## Supporting Information

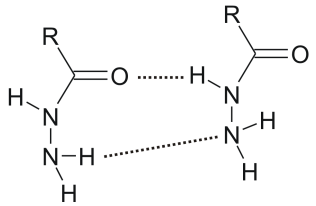
**Table S1.** Assignments of graph sets of carbohydrazide containing molecules in the Cambridge Structural Database.

| CSD Ref Code | Chemical Name | Graph Set |
|---|---|---|
| 1) BANFIG | 3-nitrobenzoic acid hydrazide | $R_2^2(10)$, C(3) |
| 2) BIZMAZ | bis(isonicotinic acid hydrazide).(2,2'-dithiodibenzoic acid) | $R_2^2(6)$, $R_2^2(4)$, $R_4^4(18)$ **t**, |
| 3) CBOHAZ04 | dicarbohydrazide | $R_2^2(6)$, $R_2^2(8)$ C12(4) |
| 4) CEDREJ | o-aminobenzoylhydrazide | $R_3^2(10)$ |
| 5) CYACHZO1 | alpha-cyanoacetohydrazide | $R_4^2(10)$ **t**, $R_4^4(18)$ **t** |
| 6) EDENIL | (E)-3-(4methoxyphenyl)acrylohydrazide | $R_2^2(10)$, $R_3^3(10)$ |
| 7) EDEVAL | 3-(3,4,5-trimethoxyphenyl)propanohydrazide | $R_4^2(10)$ **t**, $R_5^6(22)$ **h** |
| 8) EDEVEP | 3,4-dimethoxybenzohydrazide | $R_4^2(10)$ **t**, $R_4^4(18)$ **t** |
| 9) GENZEF | Pyrazine-2-carbohydrazide | $R_2^2(10)$ |
| 10) GINNEX | 2-bromo-5-methoxybenzohydrazide | C(4) |
| 11) GIRYEM | Nicotinohydrazide | $R_2^2(10)$ |
| 12) HIRBAM | 3-hydroxy-4-methoxybenzohydrazide | $R_2^2(8)$ NN not symmetrical |
| 13) HUMHUS | 4-chlorobenzohydrazide | $R_2^2(10)$, C(3) |
| 14) INICAC01 | Isonicotinic acid hydrazide | C(3), |
| 15) JEYTAJ | 3,5-diflourobenzohydrazide | $R_2^2(10)$, C(3) |
| 16) KEQYAH | 5-Flouro-1H-indole2-carbohydrazide | $R_2^2(10)$ |
| 17) KERWIO | Pyridine-2,6-dicarbohydrazide | $R_2^2(10)$, $R_2^2(8)$ NN not symmetrical |
| 18) LECKUA | 5-Methyl-3,4-diphenyl-1H-pyrrole-2-carbohydrazide | $R_2^2(6)$ |
| 19) MEHMES | 3,5-Dimethoxybenzohydrazide | $R_2^2(10)$, C(4) |
| 20) MESCZB | S-Methylthiocarbazate | $R_2^2(8)$, $R_4^2(10)$ **t** |
| 21) MFRCAH10 | 3-Methyl-4-furoxancarbohydrazide | $R_2^2(6)$, $R_2^2(10)$ |
| 22) MFURCH10 | 4-Methyl-3-furoxancarbohydrazide | $R_2^2(6)$, $R_2^2(10)$ |
| 23) MUGMEG | 1,1'-bis(Hydrazinopropanoyl)-2,2'-biimidazole | $R_3^3(10)$ |
| 24) NAPDEN | 1-Acetylhydrazino-3-hydrazinocarbonyl-4,6-dimethoxybenze | $R_3^3(10)$ |
| 25) NIQPIN | 2-(3-Methoxyphenyl)acetohydrazide | $R_2^2(10)$, C(4) |
| 26) NIQPOT | 3-Hydroxybenzohydrazide | $R_2^2(10)$, $R_4^2(10)$ **t** |

