

Efficient synthesis of novel spiro-furo-pyrido-pyrimidine-indolines by manganese ferrite nanoparticles as a highly active magnetically reusable nanocatalyst in water

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SUPPORTING INFORMATION

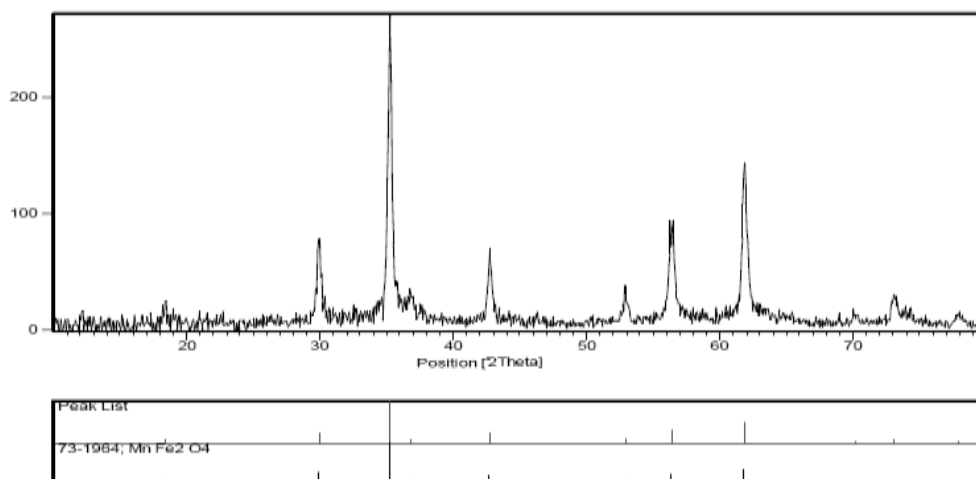
<i>List of contents</i>	<i>Page</i>	<i>List of contents</i>	<i>Page</i>
Title, author's name, address	1	¹³ C NMR of 4f	21
General methods and characterization data	2-9	¹ H NMR of 4g	22
XRD pattern of MnFe ₂ O ₄ nanoparticles (S1)	3	¹³ C NMR of 4g	23
Magnetization curve for the prepared nanoMnFe ₂ O ₄ (S2)	3	¹ H NMR of 4h	24
TEM image of nanoMnFe ₂ O ₄ (S3)	3	¹ H NMR of 4i	25
¹ H NMR of 4a	10	¹³ C NMR of 4i	26
¹³ C NMR of 4a	11	¹ H NMR of 4j	27
¹ H NMR of 4b	12	¹ H NMR of 4k	28
¹³ C NMR of 4b	13	¹³ C NMR of 4k	29
¹ H NMR of 4c	14	¹ H NMR of 4l	30
¹³ C NMR of 4c	15		
¹ H NMR of 4d	16		
¹³ C NMR of 4d	17		
¹ H NMR of 4e	18		
¹³ C NMR of 4e	19		
¹ H NMR of 4f	20		

Experimental Part

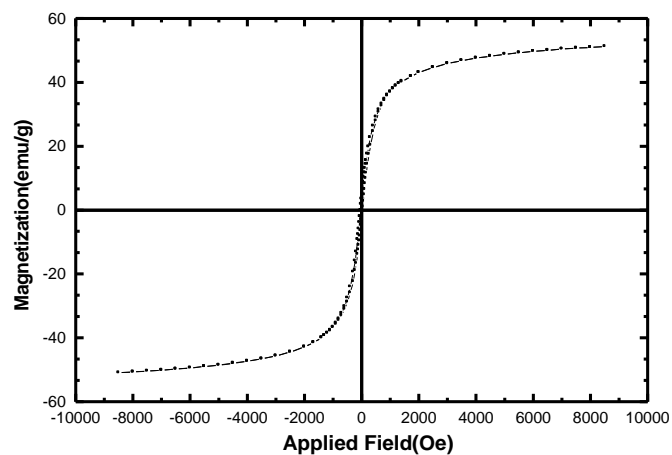
General. The chemicals used in this work were obtained from Fluka and Merck and were used without purification. Melting points were measured on an Electrothermal 9200 apparatus. Mass spectra were recorded on a Shimadzu QP 1100 Ex mass spectrometer operating at an ionization potential of 70 eV. IR spectra were recorded as KBr pellets on a Perkin-Elmer 781 spectrophotometer and an Impact 400 Nicolet FT-IR spectrophotometer. ^1H NMR and ^{13}C NMR spectra were recorded in DMSO- d_6 solvents on a Bruker DRX-400 spectrometer with tetramethylsilane as internal reference. The elemental analyses (C, H, N) were obtained from a Carlo ERBA Model EA 1108 analyzer. X-ray diffraction (XRD) pattern of the as-synthesized material was obtained using a Holland Philips Xpert X-ray powder diffraction (XRD) diffractometer (CuK α radiation, $\lambda = 0.154056$ nm), at a scanning speed of $2^\circ/\text{min}$ from 10° to 100° (2θ). The nanocatalyst was determined using a Philips EM 208 transmission electron microscope (TEM) operated at a 100 kV accelerating voltage. The purity determination of the substrates and reaction monitoring were accomplished by TLC on silica-gel polygram SILG/UV 254 plates (from Merck Company).

Typical experimental procedure for the preparation of catalyst

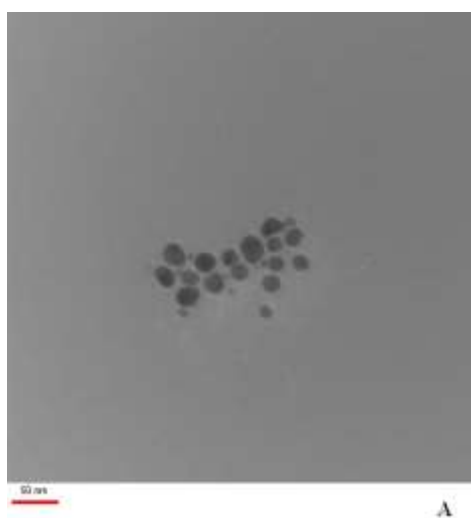
MnFe_2O_4 nanoparticles have been prepared following the reported standard protocol by co-precipitation of MnCl_2 and FeCl_3 in water in the presence of sodium hydroxide. Briefly, $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ were taken in molar ratio of $\text{Mn}^{2+}:\text{Fe}^{3+} = 1:2$ to prepare $0.3 \text{ mol} \cdot \text{L}^{-1}$ metal ion solution of 100 ml containing $0.1 \text{ mol} \cdot \text{L}^{-1}$ Mn^{2+} and $0.2 \text{ mol} \cdot \text{L}^{-1}$ Fe^{3+} , then was slowly dropped into 100 ml NaOH solution of $3 \text{ mol} \cdot \text{L}^{-1}$ at the preheated temperature of 95°C . After aging for 2 h with continuous stirring, the mixture was filtered, washed and dried at 60°C for 12 h. The XRD patterns of calcinated precipitate indicate that the powder is mainly composed of MnFe_2O_4 (S1) and the vibrating sample magnetometer curve of synthesized MnFe_2O_4 nanoparticles at room temperature shows that the hysteresis loop for the sample is completely reversible (S2). Also, the TEM image of MnFe_2O_4 was shown that manganese ferrite nanoparticles have a mean diameter of about 25-30 nm (S3).



S1. XRD pattern of MnFe₂O₄ nanoparticles



S2. Magnetization curve for the prepared nano MnFe₂O₄.

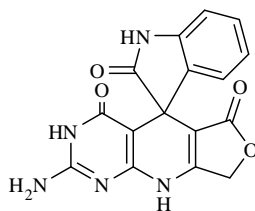


S3. The TEM image of MnFe₂O₄ before the reaction

Typical procedure for the preparation of spiro-furo-pyrido-pyrimidine-indoline derivatives 4(a-l).

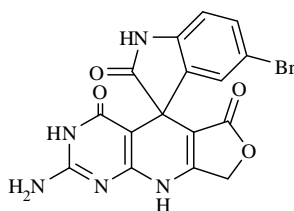
Isatin (1 mmol), tetronic acid (1 mmol) and 2, 6-diaminopyrimidin-4(3H)-one or uraciles (1 mmol) were placed in a 10 ml round-bottomed flask in H₂O (5 mL). Sequentially MnFe₂O₄ (5 mol%) was added. The resulting mixture was stirred at 90 °C for 6h and monitored by TLC until conversion of the starting materials is satisfactory. After completion of the reaction, the reaction mixture was magnetically concentrated with the aid of a magnet to separate the catalyst. Catalyst was separated and washed several times with acetone followed by water, then dried under vacuum and reutilized four times for the same reaction. The reaction mixture was filtered and the precipitate washed with water and recrystallized by EtOH to afford the pure product and was identified by physical and spectroscopic data.

2-Amino-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4a):



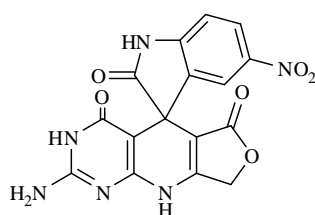
White powder (Yield: 82%, 0.277 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3420, 3156, 1751, 1713, 1638; ¹H NMR (DMSO-*d*₆, 400 MHz): δ_{ppm} : 4.84 (2H, s, OCH₂), 6.58-7.06 (6H, m, ArH and NH₂), 10.06 (1H, s, NH), 10.19 (1H, s, NH), 10.47 (1H, s, NH); ¹³C NMR (DMSO-*d*₆, 100 MHz): δ_{ppm} : 47.4, 65.7, 91.0, 98.7, 108.9, 121.5, 123.8, 127.9, 136.0, 142.7, 155.0, 155.4, 158.8, 161.2, 170.2, 179.0; Anal. Calcd for C₁₆H₁₁N₅O₄: C, 56.98; H, 3.29; N, 20.76; Found C, 56.97; H, 3.31; N, 20.71, MS: m/z 337.

2-Amino-5'-bromo-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4b):



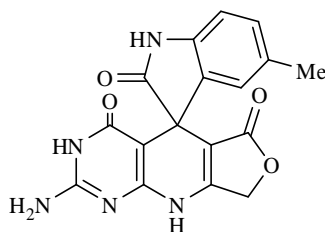
Cream powder (Yield: 80%, 0.332 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3465, 3249, 1729, 1645, 1519; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 4.86 (2H, s, OCH₂), 6.65-7.24 (5H, m, ArH and NH₂), 10.15 (1H, s, NH), 10.37 (1H, s, NH), 10.52 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 47.7, 65.9, 90.5, 97.9, 110.9, 113.2, 126.6, 130.7, 138.3, 142.2, 155.1, 155.6, 159.3, 161.2, 170.2, 178.7; Anal. Calcd for C₁₆H₁₀BrN₅O₄: C, 46.17; H, 2.42; N, 16.83; Found C, 46.21; H, 2.39; N, 16.88; MS: m/z, 416, 414.

2-Amino-5'-nitro-5'-nitro-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4c):



Yellow powder (Yield: 77%, 0.294 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3449, 3173, 1727, 1640, 1594; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 4.91 (2H, s, OCH₂), 6.91-8.10 (5H, m, ArH and NH₂), 10.29 (1H, s, NH), 10.59 (1H, s, NH), 11.01 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 47.5, 66.1, 90.0, 97.3, 109.0, 119.3, 125.8, 136.8, 142.4, 149.5, 155.2, 155.7, 159.7, 161.3, 170.2, 179.7; Anal. Calcd for C₁₆H₁₀N₆O₆: C, 50.27; H, 2.64; N, 21.98. Found C, 50.24; H, 2.60; N, 21.93; MS: m/z 382.

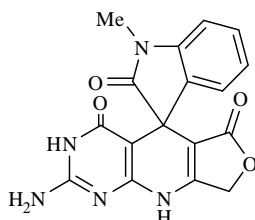
2-Amino-5'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4d):



Cream powder (Yield: 79%, 0.277 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3392, 3271, 1732, 1641, 1518 ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 2.14 (3H, s, CH₃), 4.84 (2H, s, OCH₂), 6.56-6.84 (5H, m, ArH and NH₂), 10.06 (1H, s, NH), 10.09 (1H, s, NH), 10.47 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 21.0, 47.4, 65.7, 91.1, 98.8, 108.6, 124.4, 128.2, 130.1, 136.1, 140.3, 155.0, 155.4, 158.7, 161.1, 170.2,

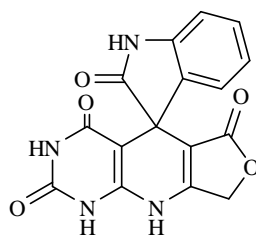
178.9; Anal. Calcd for $C_{17}H_{13}N_5O_4$: C, 58.12; H, 3.73; N, 19.93. Found C, 58.09; H, 3.78; N, 19.88; MS: m/z 351.

