	127.5(4)	N3 - C10 - C11	118.0(5)	C12 - C11 - C10	112.9(5)
O1 - Ni - N2	83.48(17)	N3 - N2 - Ni	114.2(3)	O1 - C10 - C11	117.6(5)	N2 - C9 - C8	127.8(5)

Hydrogen bond *D* - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N1 - H1N------O2	0.88(4)	1.90(5)	2.766(6)	167(6)
N4 - H4N------O2	0.88(3)	2.05(3)	2.887(6)	158(6)

Table 23:
Crystallographic data for NiL3.

Empirical formula	$[Ni(C_{19}H_{14}BrN_4O)_2]$
Formula weight	849.23
T (K)	100(2)
Radiation ə (Ù)	Mo KŬ(0.71073)
Crystal system, space group	Monoclinic, P2(1)/c
Crystal size (mm)	0.20 x 0.10 x 0.10
a (Ù)	10.040(3)
b (Ù)	8.749(3)
c (Ù)	18.619(7)
Ŭ(º)	90
ɓ (º)	98.95(3
ɔ (º)	90
V ($Ù^3$)	1615.7(9)
Z	2
D_x (mgm^{-3})	1.746
ε (mm^{-1})	3.125
F (000)	856
ɗ range for data collection (º)	2.05 to 30.92
Limiting indices	-13 Ò*h* Ò14, -12 Ò*k* Ò12, -26 Ò*l* Ò21
Reflection collected / unique	0.4871
Data / restraints / parameters	4729 / 0 / 232
Completeness to ɗ = 25.00	100.0 %
Refinement method	Full-matrix least-squares on F^2
Goodness of fit F^2	0.703
Final R indices [I > 2ů(I)]	R_1 = 0.085, wR_2 = 0.166
R indices (all data)	R_1 = 0.335, wR_2 = 0.229
Largest difference peak and hole (e $Ù^{-3}$)	1.22 and -0.80

Table 24:
Selected bond lengths (Ù) and angles (º) for NiL3.

Bond lengths (Å)							
Ni - O1	1.825(6)	Ni ï N3ô	1.884(8)	O1 - C10	1.285(12)	N1 ï C10	1.305(13)
Ni ï N3	1.884(8)	N1 - N3	1.377(11)	C9 ï C10	1.473(13)	C11 - C12	1.458(14)
Ni - O1ô	1.825(6)	N3 - C11	1.310(12)	C10 - C11	1.510(7)	Br ï C17	1.843(12)
Bond angles (º)							
O1 - Ni - O1ô	180.0(4)	O1ô- Ni - N2	96.52(17)	C11 - N3 ï N1ô	117.3(8)	C10 - O1 - Ni	112.3(6)
O1 - Ni ï N3ô	83.0(4)	N3 - Ni ï N3ô	180.0(6)	N1 - C10 - O1	122.0(10)	C9 ï N1 - N3ô	109.7(9)
O1ô- Ni ï N3ô	97.0(3)	C11 ï N3 - Ni	129.7(7)	N3 - C11 - C12	128.1(10)		
O1 - Ni ï N3	97.0(3)	N1ôï N3 - Ni	112.9(7)	O1 - C10 ï C9	119.8(9)		

Hydrogen bond *D* - H----*A*	*D* - H	H----*A*	*D*----*A*	*D* - H----*A*
N5- H5N------N1	0.88	2.54	3.193(15)	131.7

Figure 27:
UV-Vis absorption bands of chloroindolecarboxaldehyde ï indolehydrazide (*L2*)

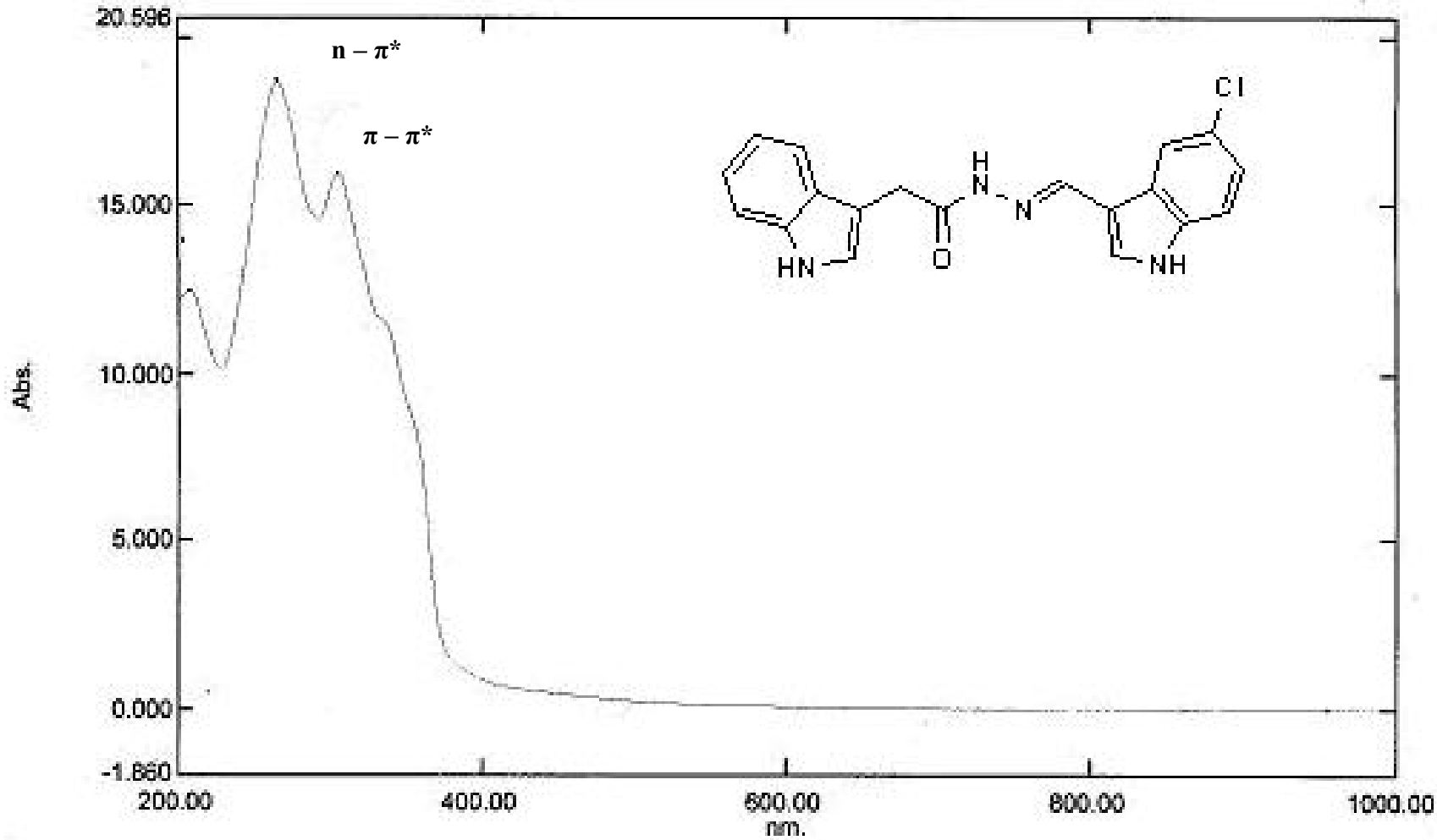

Figure 28:
UV-Vis absorption bands of Cu(chloroindolecarboxaldehyde ï indolehydrazide)$_2$ (*CuL2*)