| | | |
|---|---|---|
| 27) NONACH | n-Nonanoic acid hydrazide | $R_3^3(10)$ |
| 28) PAZDAW | 3-(3,5-Di-t-butyl-4-hydroxyphenyl)propionohydrazide | $R_2^2(6)$, $R_2^2(10)$ |
| 29) QETNUZ | N-2-Bromobenzoylhydrazide | $R_3^3(10)$ |
| 30) RATMEF | Formylhydrazine | $R_2^1(5)$ |
| 31) RIZCAF | 2,2'-(Biphenyl)-2,2'-diyldioxy)diacetohydrazide | $R_2^2(6)$, $R_2^2(10)$, $R_4^2(8)$ **t** |
| 32) SOQRUL | 6-(t-Butyldimethylsilyl)-1-carbazoyl-5-dimethylaminosulfonyl-2-phenylcarbamoylimidazo(4,5-c)pyridazine | $R_2^2(6)$, $R_2^2(10)$ |
| 33) TICDUF | 1-(2,4-dihydroxyphenyl)ethanone-((1H-indol-3-yl)acetyl)hydrazone 2-(1H-indol-3-yl)acetohydrazide | $R_3^3(10)$ |
| 34) UFAPAT | 3-(4-Mthoxyphenyl)propanohydrazide | $R_3^3(10)$ |
| 35) VAWGAB01 | (Z)-2-Hydroxybenzohydrazide | $R_2^2(6)$ |
| 36) VIPKIO01 | trans,trans-Oxalyldihydrazide | $R_2^2(8)$ NN not symmetrical, $R_4^4(14)$ |
| 37) VIPKIO03 | trans,trans-Oxalyldihydrazide | $R_3^3(10)$ |
| 38) VIPKIO04 | trans,trans-Oxalyldihydrazide | $R_3^3(10)$ |
| 39) VIPKIO05 | trans,trans-Oxalyldihydrazide | $R_2^2(10)$, C(3) |
| 40) VOPJEP | Benzhydrazide | $R_2^2(10)$, C(3) |
| 41) WADCUA | 3-Carbazoyl-5-methylpyridazin -6(1H)-one | $R_2^2(10)$ |
| 42) WEPVIX | 2,6-Dichlorobenzohydrazide | C(4) |
| 43) WEPVOD | 2,4-Dichlorobenzohydrazide | $R_2^2(6)$, $R_2^2(10)$ |
| 44) WERHUX01 | N-Picolnoylhydrazide | $R_2^2(10)$ |
| 45) XEZBAG | 2-Methylpropionohydrazide | $R_2^2(10)$ $R_4^2(10)$ **t** |
| 46) YIFSOW | 2-(2,4-Diclorophenylsulfanyl)acetohydazide | $R_2^2(6)$, $R_2^2(10)$ |
| 47) YIZRAB | 4-Bromobenzohydrazide | $R_2^2(10)$, C(3) |

Footnote

[a] "$R_2^2(8)$ NN not symmetrical" refers to the following hydrogen bond ring:




[b] **t** stands for a ring made up of four molecules (tetramer); **h** stands for six molecules (hexamer).

Supplementary Data for **1**

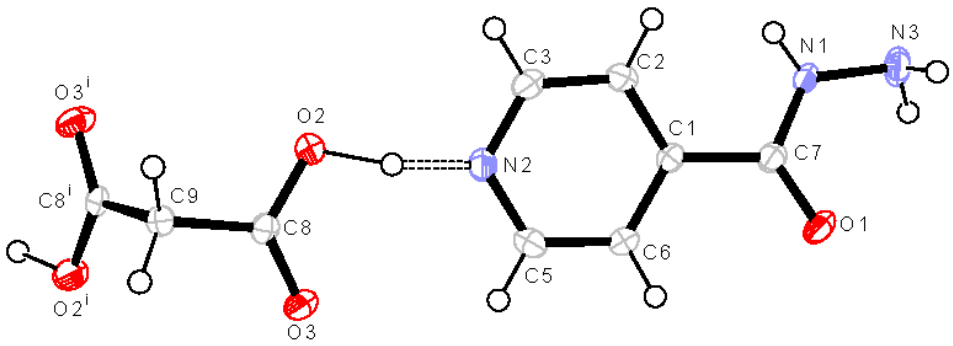



**Figure S1**. The asymmetric unit of **1** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown. Atoms labelled with (i) are at symmetry position (-x, 1-y, z).