2-Amino-1'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4e):



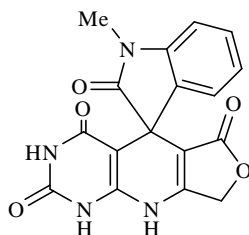
Cream powder (Yield: 76%, 0.267 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3490, 3376, 3211, 1680, 1647; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 3.08 (3H, s, CH_3), 4.90 (2H, s, OCH_2), 6.65-7.20- (6H, m, ArH and NH_2), 10.11 (1H, s, NH), 10.35 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 26.7, 46.9, 65.8, 90.8, 98.4, 107.8, 122.4, 123.5, 128.3, 135.1, 144.1, 155.0, 155.4, 159.0, 161.1, 170.2, 177.6; Anal. Calcd for $C_{17}H_{13}N_5O_4$: C, 58.07; H, 3.73; N, 19.93;. Found C, 58.10; H, 3.76; N, 19.90; MS: m/z 351.

1H-Spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4f):



White powder (Yield: 83%, 0.280 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3175, 3103, 1693, 1650, 1538; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 4.90 (2H, s, OCH_2), 6.69-7.08 (4H, m, ArH), 10.02 (1H, s, NH), 10.33 (1H, s, NH), 10.83 (1H, s, NH), 11.59 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 46.8, 66.4, 89.1, 101.0, 109.1, 121.6, 124.0, 128.4, 135.1, 142.8, 146.0, 150.4, 157.3, 162.3, 169.6, 178.3; Anal. Calcd for $C_{16}H_{10}N_4O_5$: C, 56.81; H, 2.98; N, 16.56; Found C, 56.84; H, 2.94; N, 16.52; MS: m/z 338.

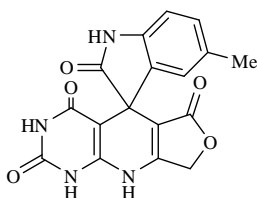
**1'-Methyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6
(3H,8H,9H)-tetraone (4g):**



White powder (Yield: 78%, 0.274 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3495, 3350, 3211, 1755, 1683; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 3.09 (3H, s, CH₃), 4.90 (2H, s, OCH₂), 6.89-7.21 (4H, m, ArH), 10.05 (1H, s, NH), 10.82 (1H, s, NH), 11.59 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 26.7, 46.3, 66.5, 89.0, 100.8, 108.0, 122.4, 123.7, 128.6, 134.2, 144.3, 146.0, 150.4, 157.5, 162.2, 169.6, 177.0; Anal. Calcd for C₁₇H₁₂N₄O₅: C, 57.96; H, 3.43; N, 15.90;. Found C, 57.92; H, 3.41; N, 15.94; MS: m/z 352.

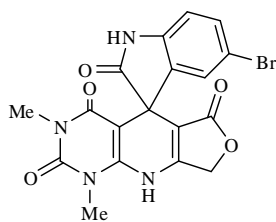
Due to very low solubility of the products **4h**, we can not report the ^{13}C NMR data for this product.

**5'-Methyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6
(3H,8H,9H)-tetraone (4h):**



Cream powder (Yield: 75%, 0.264 g). mp: >350 °C. IR (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 3483, 3176, 1759, 1694; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 2.15 (3H, s, CH₃), 4.89 (2H, s, OCH₂), 6.59-6.88 (3H, m, ArH), 9.97 (1H, s, NH), 10.22 (1H, s, NH), 10.81 (1H, s, NH), 11.53 (1H, s, NH); Anal. Calcd for C₁₇H₁₂N₄O₅: C, 57.96; H, 3.43; N, 15.90;. Found C, 57.98; H, 3.38; N, 15.86; MS: m/z 352.

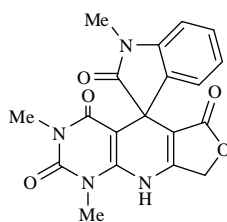
5'-Bromo-1,3-dimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4i):



Cream powder (Yield: 76%, 0.338 g). mp: >350 °C. IR (KBr) (ν_{\max} / cm^{-1}): 3443, 3162, 1765, 1645; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 3.12 (3H, s, CH₃), 3.38 (3H, s, CH₃), 3.44 (3H, s, CH₃), 4.97 (2H, s, OCH₂), 6.91-7.39 (3H, m, ArH), 10.45 (1H, s, NH), 11.66 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 27.59, 31.8, 49.8, 65.7, 92.1, 95.2, 115.4, 119.1, 123.8, 130.4, 132.0, 136.6, 151.6, 153.3, 157.4, 159.4, 170.9, 180.0; Anal. Calcd for C₁₈H₁₃BrN₄O₅: C, 48.56; H, 2.94; N, 12.58. Found C, 48.60; H, 2.90; N, 12.55; MS: m/z 446, .444.

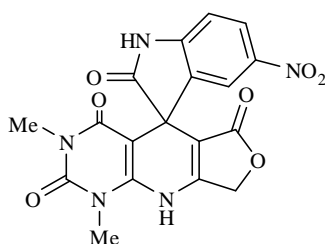
Due to very low solubility of the products **4j**, we can not report the ^{13}C NMR data for this product.

1,1',3-Trimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline] [2,2',4,6(3H,8H,9H)-tetraone (4j):



White powder (Yield: 72%, 0.273 g). mp: >350 °C. IR (KBr) (ν_{\max} / cm^{-1}): 3447, 3252, 1680, 1642; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 2.99 (3H, s, CH₃), 3.12 (3H, s, CH₃), 3.44 (3H, s, CH₃), 4.97 (2H, s, CH₂), 6.91-7.20 (4H, m, ArH), 10.58 (1H, s, NH); Anal. Calcd for C₁₉H₁₆N₄O₅: C, 60.00; H, 4.24; N, 14.73. Found C, 60.04; H, 4.21; N, 14.68; MS: m/z 380.

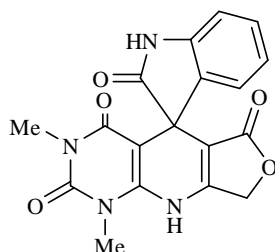
1,3-Dimethyl-5'-nitro-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline] [2,2',4,6(3H,8H,9H)-tetraone (4k):



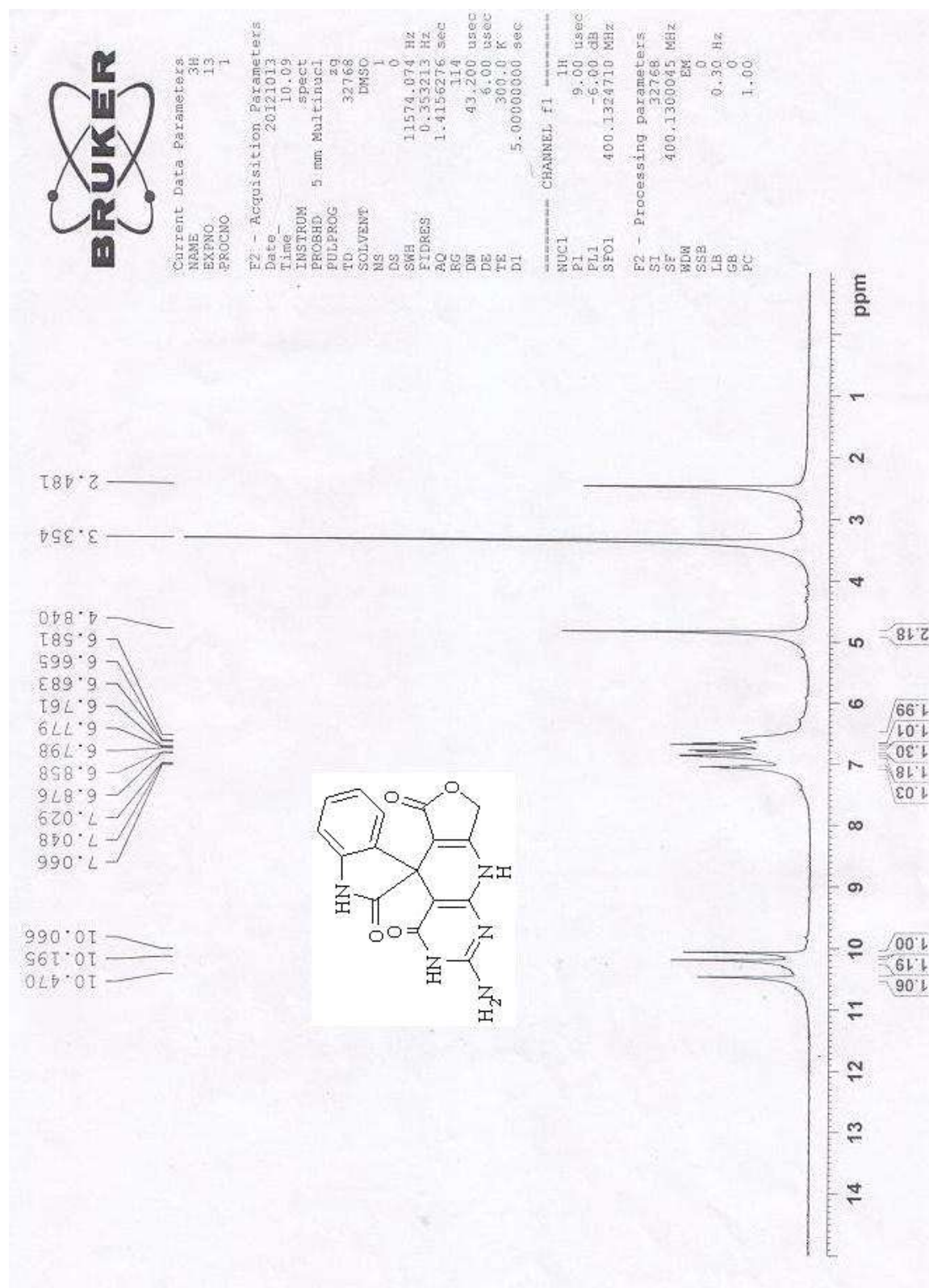
Yellow powder (Yield: 74%, 0.304 g). mp: >350 °C. IR (KBr) (ν_{\max} / cm^{-1}): 3525, 3114, 1739, 1651. ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 3.01 (3H, s, CH_3), 3.39 (3H, s, CH_3), 5.05 (2H, s, OCH_2), 7.14-8.10 (3H, m, ArH), 11.02 (1H, s, NH), 11.86 (1H, s, NH); ^{13}C NMR (DMSO- d_6 , 100 MHz): δ_{ppm} : 27.6, 31.9, 49.8, 66.0, 94.1, 95.7, 117.7, 122.2, 124.3, 125.3, 142.9, 143.3, 151.6, 153.4, 157.5, 159.3, 170.6, 179.7; Anal. Calcd for $\text{C}_{18}\text{H}_{13}\text{N}_5\text{O}_7$: C, 52.56; H, 3.19; N, 17.03. Found C, 52.53; H, 3.24; N, 17.07; MS: m/z 411.

Due to very low solubility of the products **4l**, we can not report the ^{13}C NMR data for this product.