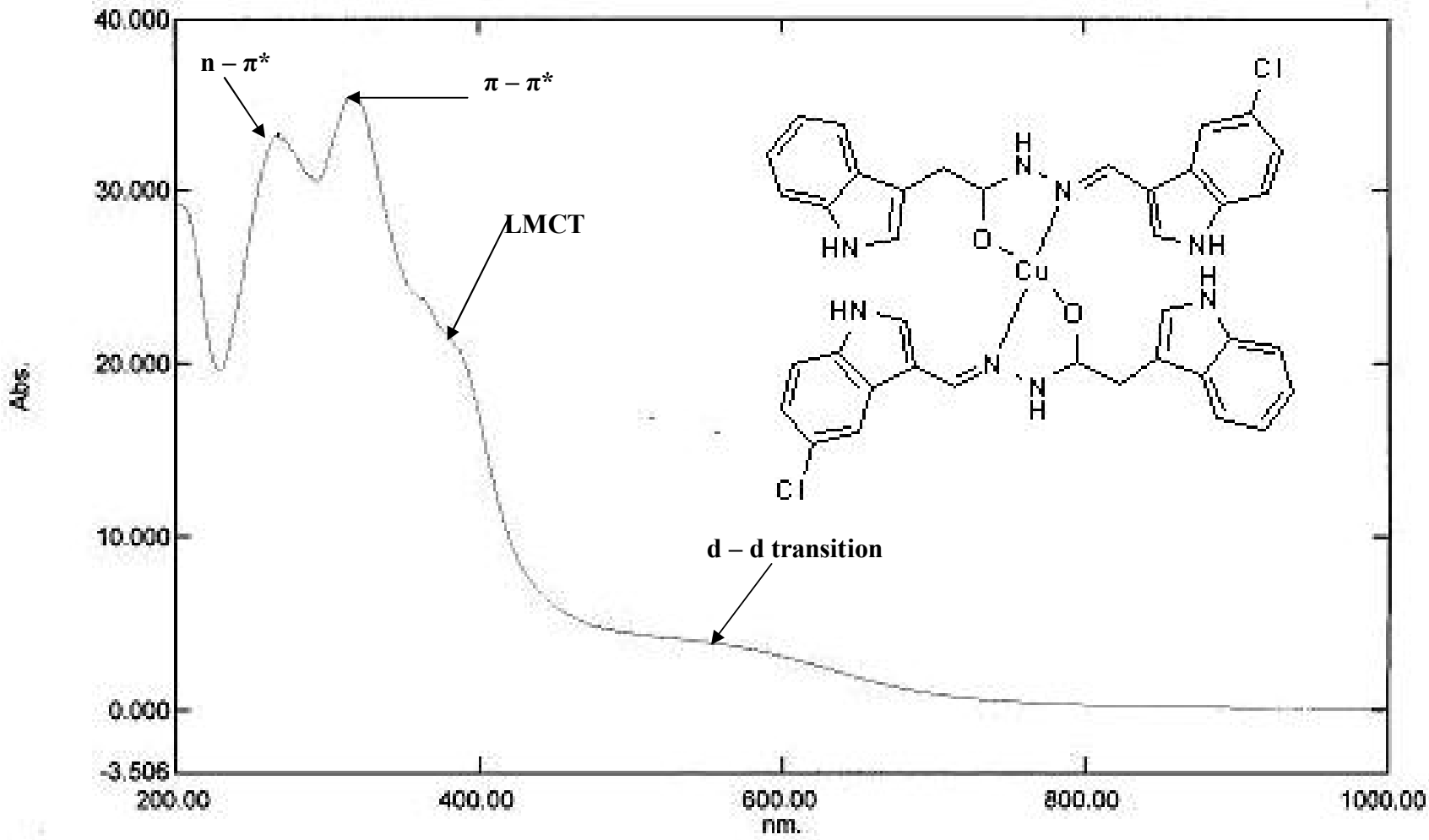

Figure 29:
UV-Vis absorption bands of Ni(chloroindolecarboxaldehyde ï indolehydrazide)$_2$ (*NiL2*)

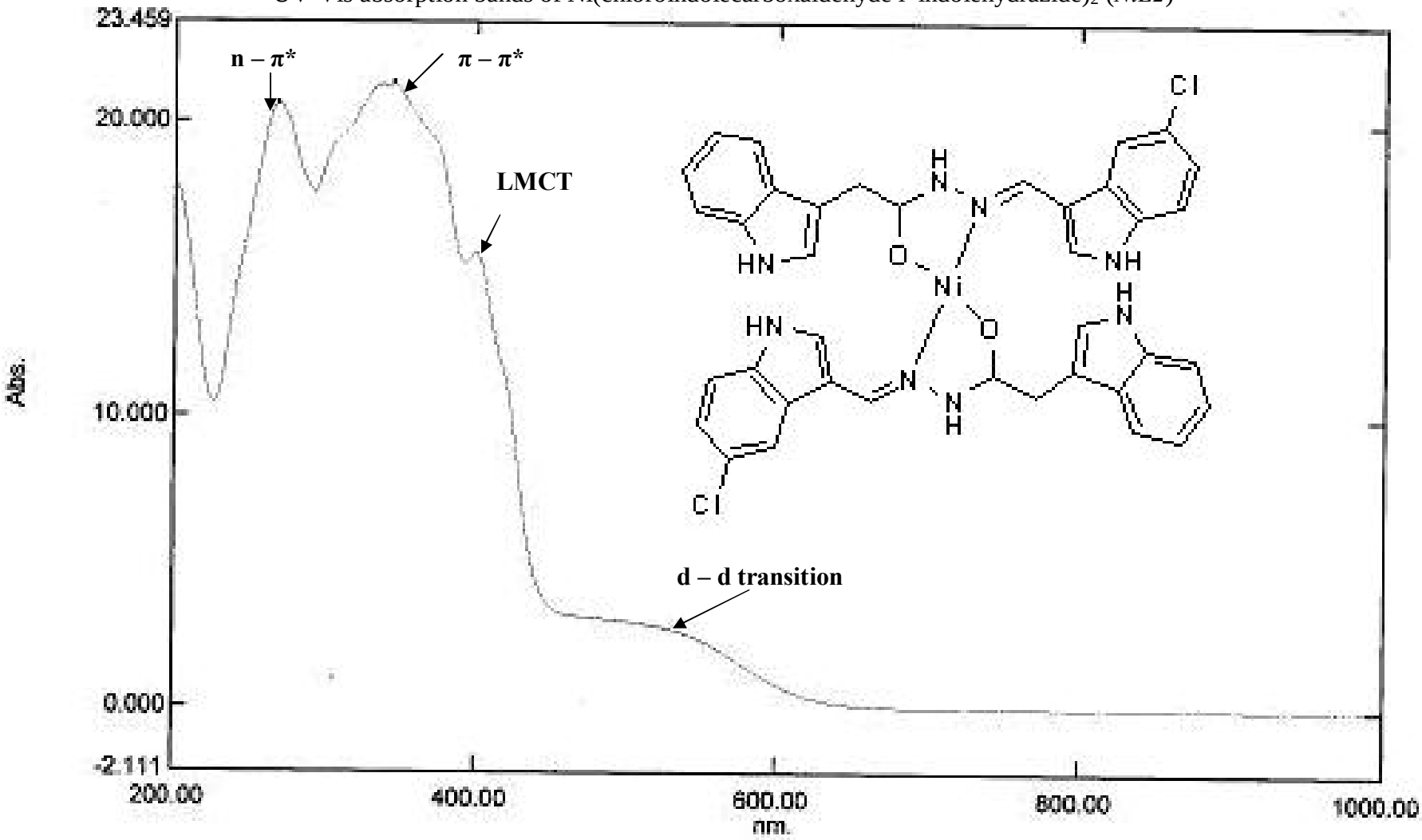

Figure 30:
UV-Vis absorption bands of bromoindolecarboxaldehyde ï indolehydrazide (*L3*)