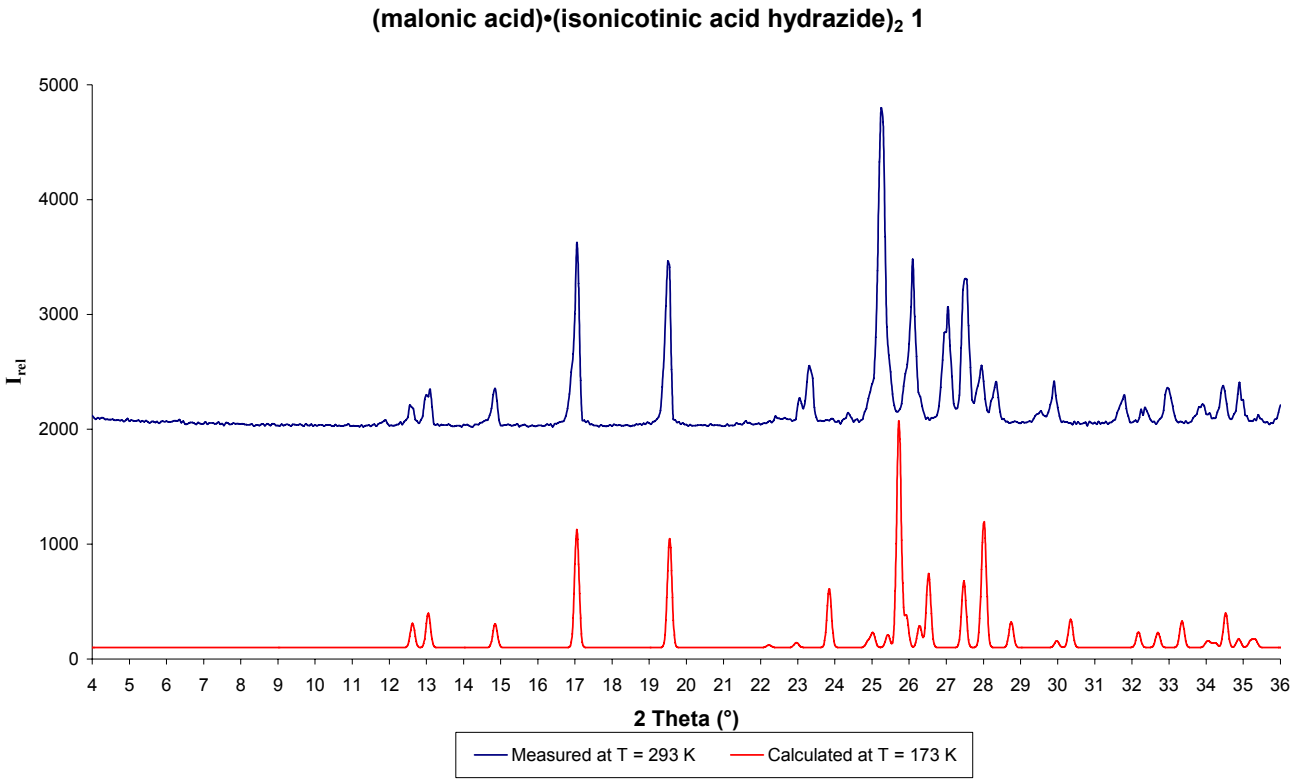



**Figure S2**. Comparative calculated and measured PXRD pattern for **1**.

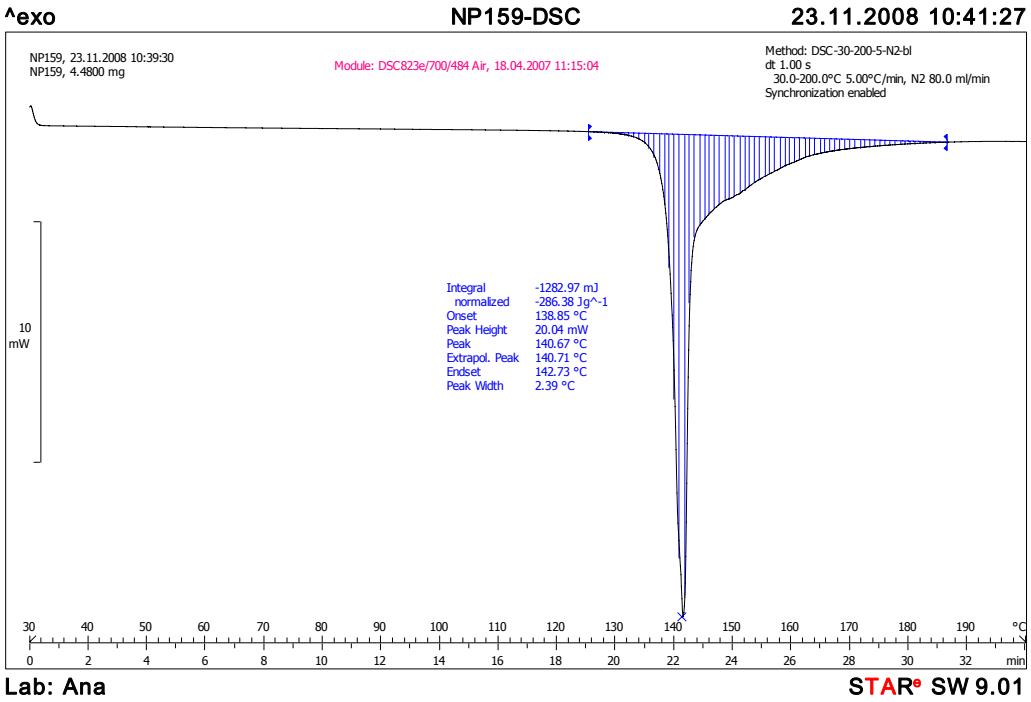



**Figure S3**. DSC trace for **1**.

Supplementary Data for **2**