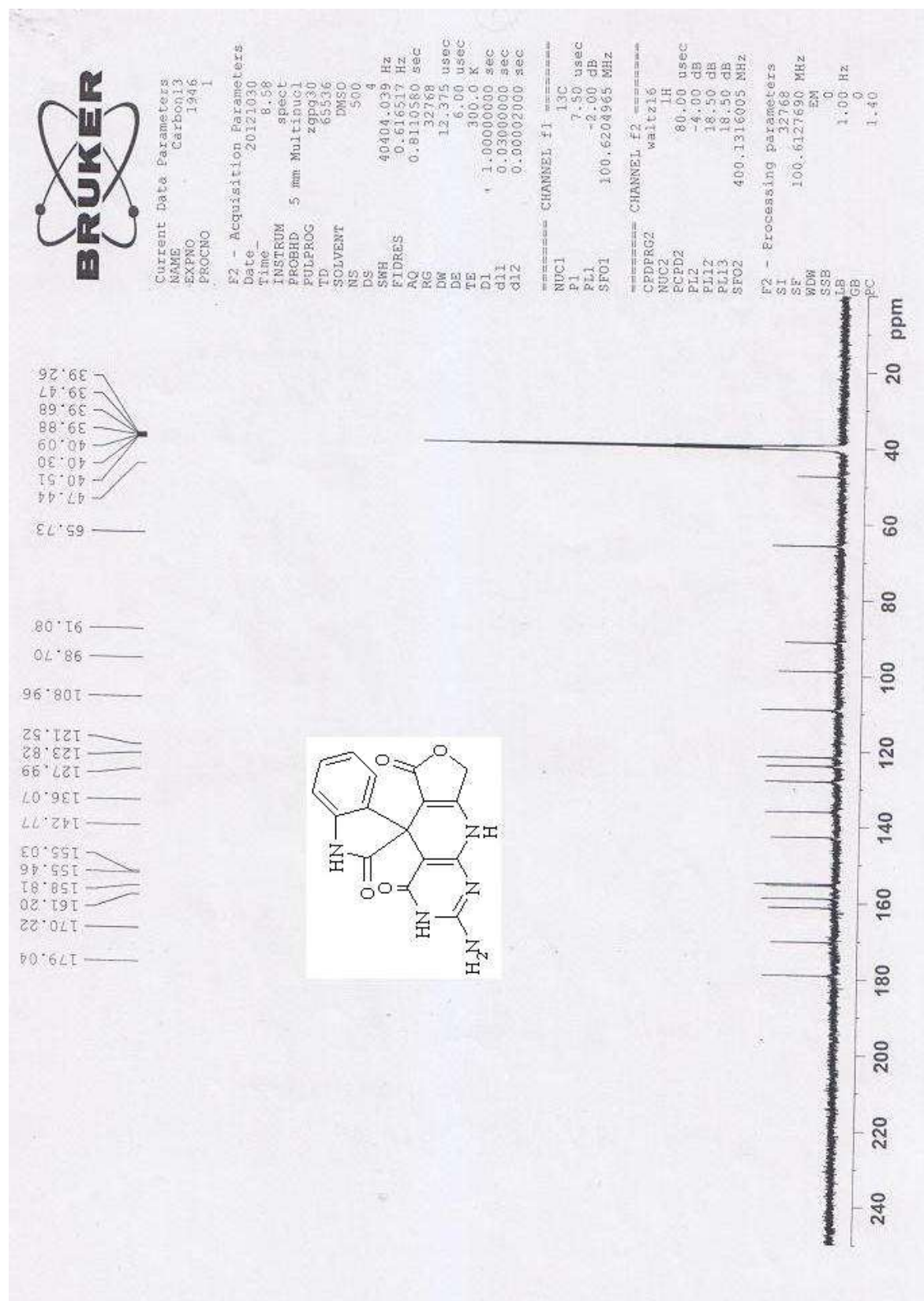
1,3-Dimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6 (3H,8H,9H)-tetraone (4l):



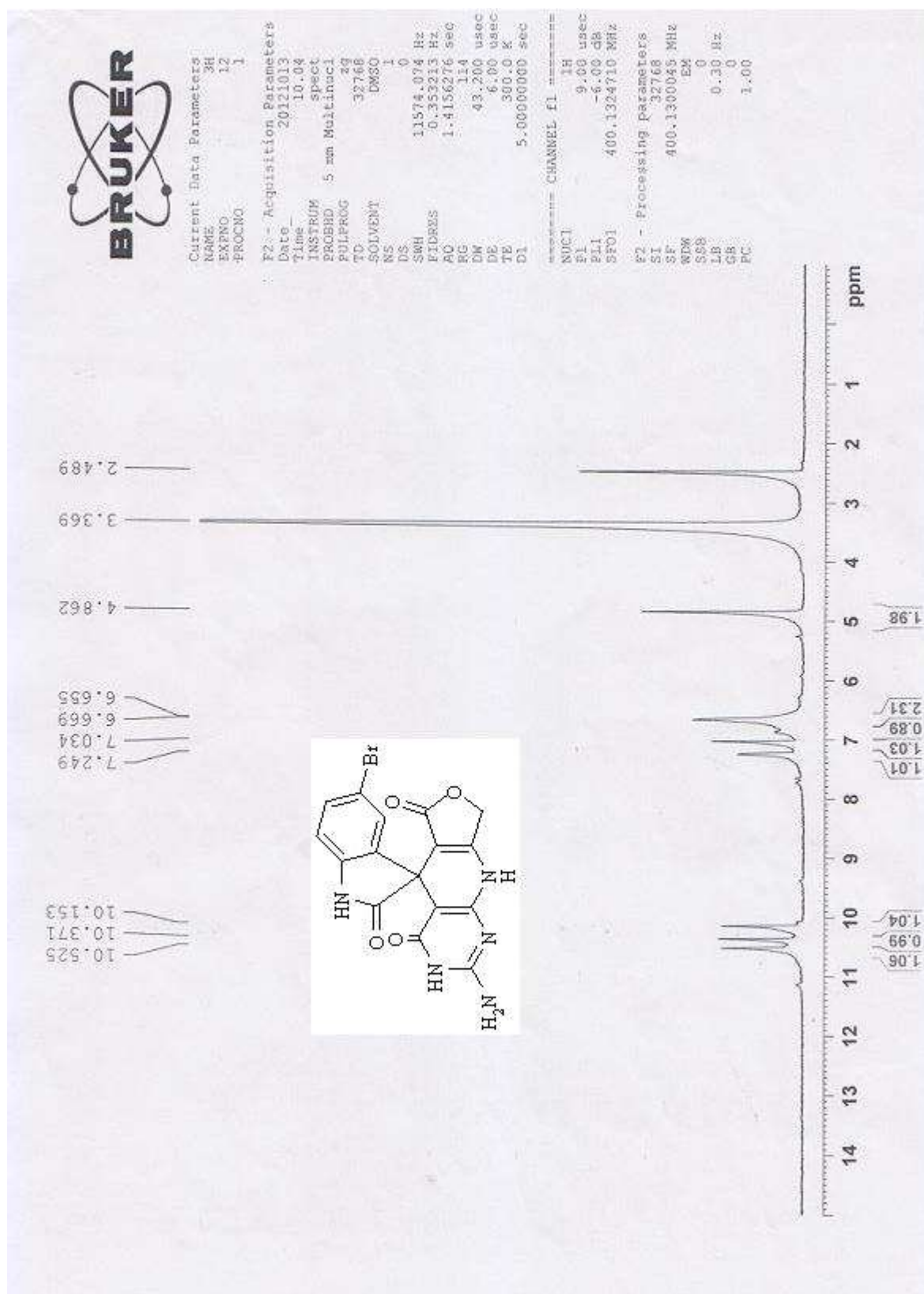
White powder (Yield: 75%, 0.274 g). mp: >350 °C. IR (KBr) (ν_{\max} / cm^{-1}): 3543, 3234, 1725, 1648; ^1H NMR (DMSO- d_6 , 400 MHz): δ_{ppm} : 3.04 (3H, s, CH_3), 3.33 (3H, s, CH_3), 4.95 (2H, s, CH_2), 6.90-7.23 (4H, m, ArH), 10.33 (1H, s, NH), 11.65 (1H, s, NH); Anal. Calcd for $\text{C}_{18}\text{H}_{14}\text{N}_4\text{O}_5$: C, 59.02; H, 3.85; N, 15.29. Found C, 59.07; H, 3.80; N, 15.34; MS: m/z 366.



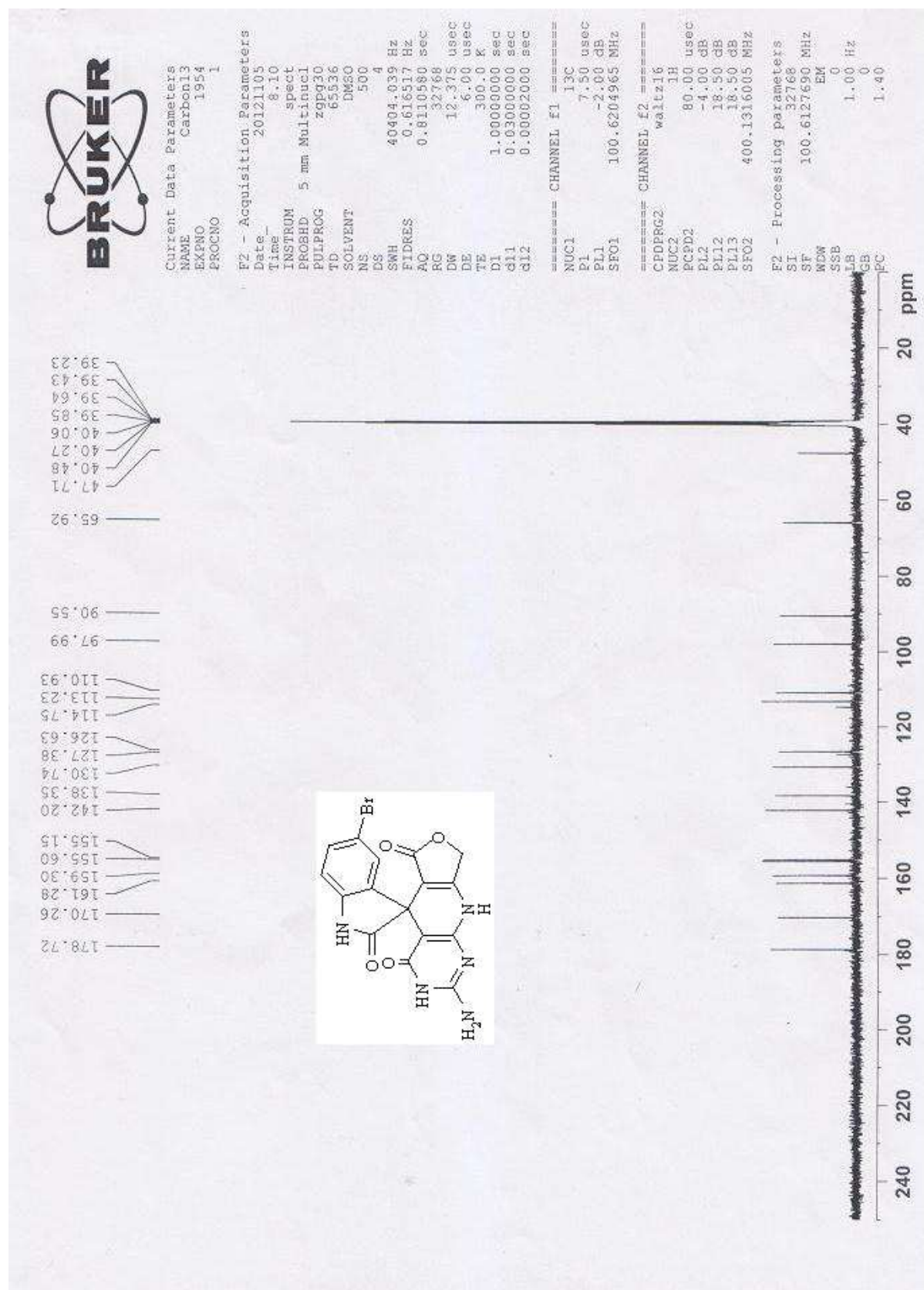
¹H NMR spectrum of 2-amino-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4a):



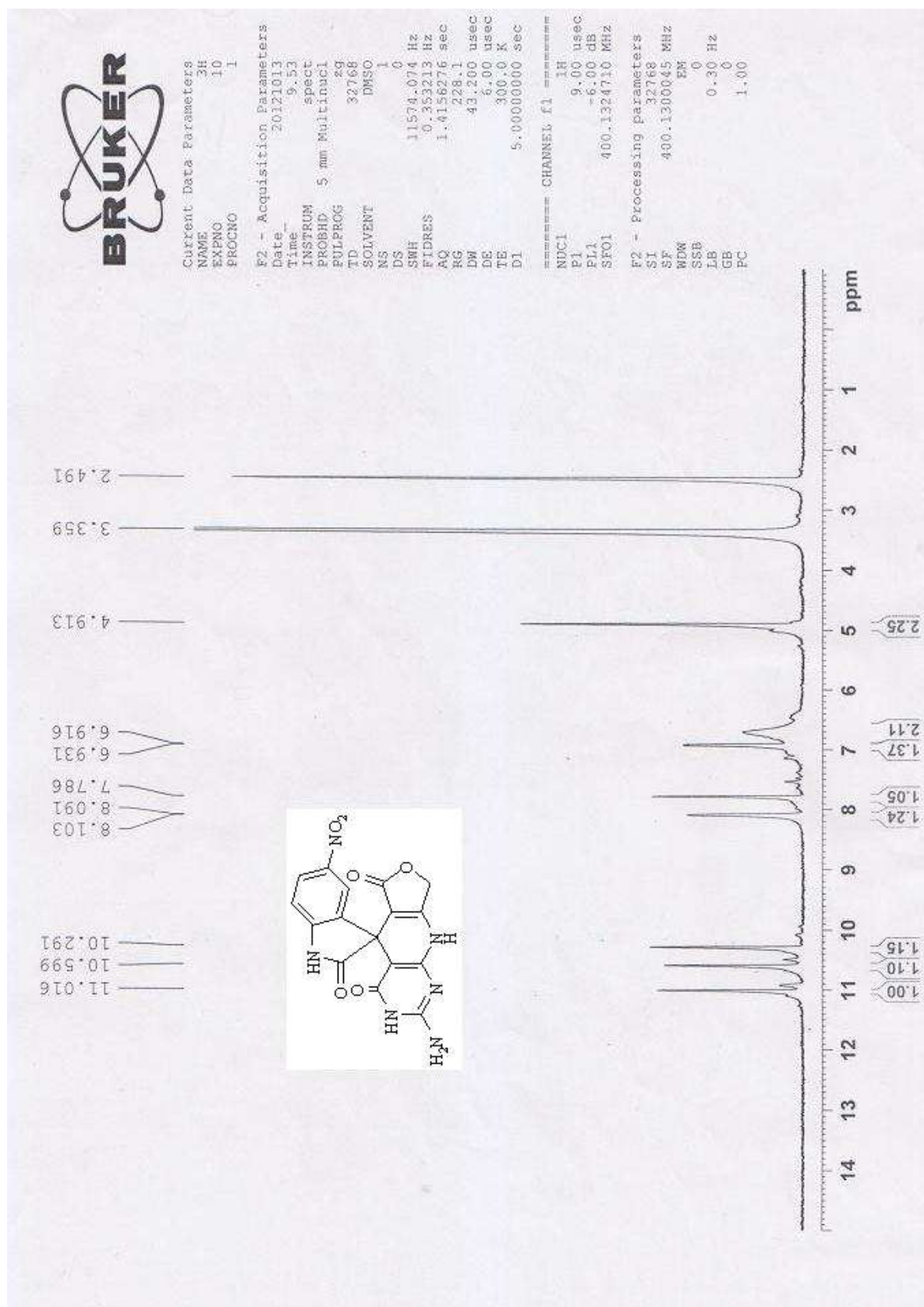
¹³C NMR spectrum of 2-amino-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4a):