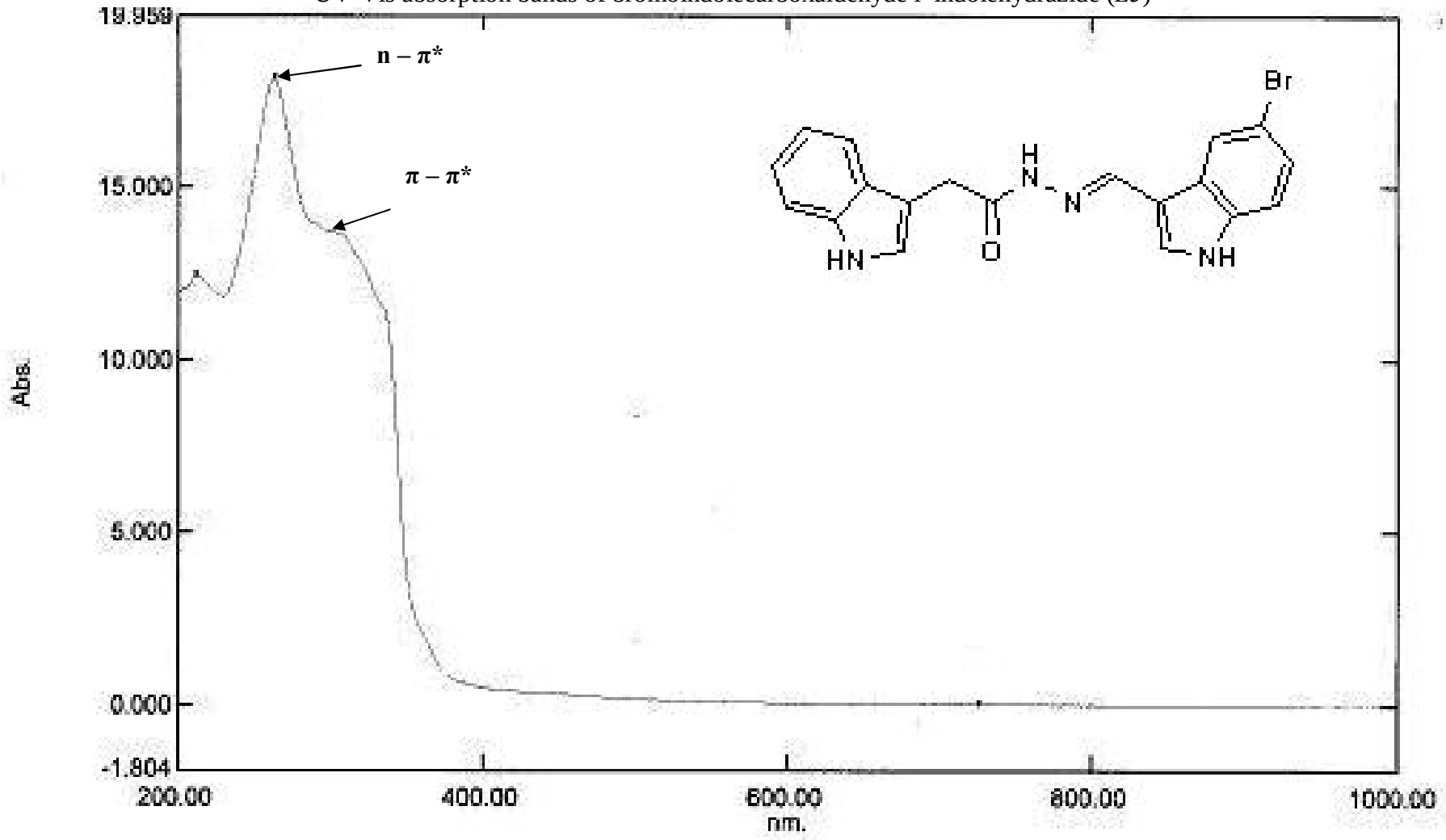

Figure 31:
UV-Vis absorption bands of Cu(bromoindolecarboxaldehyde ï indolehydrazide)$_2$ (*CuL3*)