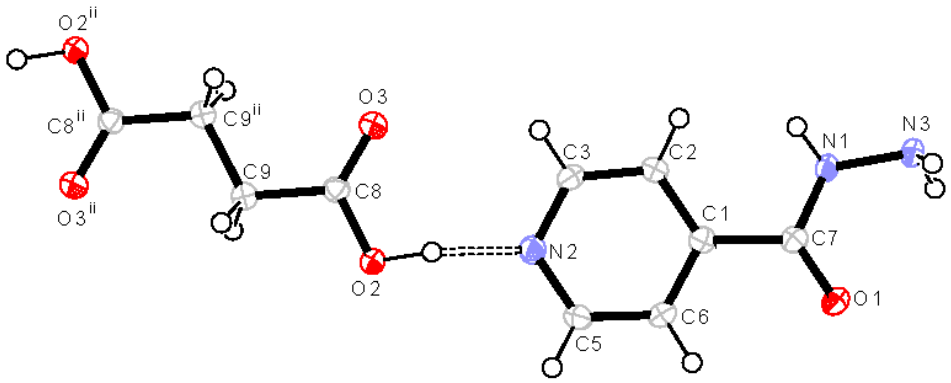



**Figure S4**. The asymmetric unit of **2** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown. Atoms labelled with (ii) are at symmetry position (-x, -y, 1-z).