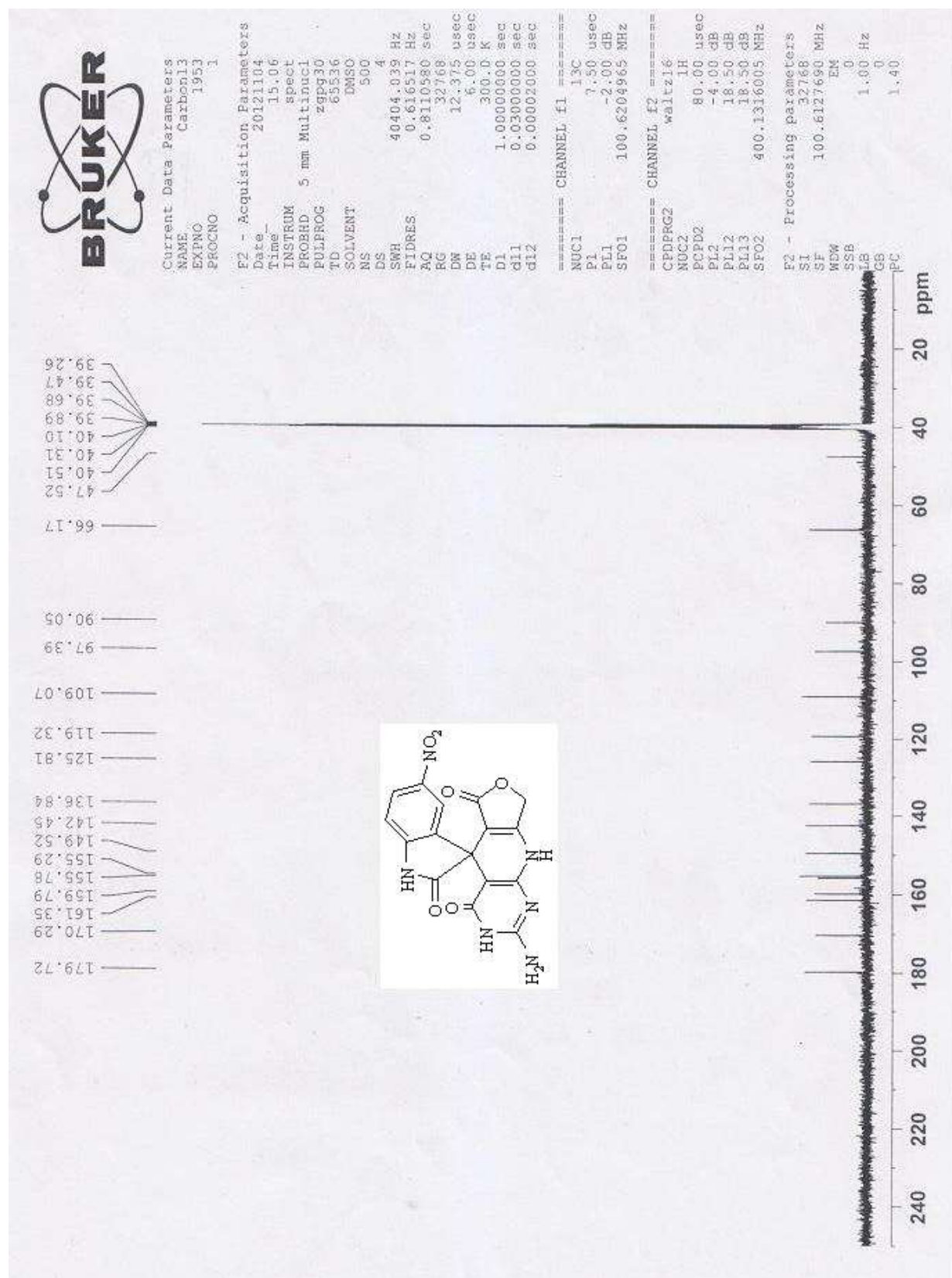
¹H NMR spectrum of 2-amino-5'-bromo-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6,8H,9H)-trione (4b):



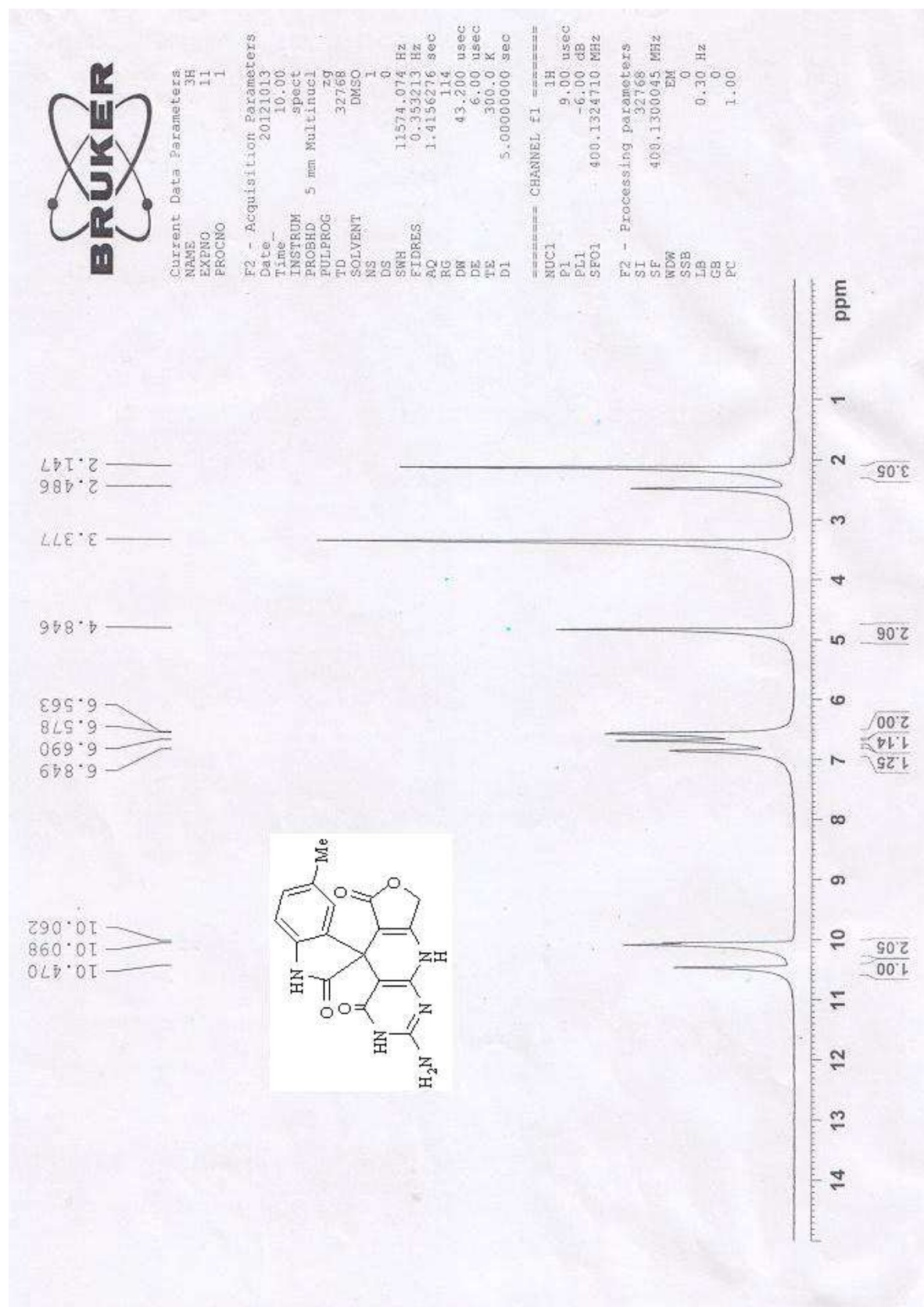
¹³C NMR spectrum of 2-amino-5'-bromo-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6,8H,9H)-trione (4b):



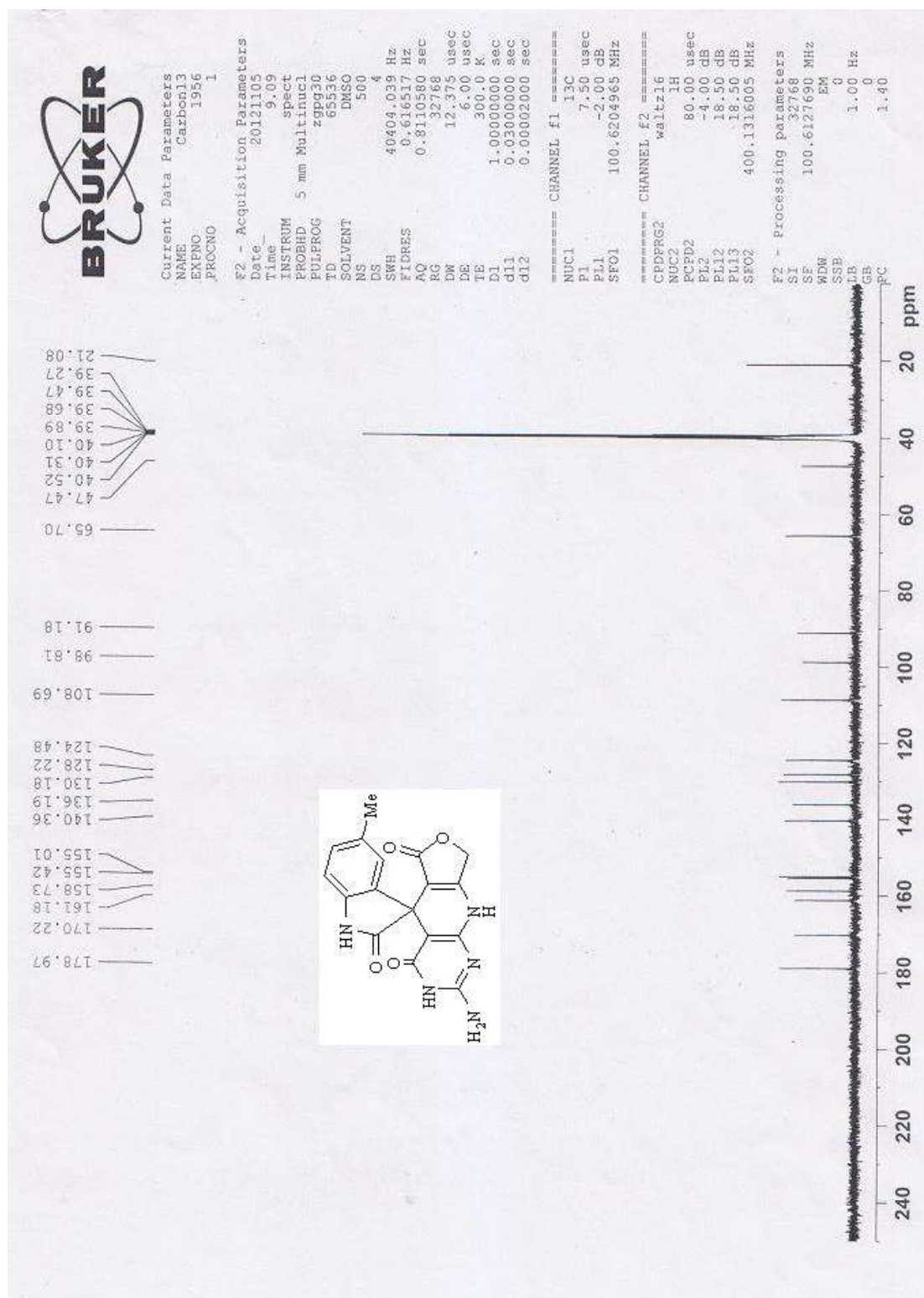
¹H NMR spectrum of 2-amino-5'-nitro-5-nitro-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4c):