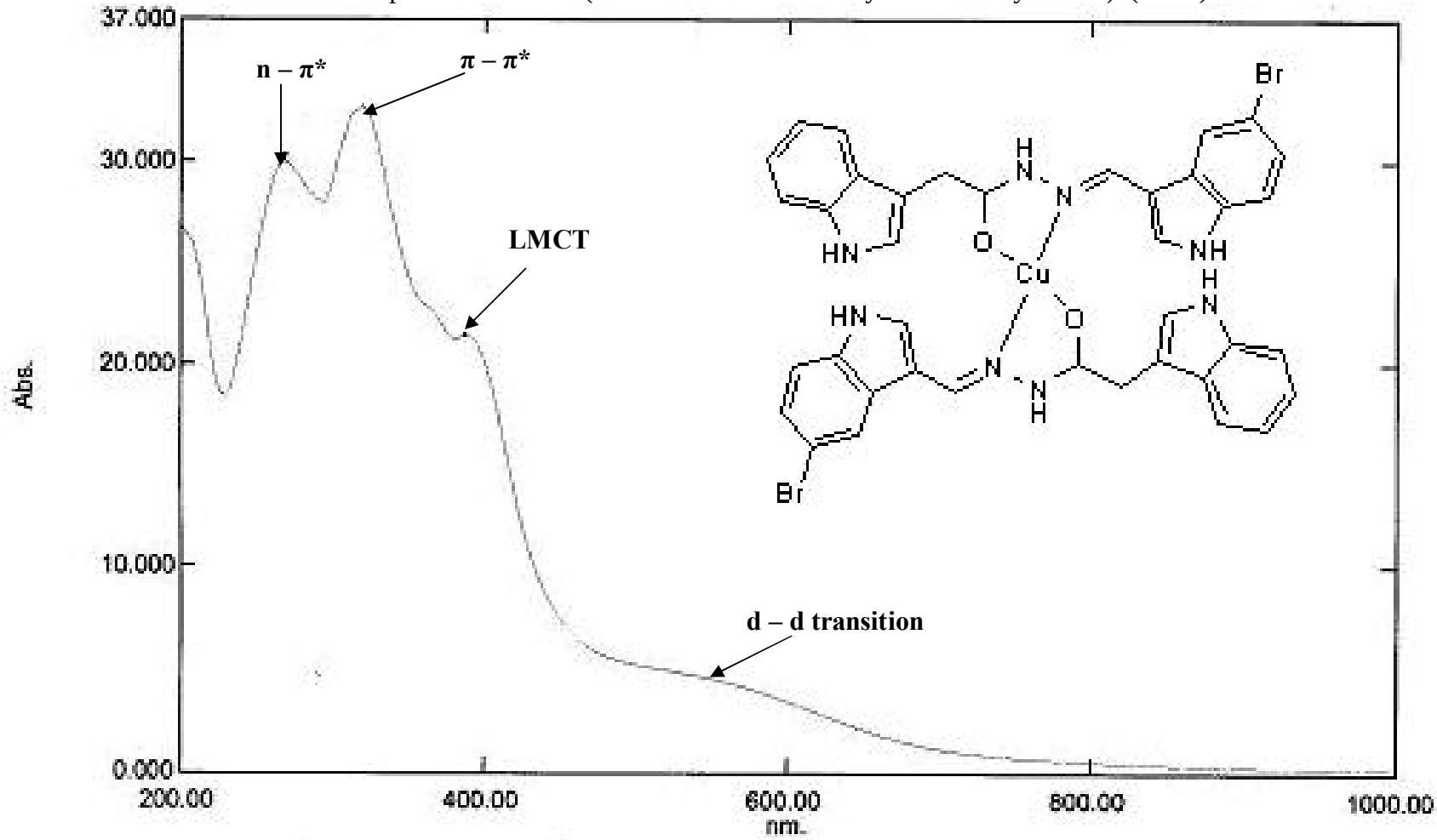

Figure 32:
UV-Vis absorption bands of Ni(bromoindolecarboxaldehyde ï indolehydrazide)$_2$ (*NiL3*)

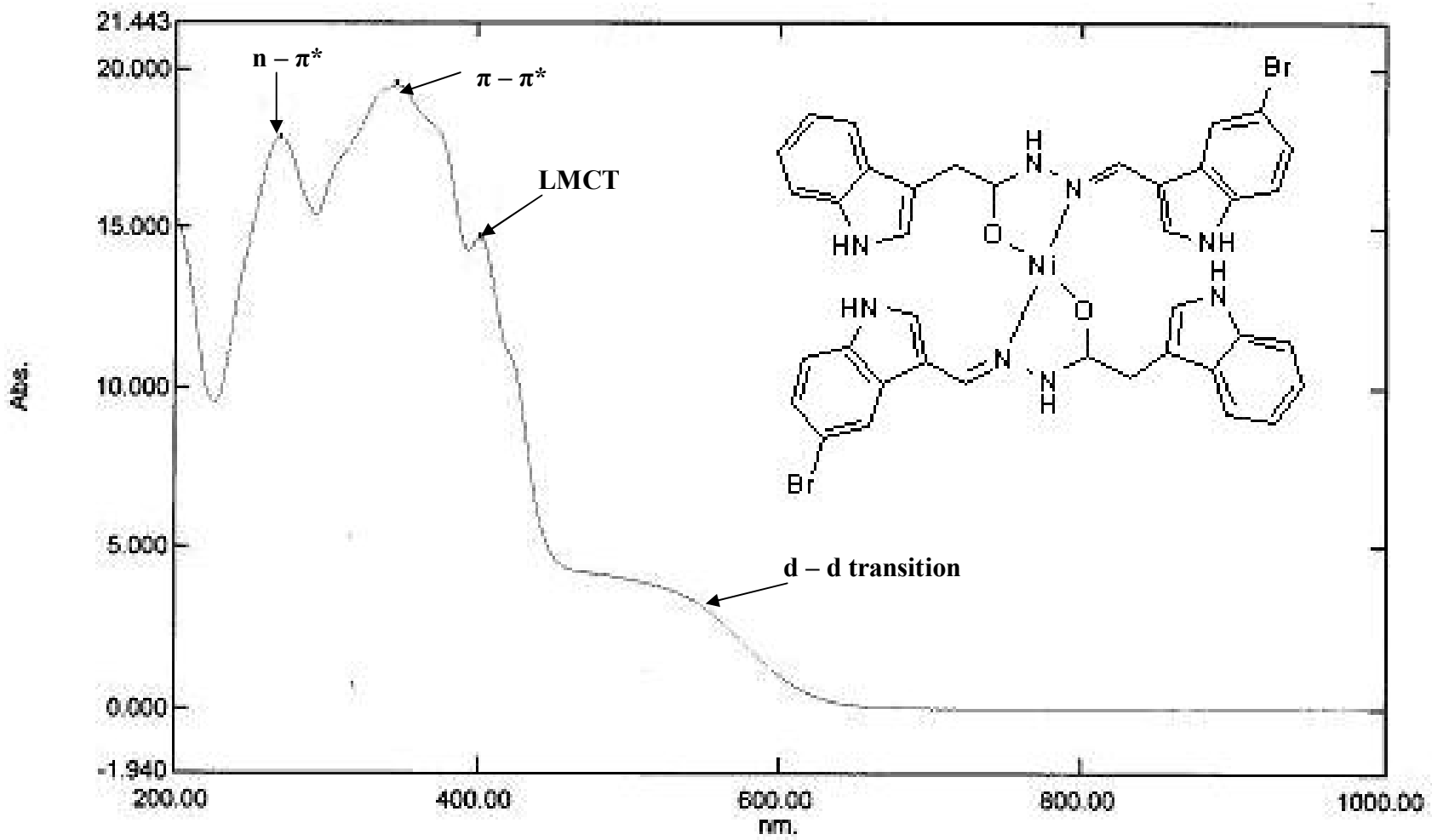

Figure 33:
IR spectrum of indolecarboxaldehyde - indolehydrazide (*L1*)