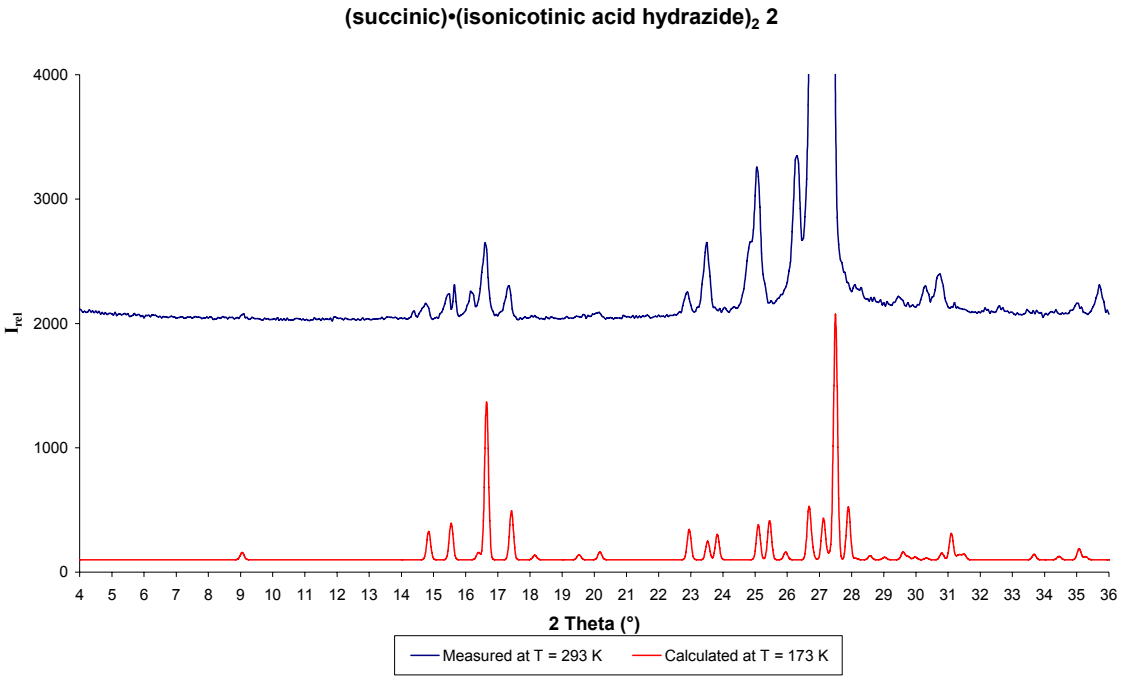



**Figure S5**. Comparative calculated and measured PXRD pattern for **2**.

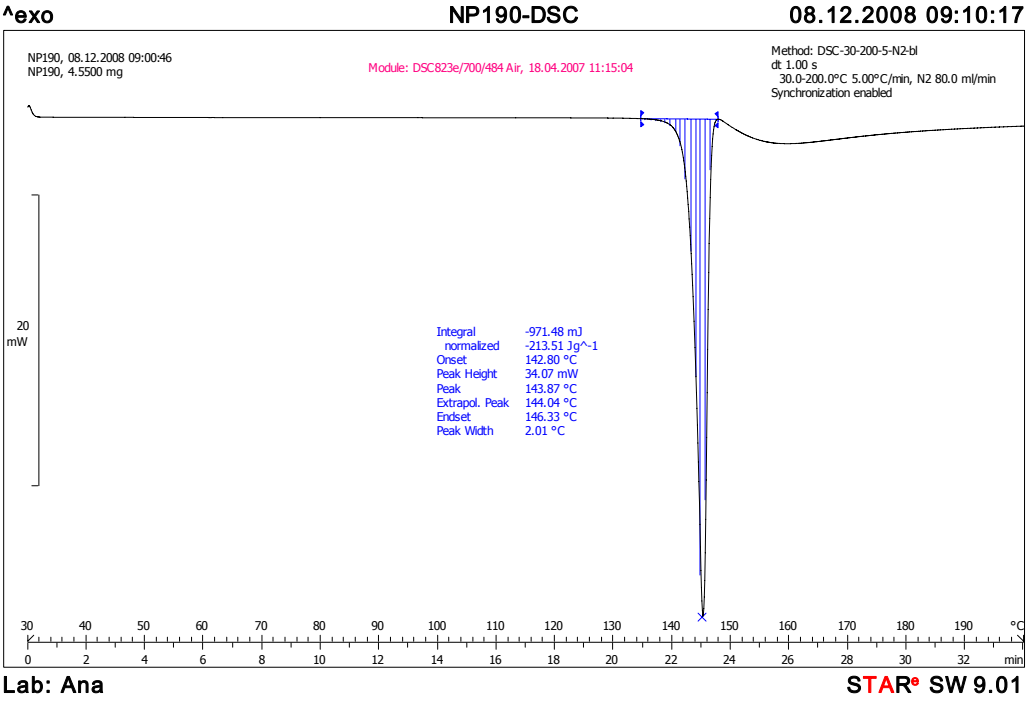



**Figure S6**. DSC trace for **2**.

Supplementary Data for **3**

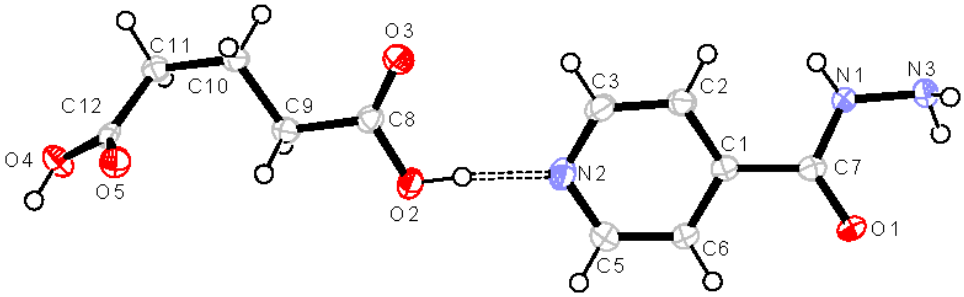