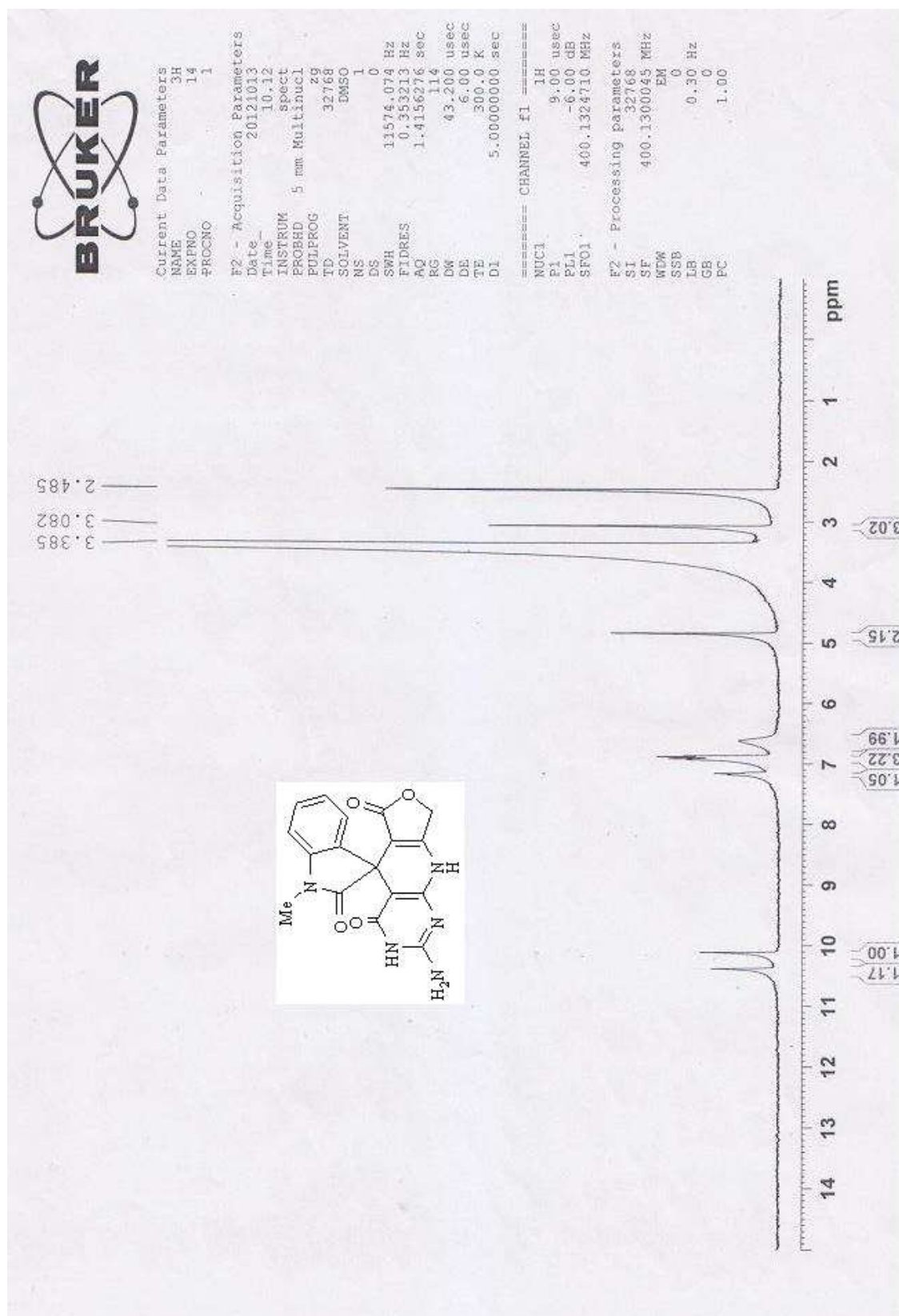
¹³C NMR spectrum of 2-amino-5'-nitro-5'-nitro-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4c):



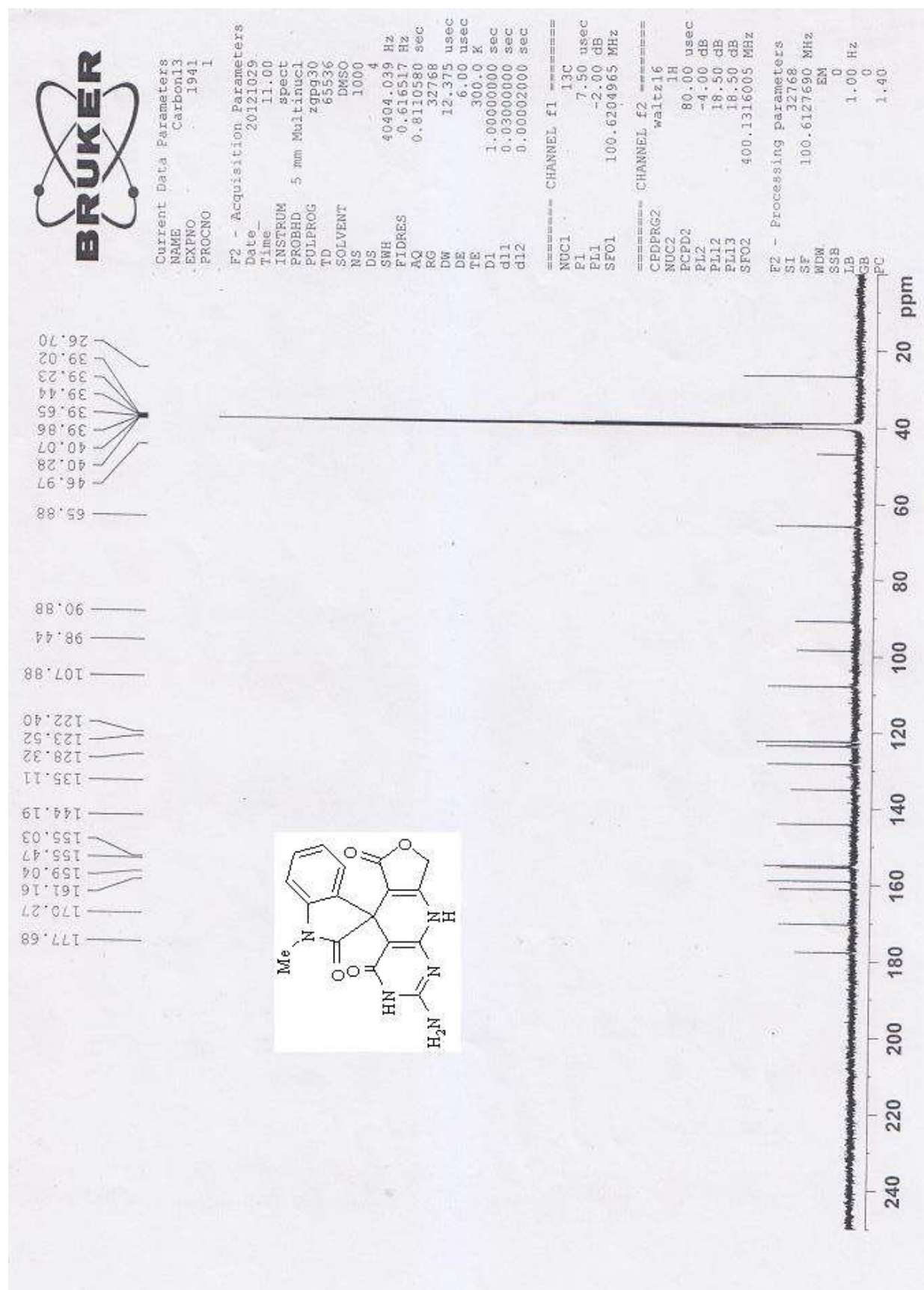
¹H NMR spectrum of 2-amino-5'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4d):



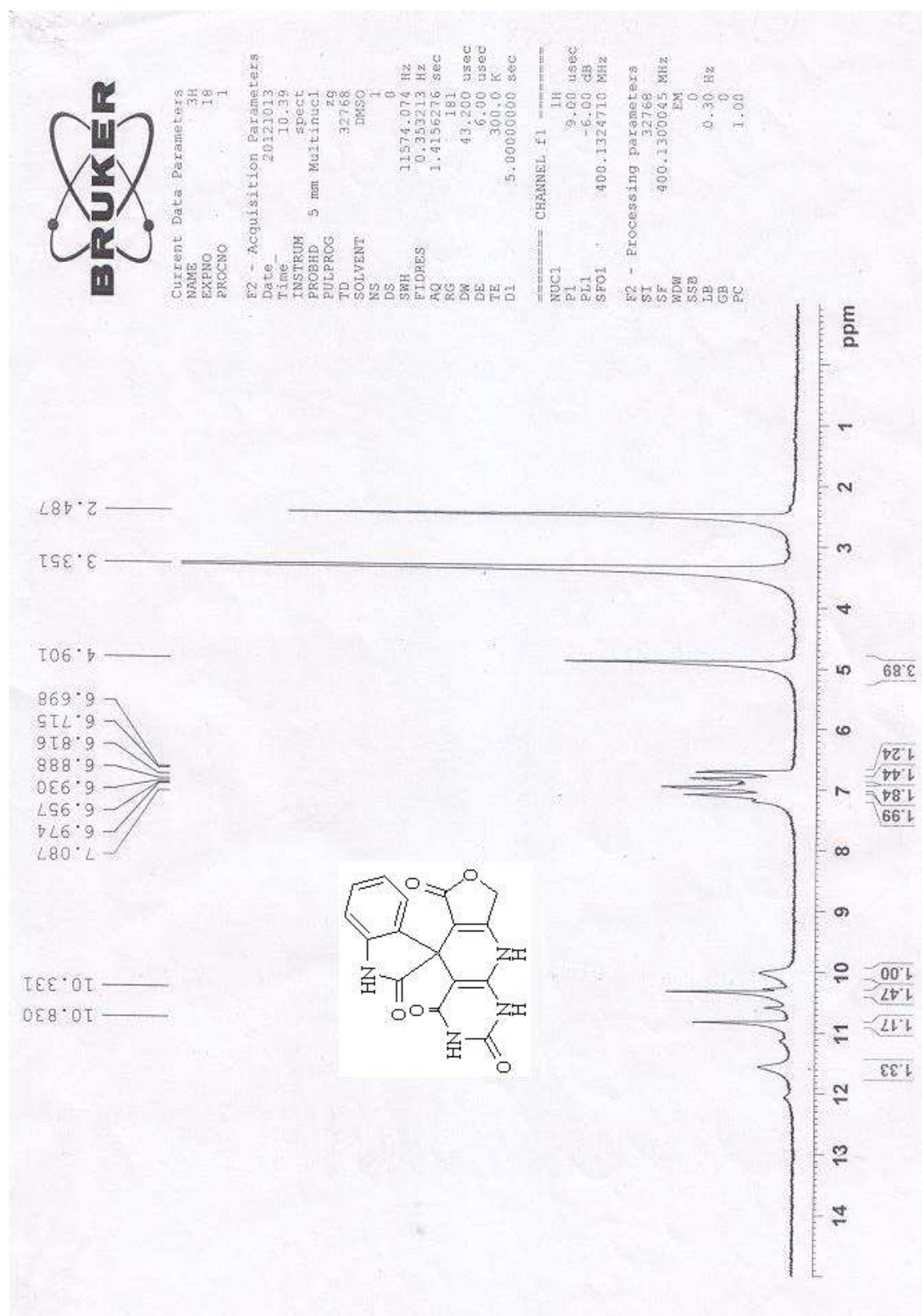
¹³C NMR spectrum of 2-amino-5'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4d):