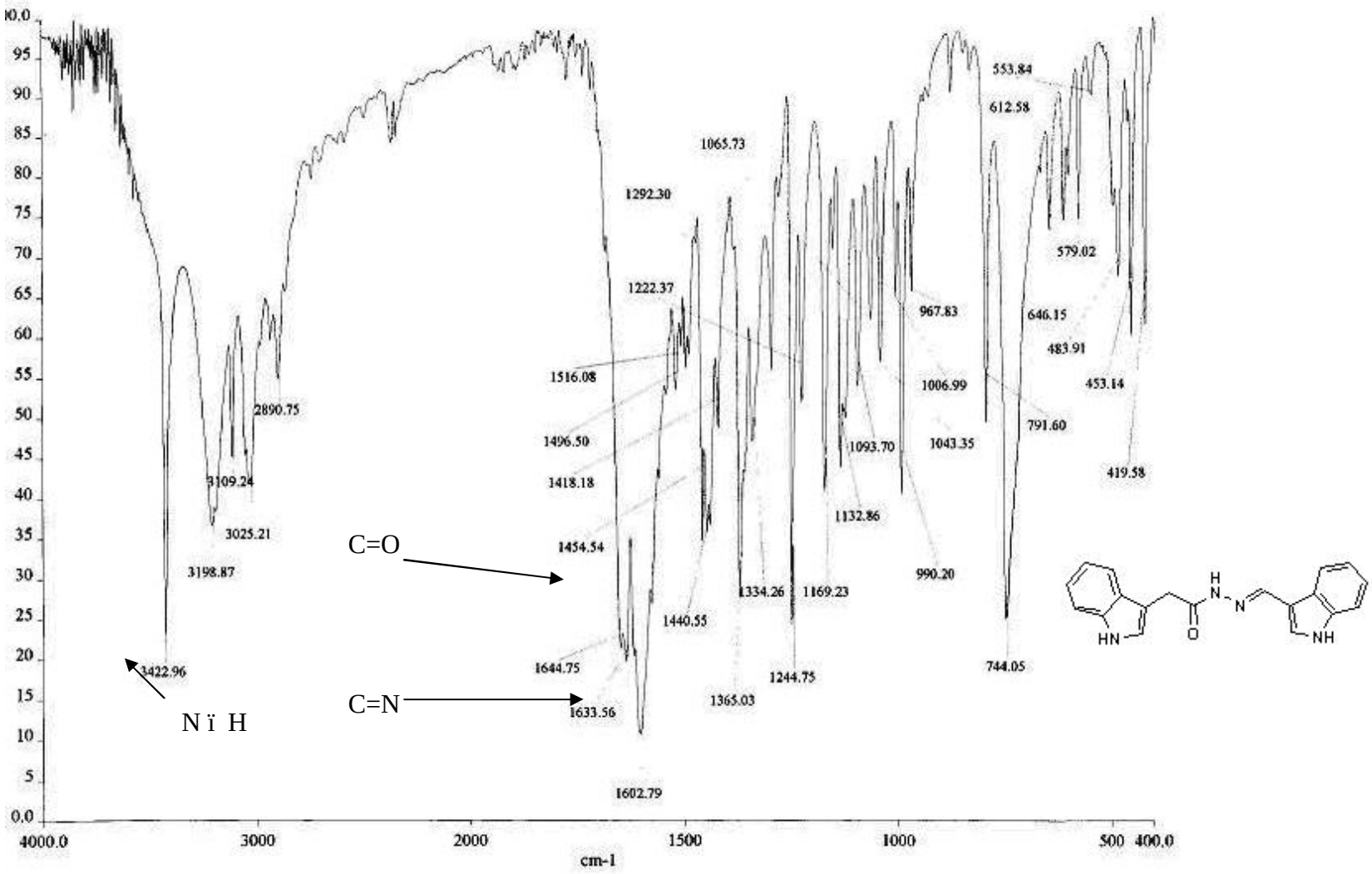

Figure 34:
IR spectrum of Cu(indolecarboxaldehyde ï indolehydrazide)$_2$ (*CuL1*)

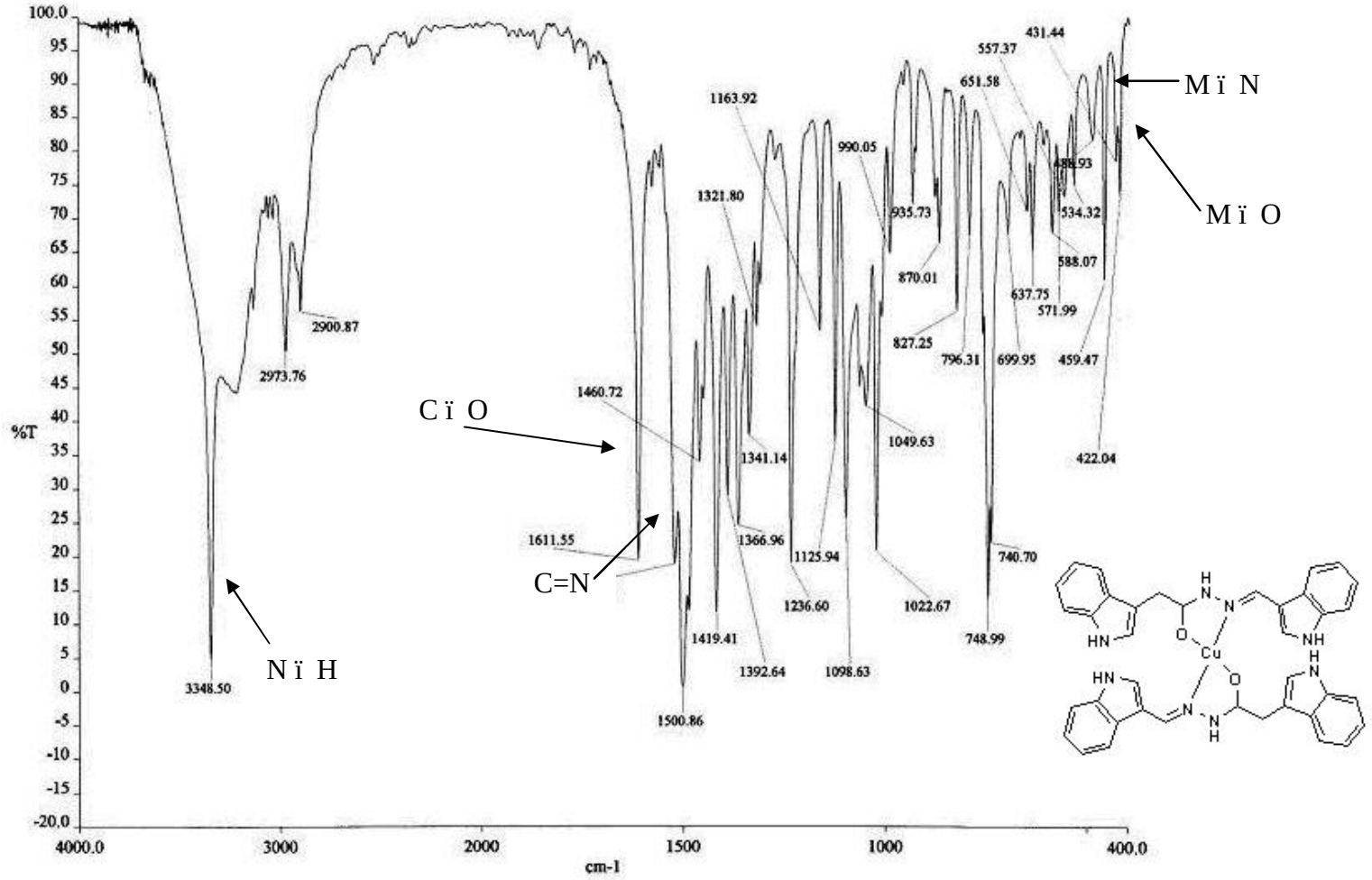

Figure 35:
IR spectrum of Ni(indolecarboxaldehyde ï indolehydrazide)$_2$ (*NiL1*)