**Figure S7**. The asymmetric unit of **3** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

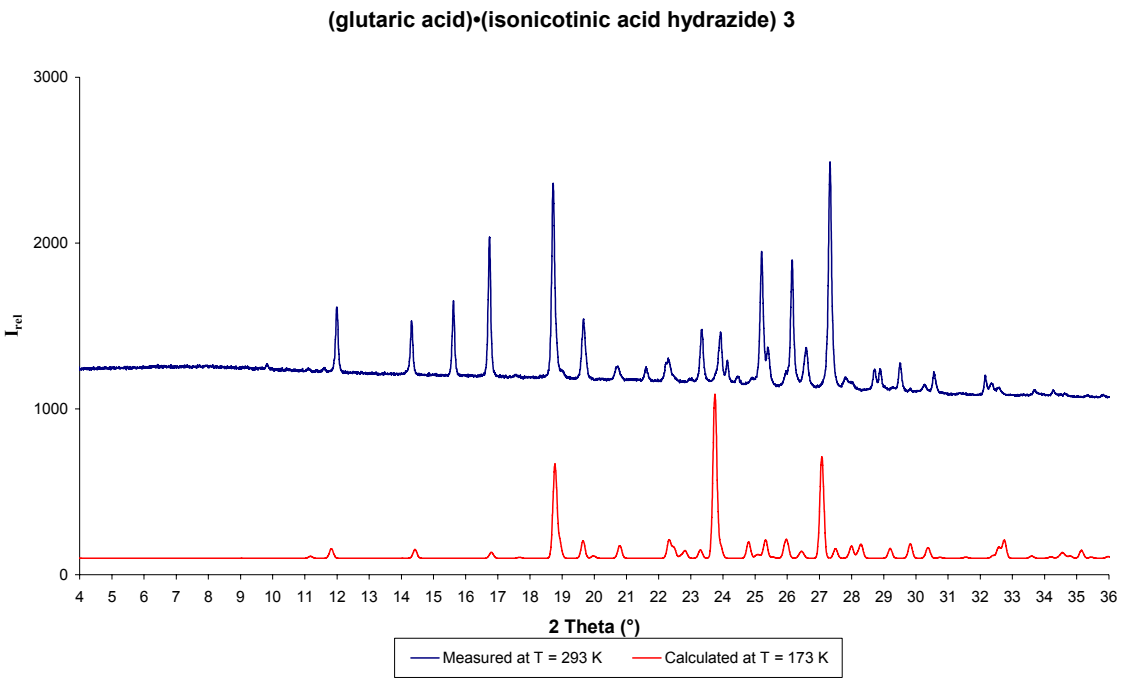



**Figure S8**. Comparative calculated and measured PXRD pattern for **3**.

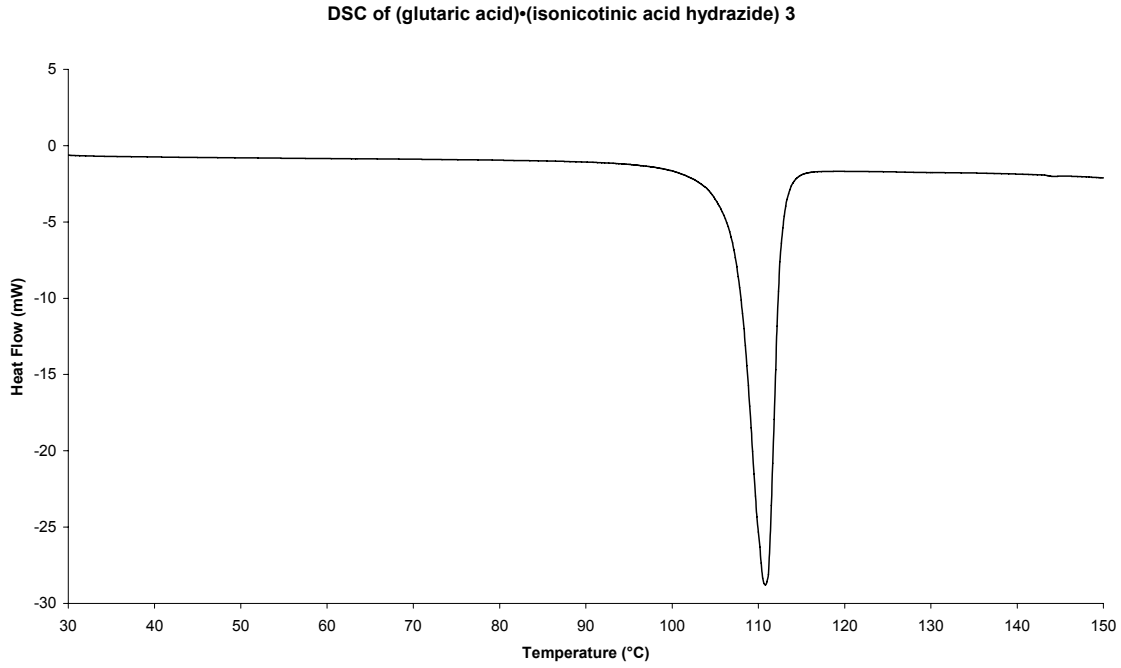



**Figure S9**. DSC trace for3**1**.

Supplementary Data for **4**

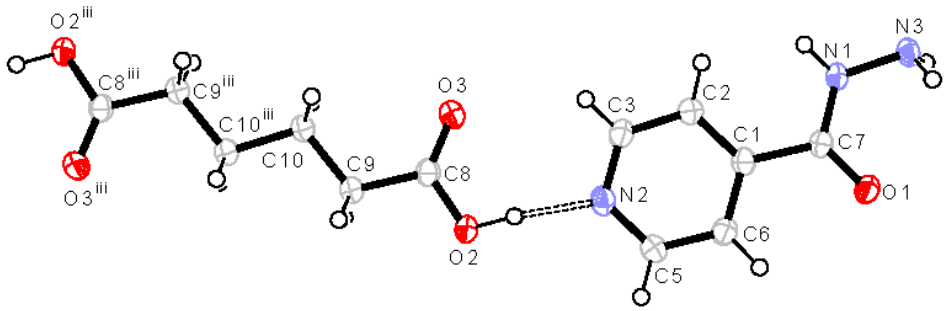



**Figure S10**. The asymmetric unit of **4** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown. Atoms labelled with (iii) are at symmetry position (-x, 2-y, -z).