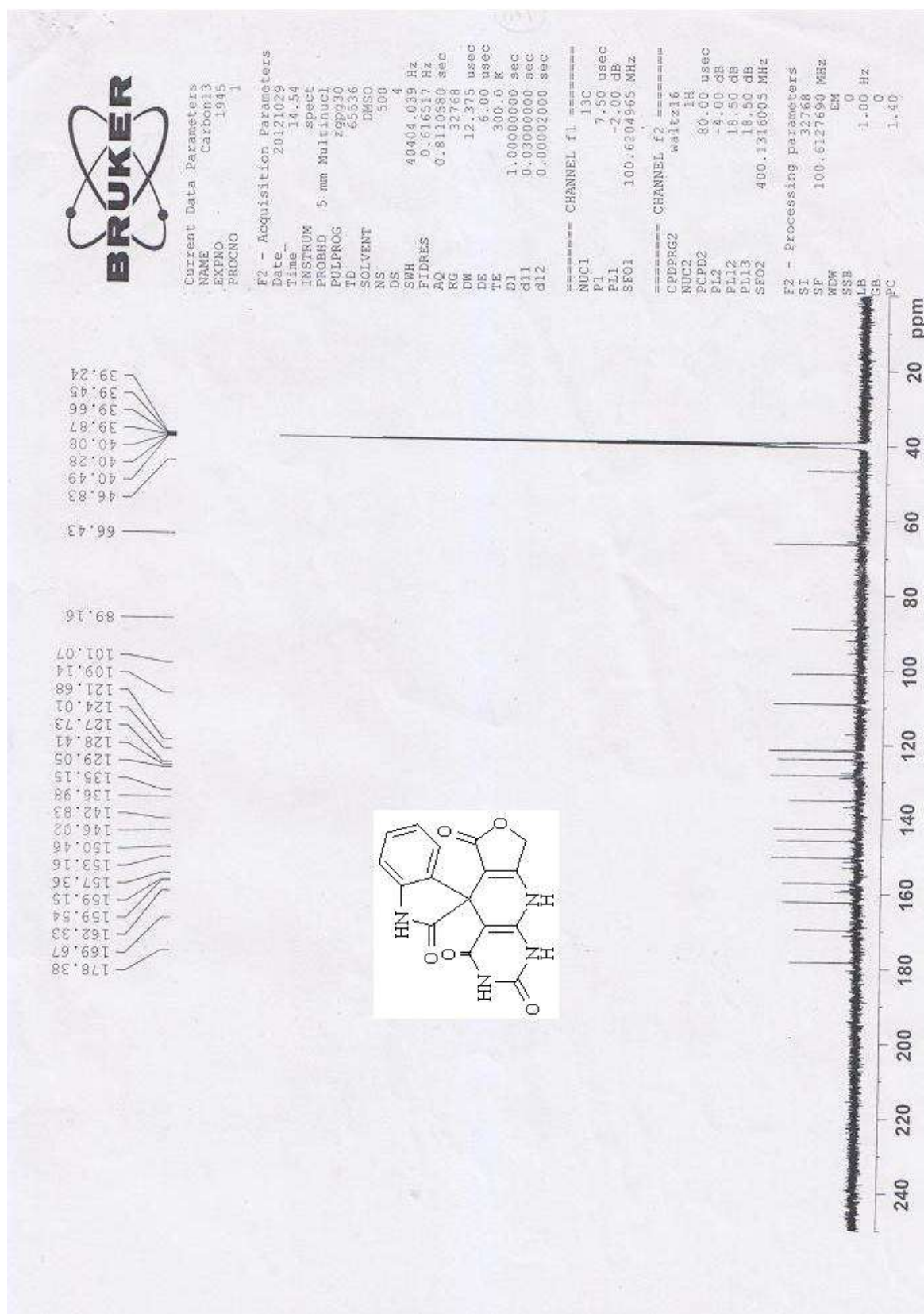
¹H NMR spectrum of 2-amino-1'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4e):



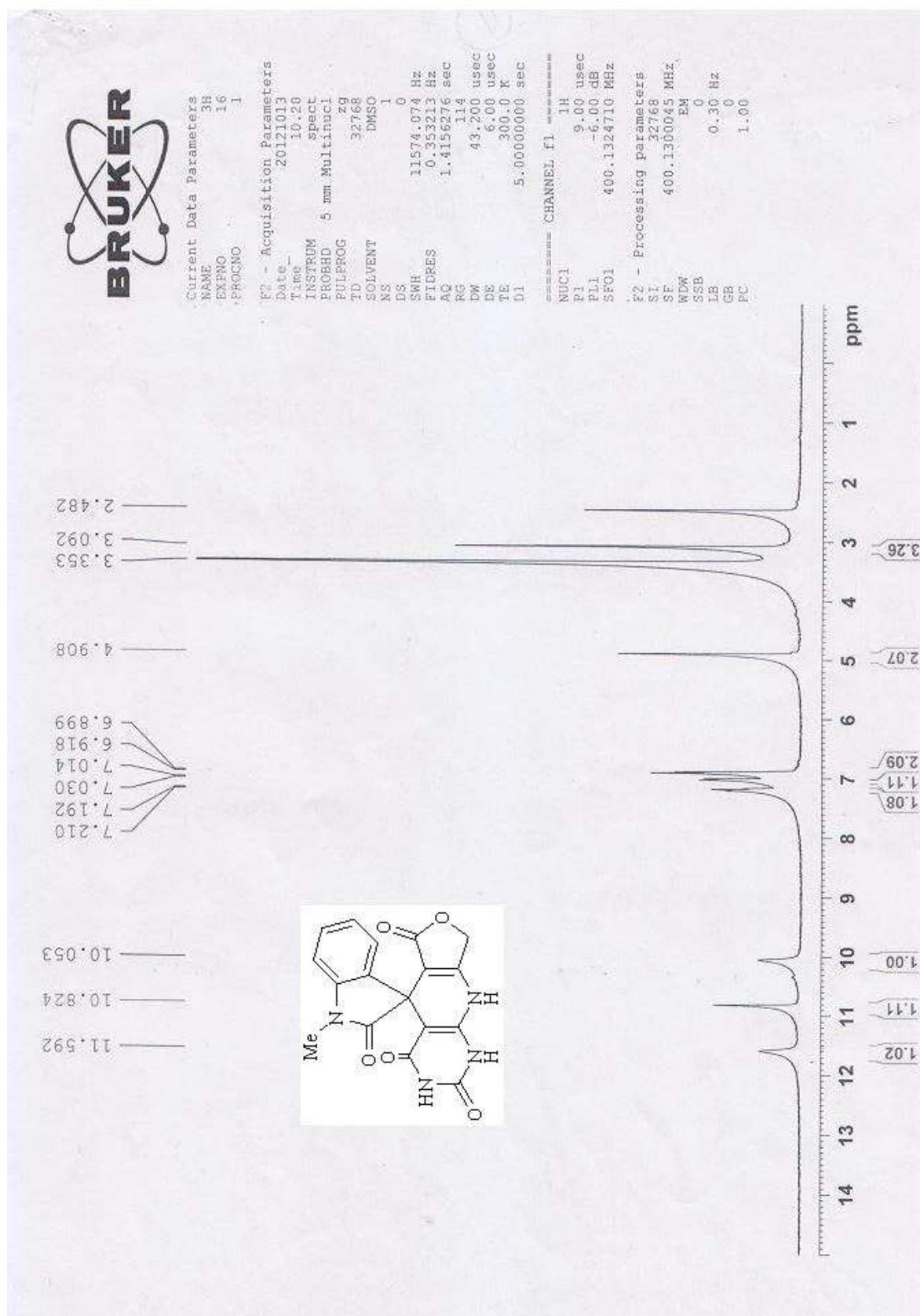
¹³C NMR spectrum of 2-amino-1'-methyl-3H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2',4,6(8H,9H)-trione (4e):



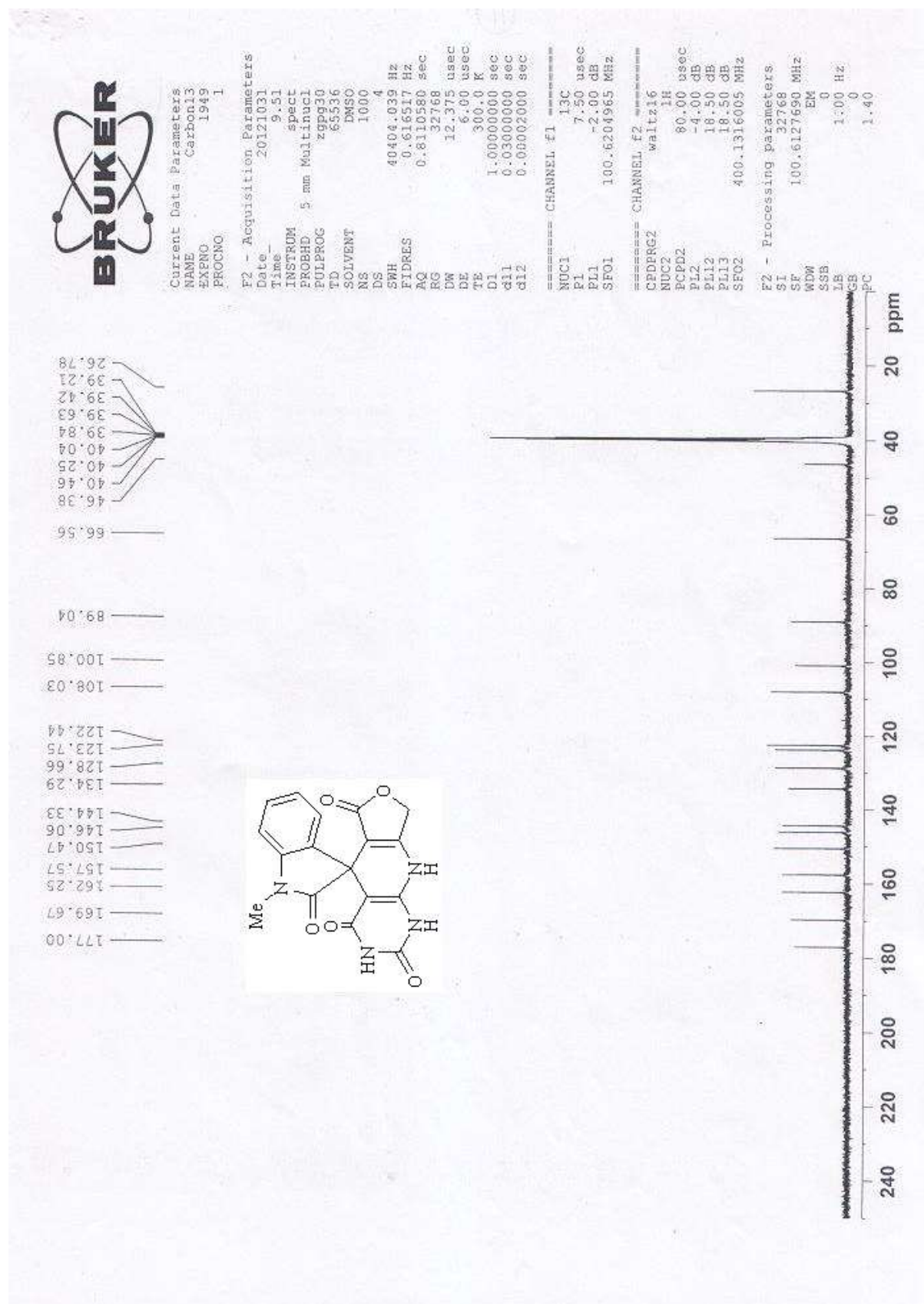
¹H NMR spectrum of 1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4f):



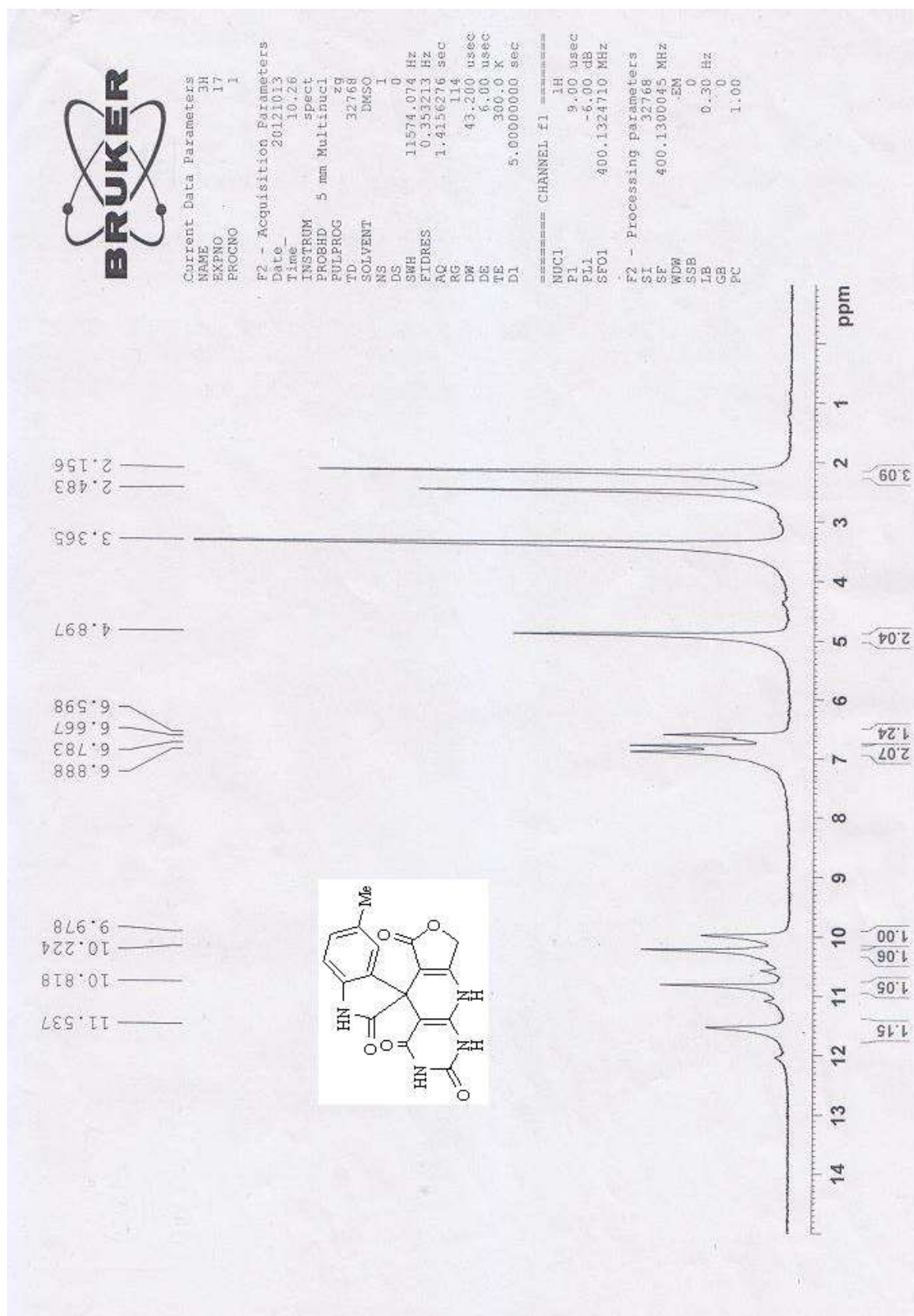
¹³C NMR spectrum of 1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4f):