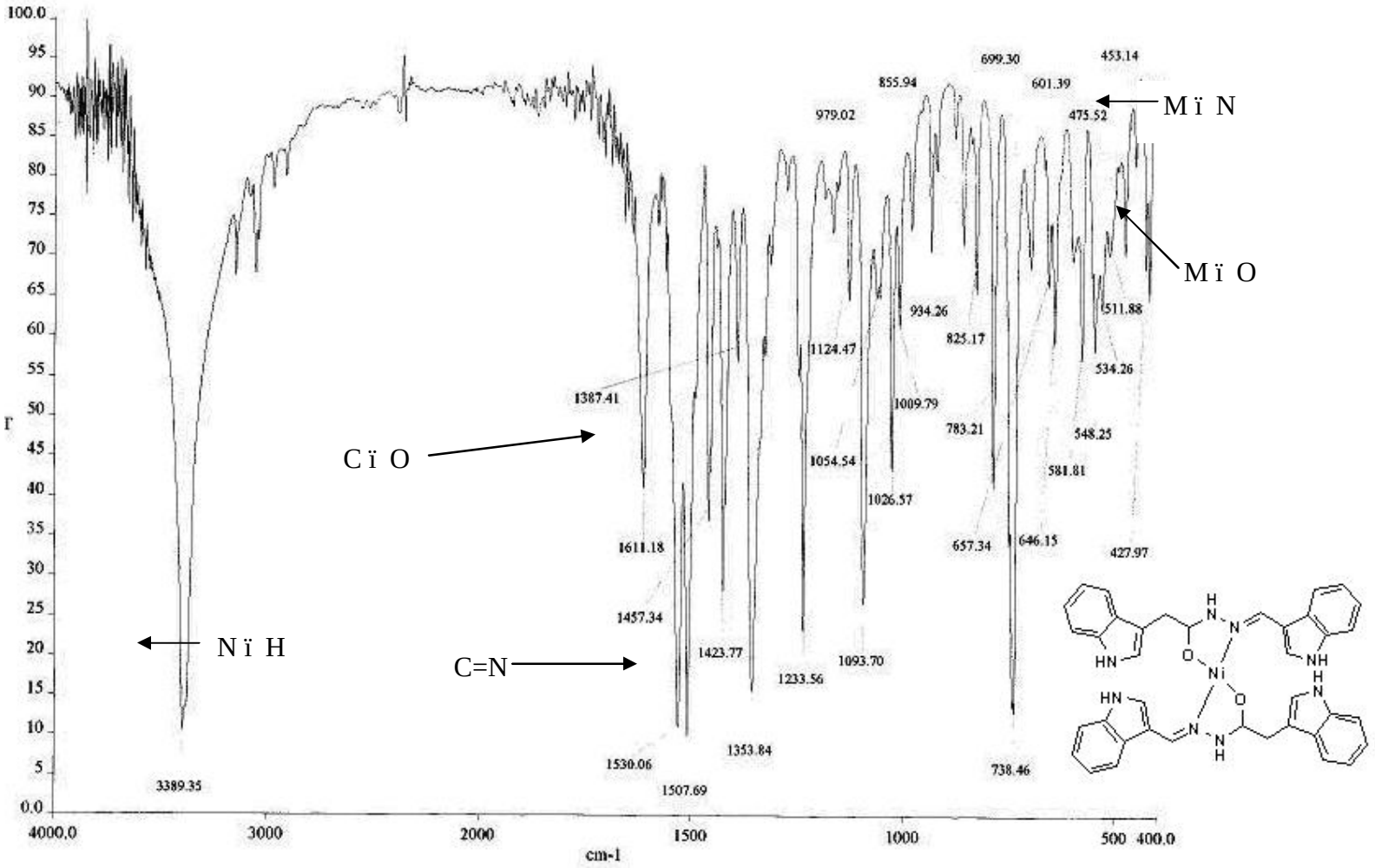

Figure 36:
IR spectrum of chloroindolecarboxaldehyde ï indolehydrazide (*L2*)

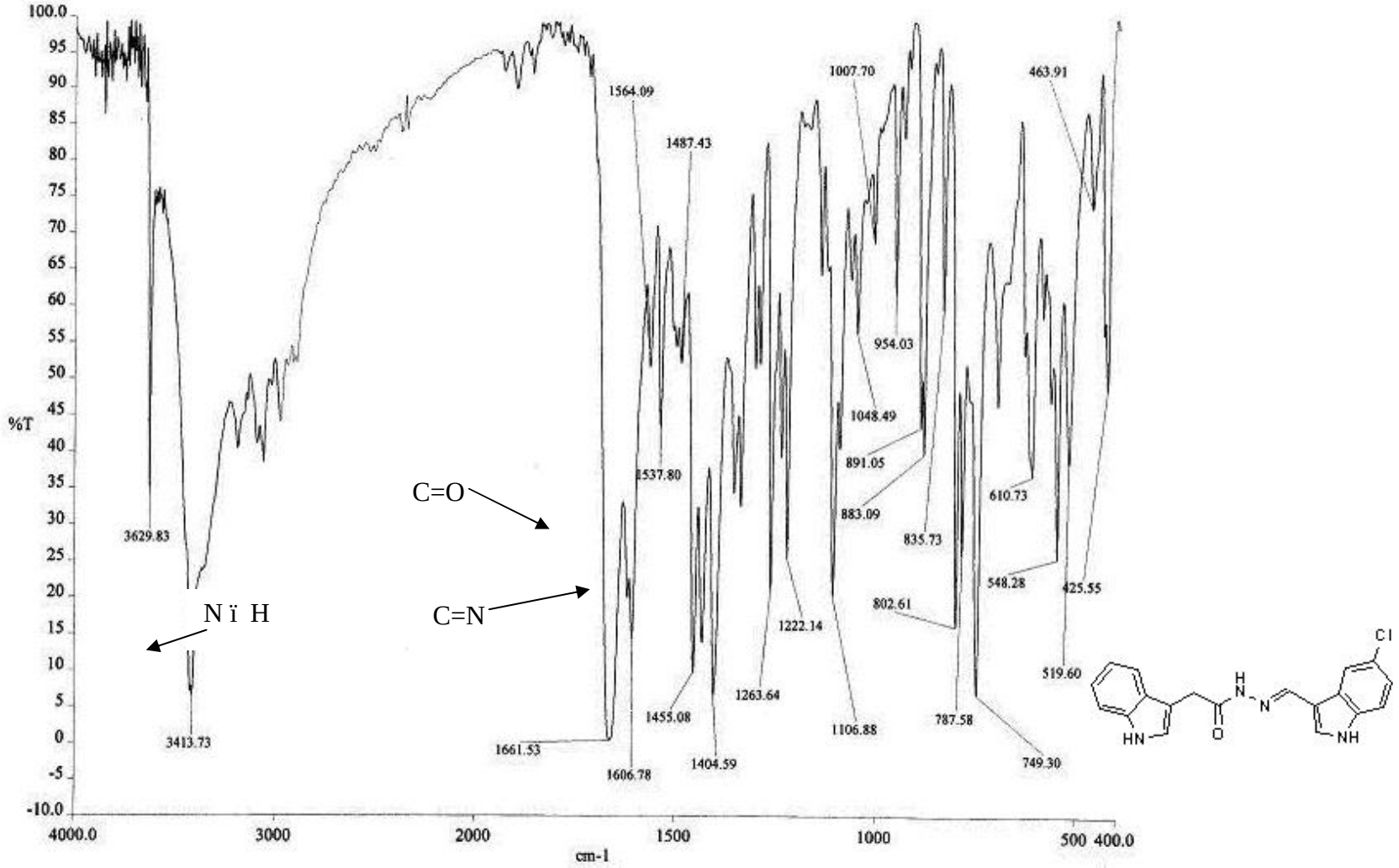

Figure 37:
IR spectrum of Cu(chloroindolecarboxaldehyde ï indolehydrazide)$_2$ (*CuL2*)