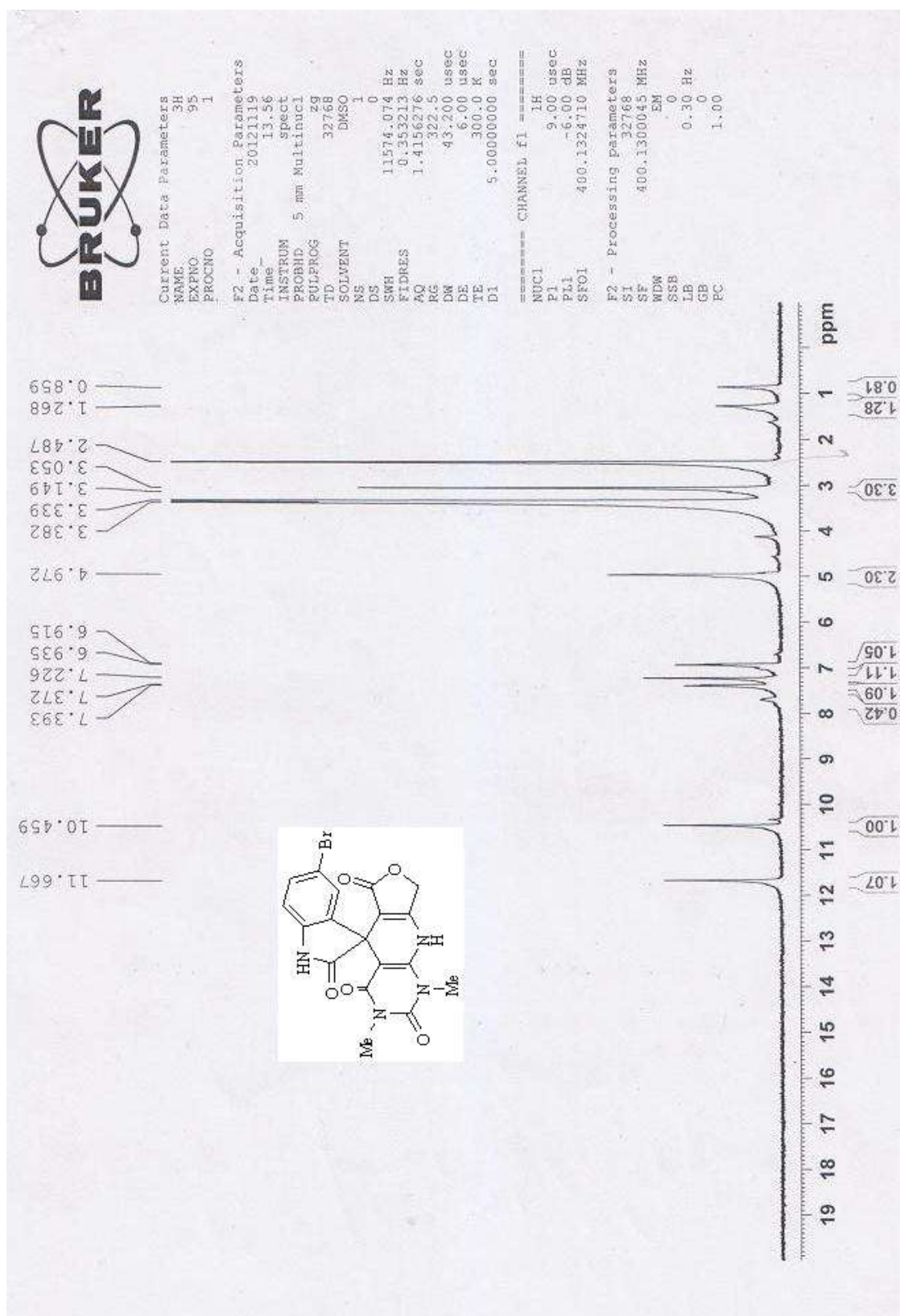
¹H NMR spectrum of 1'-methyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6 (3H,8H,9H)-tetraone (4g):



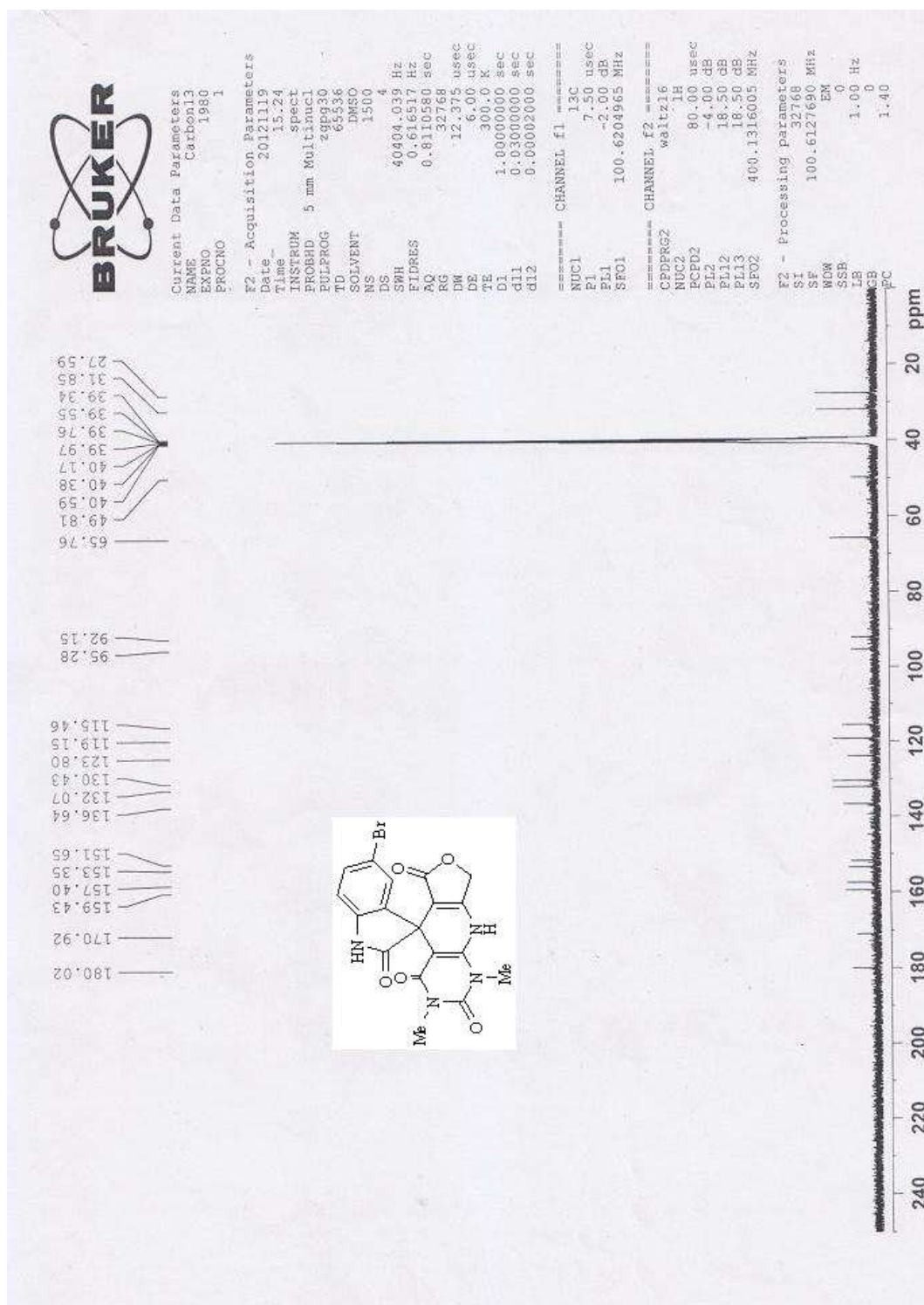
¹³C NMR spectrum of 1'-methyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6 (3H,8H,9H)-tetraone (4g):



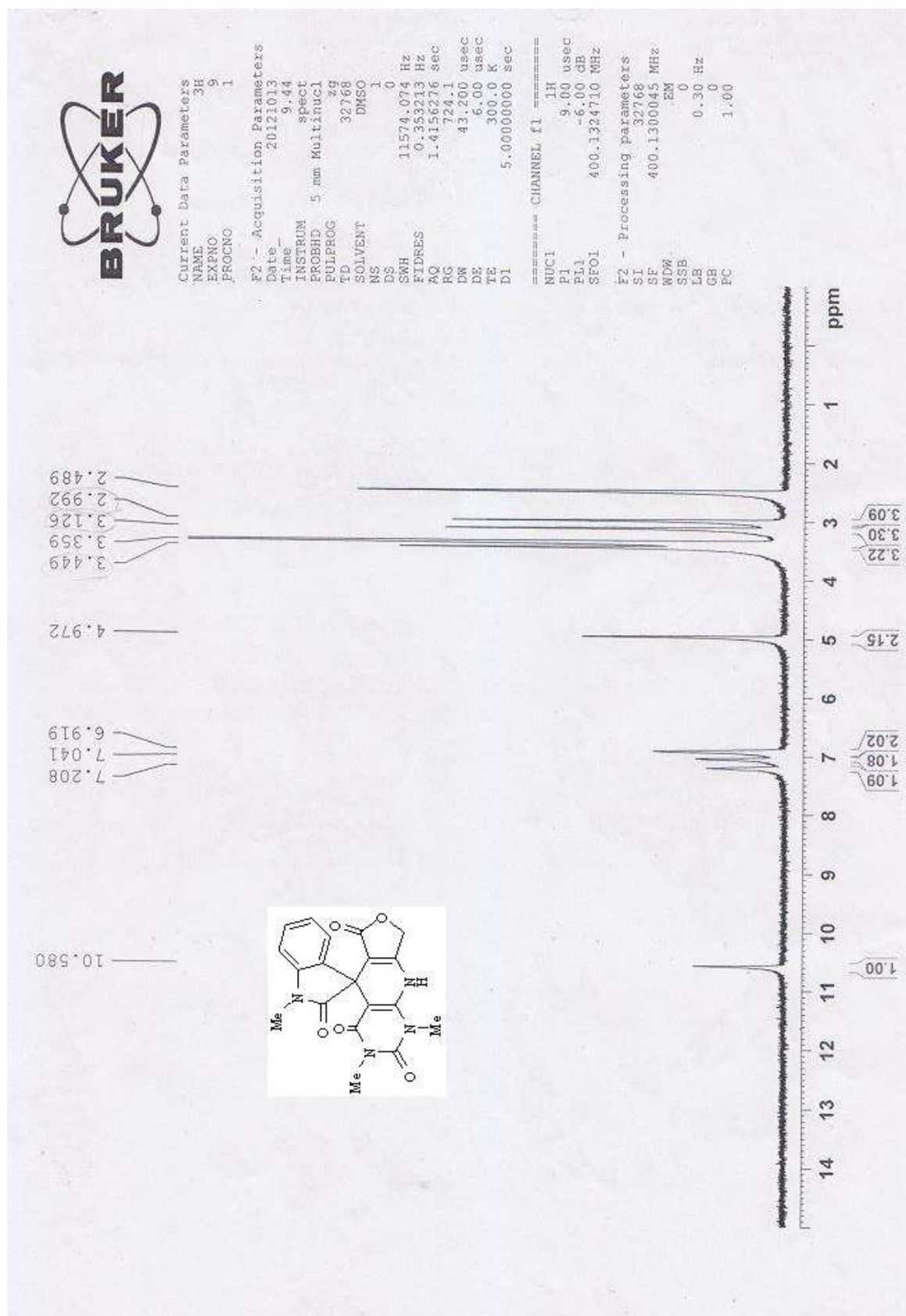
¹H NMR spectrum of 5'-methyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6 (3H,8H,9H)-tetraone (4h):