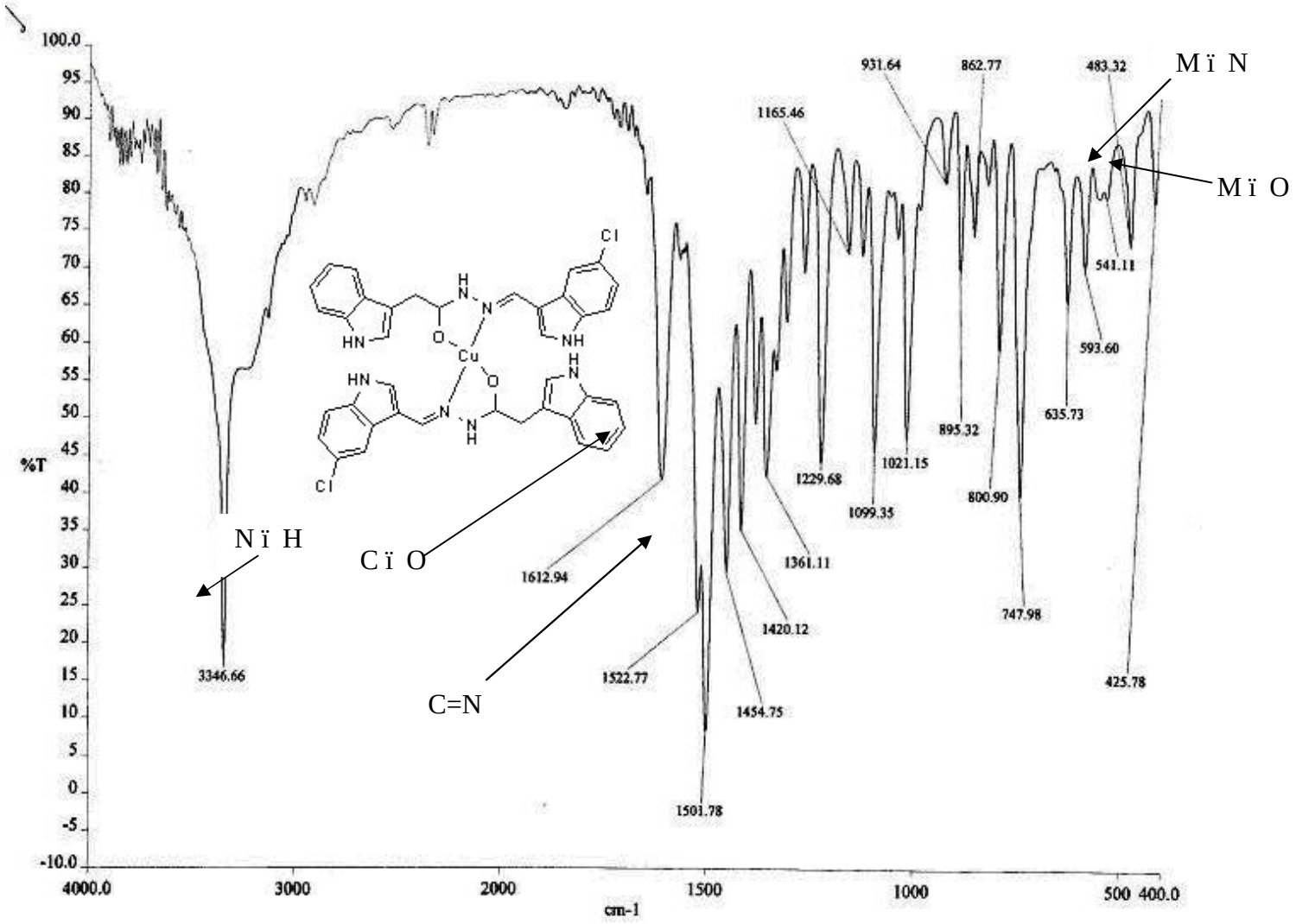

Figure 38:
IR spectrum of Ni(chloroindolecarboxaldehyde ï indolehydrazide)$_2$ (*NiL2*)

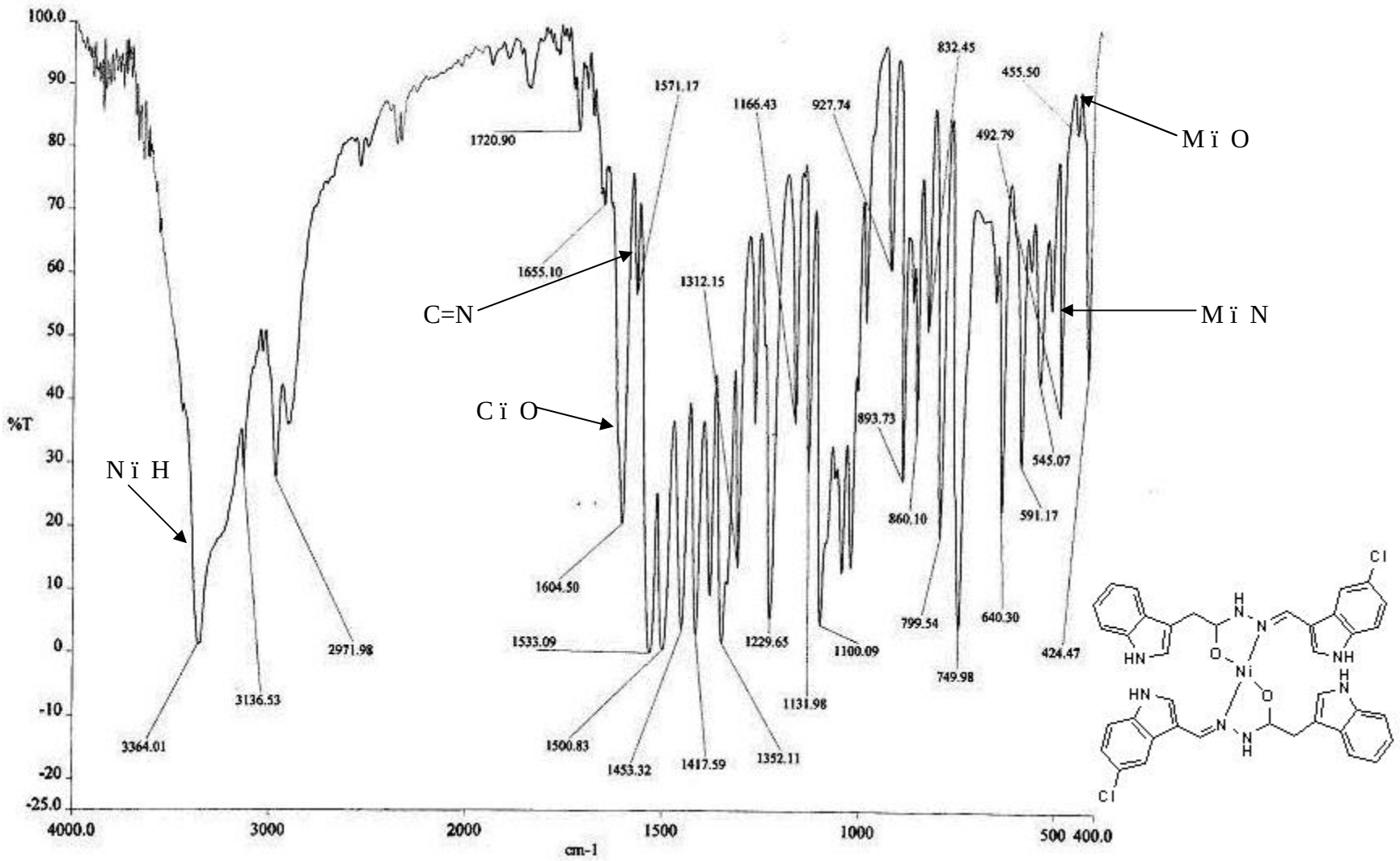

Figure 39:
IR spectrum of bromoindolecarboxaldehyde ï indolehydrazide (*L3*)