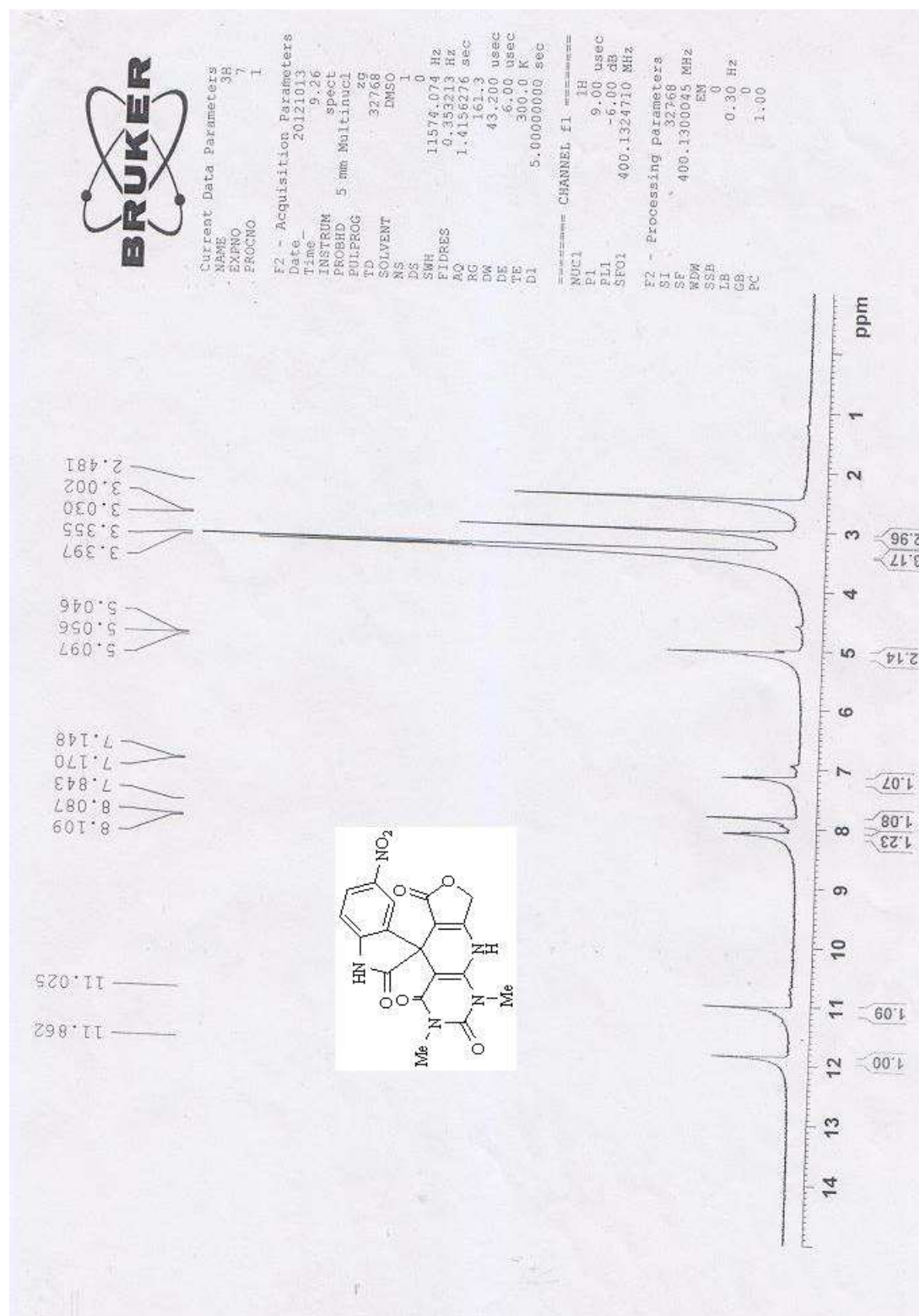
¹H NMR spectrum of 5'-bromo-1,3-dimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4i):



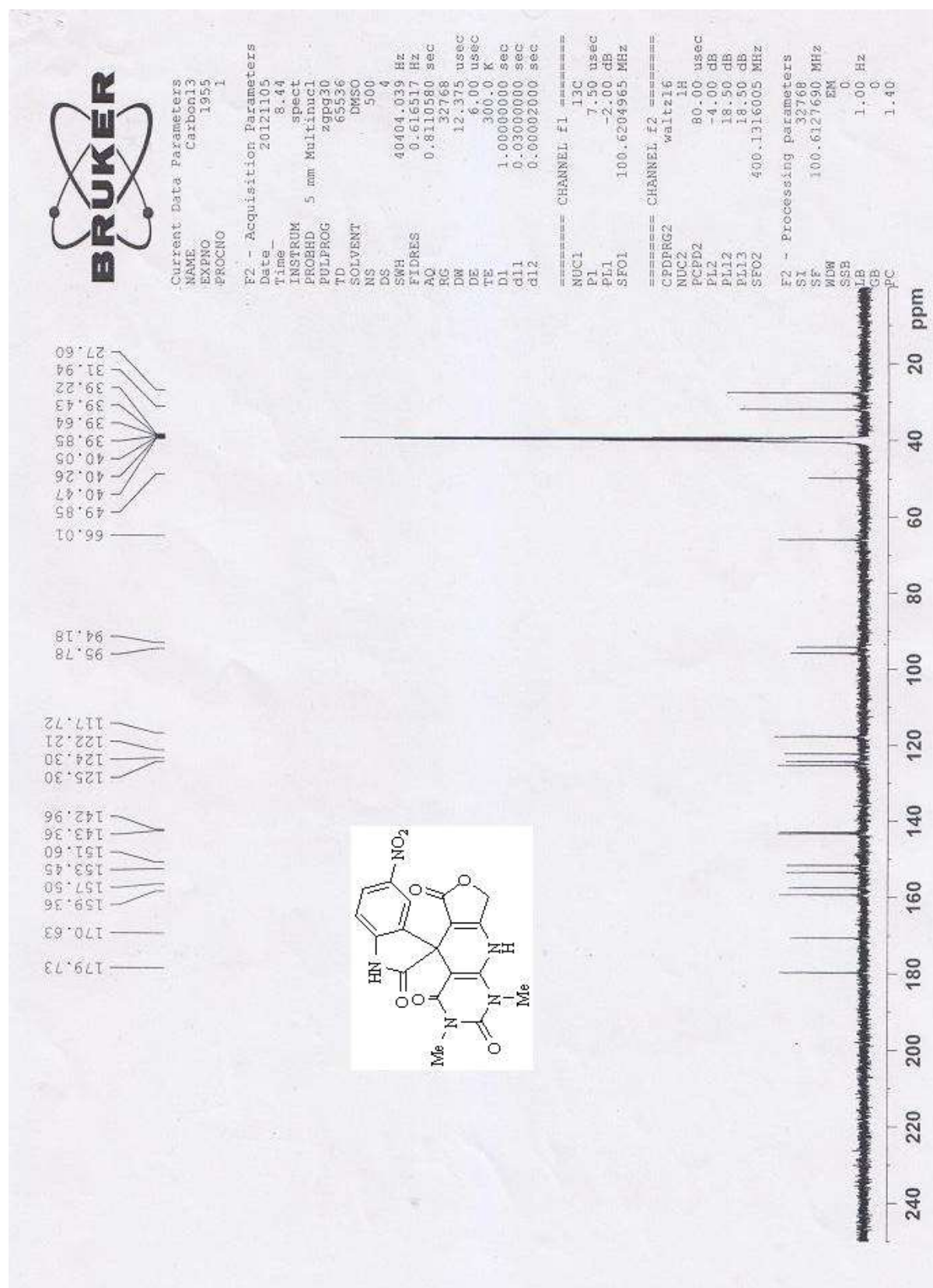
¹³C NMR spectrum of 5'-bromo-1,3-dimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6(3H,8H,9H)-tetraone (4i):



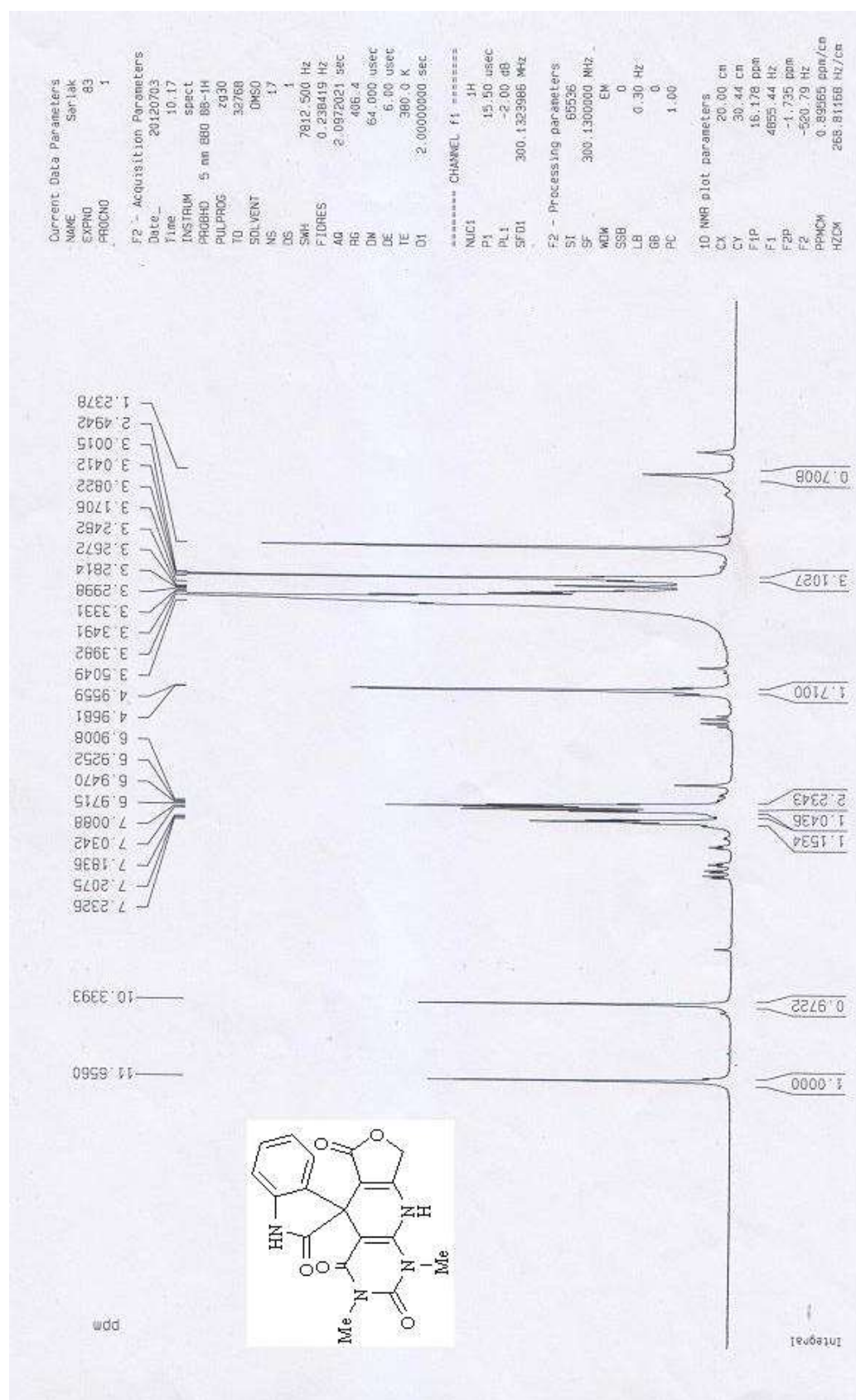
¹H NMR spectrum of 1,1',3-trimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline] 2,2',4,6(3H,8H,9H)-tetraone (4j):



¹H NMR spectrum of 1,3-dimethyl-5'-nitro-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline] 2,2',4,6(3H,8H,9H)-tetraone (4k):



¹³C NMR spectrum of 1,3-dimethyl-5'-nitro-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline] 2,2',4,6(3H,8H,9H)-tetraone (4k):



¹H NMR of spectrum 1,3-dimethyl-1H-spiro[furo[3,4-g]pyrido[2,3-d]pyrimidine-5,3'-indoline]-2,2',4,6 (3H,8H,9H)-tetraone (4I):