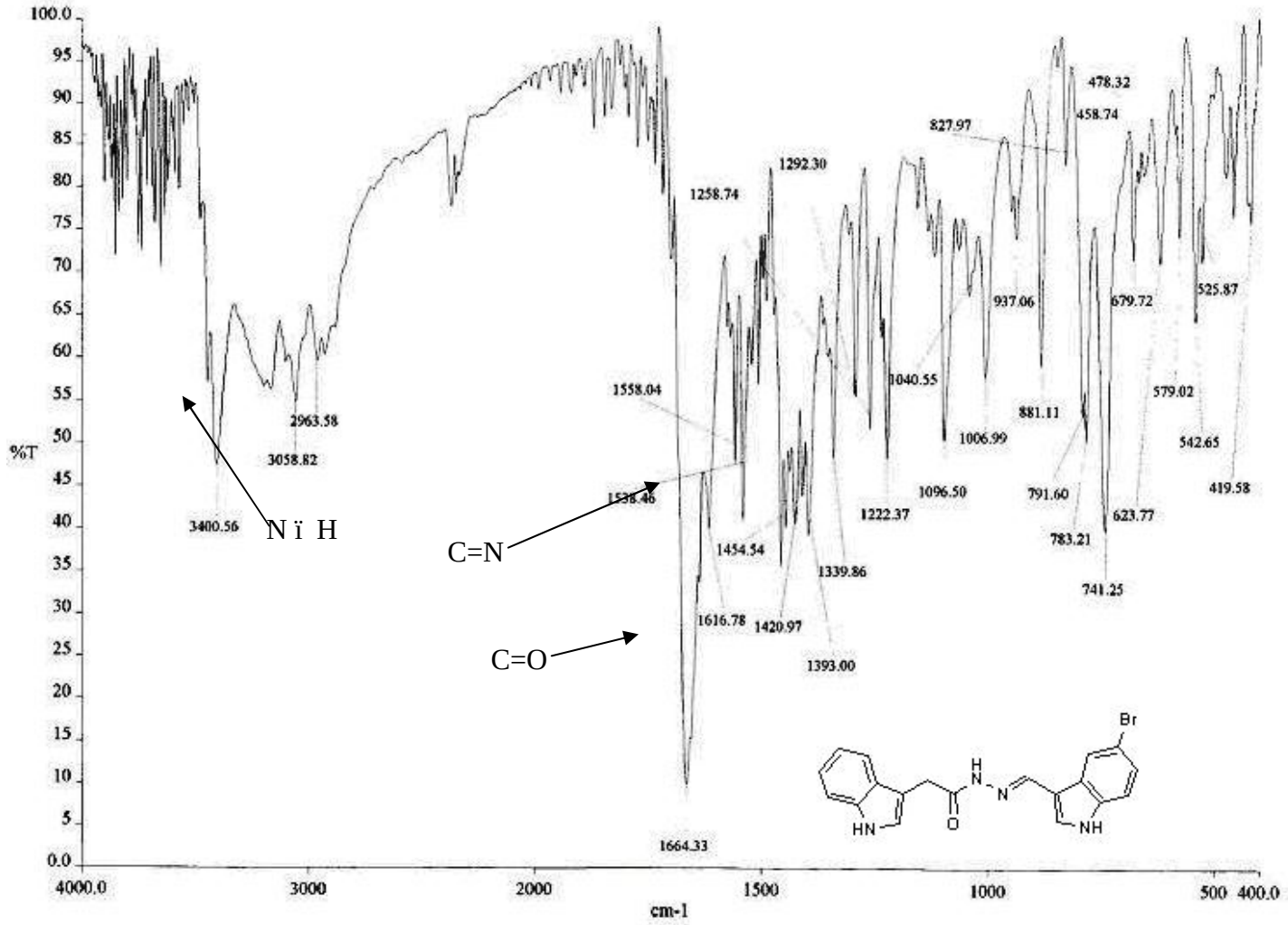

Figure 40:
IR spectrum of Cu(bromoindolecarboxaldehyde ï indolehydrazide)$_2$ (*CuL3*)

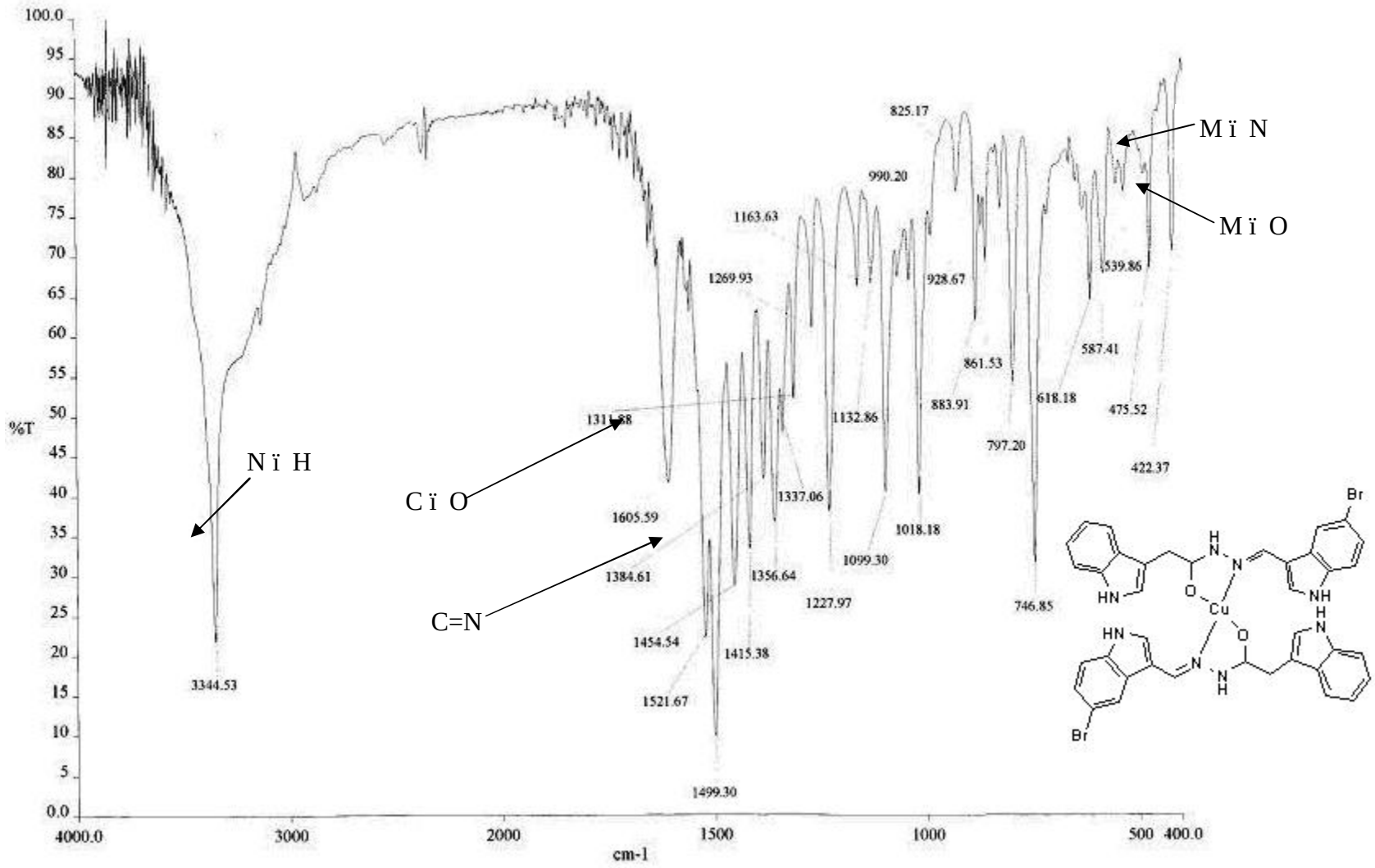

Figure 41:
IR spectrum of Ni(bromoindolecarboxaldehyde ï indolehydrazide)$_2$ (*NiL3*)