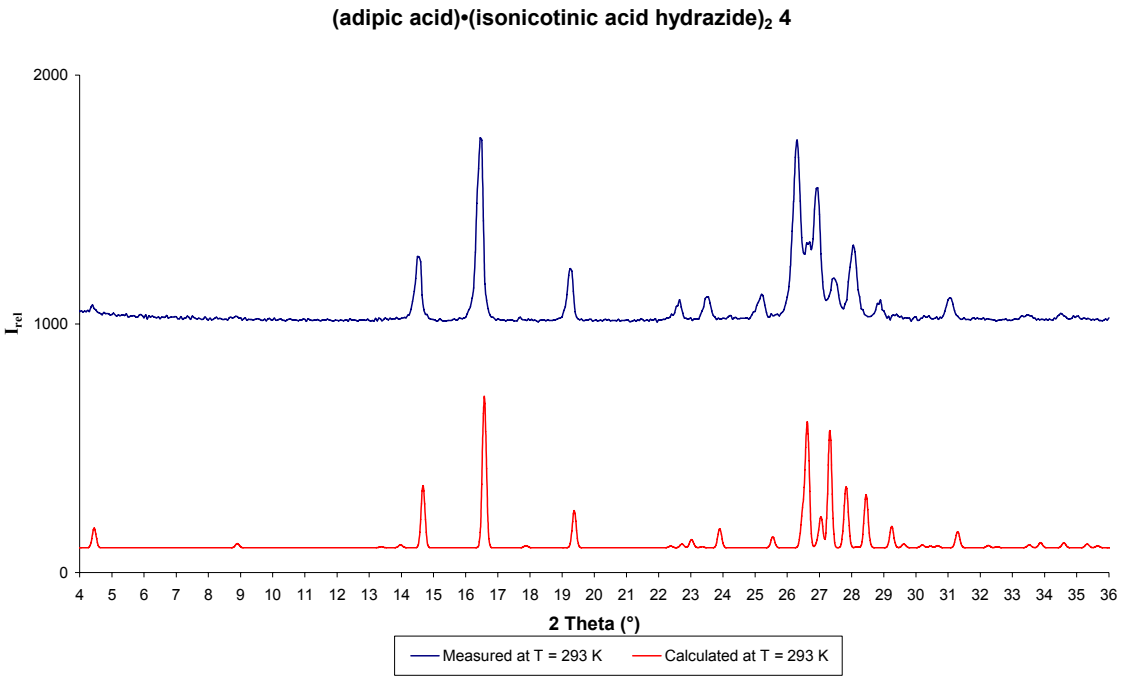



**Figure S11**. Comparative calculated and measured PXRD pattern for **4**.

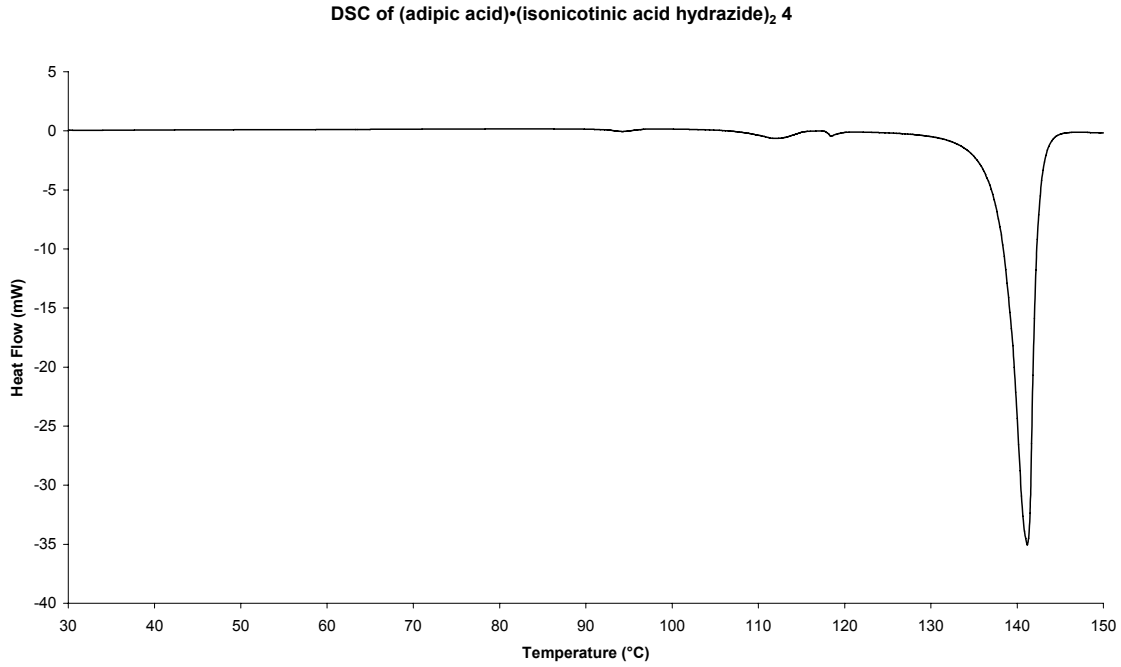



**Figure S12**. DSC trace for **4**.

Supplementary Data for **5**

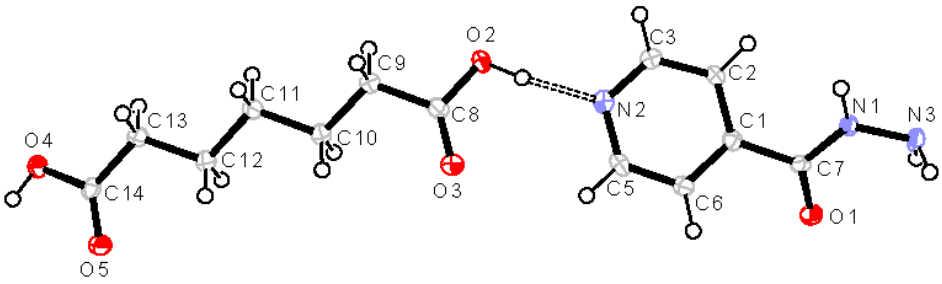



**Figure S13**. The asymmetric unit of **5** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

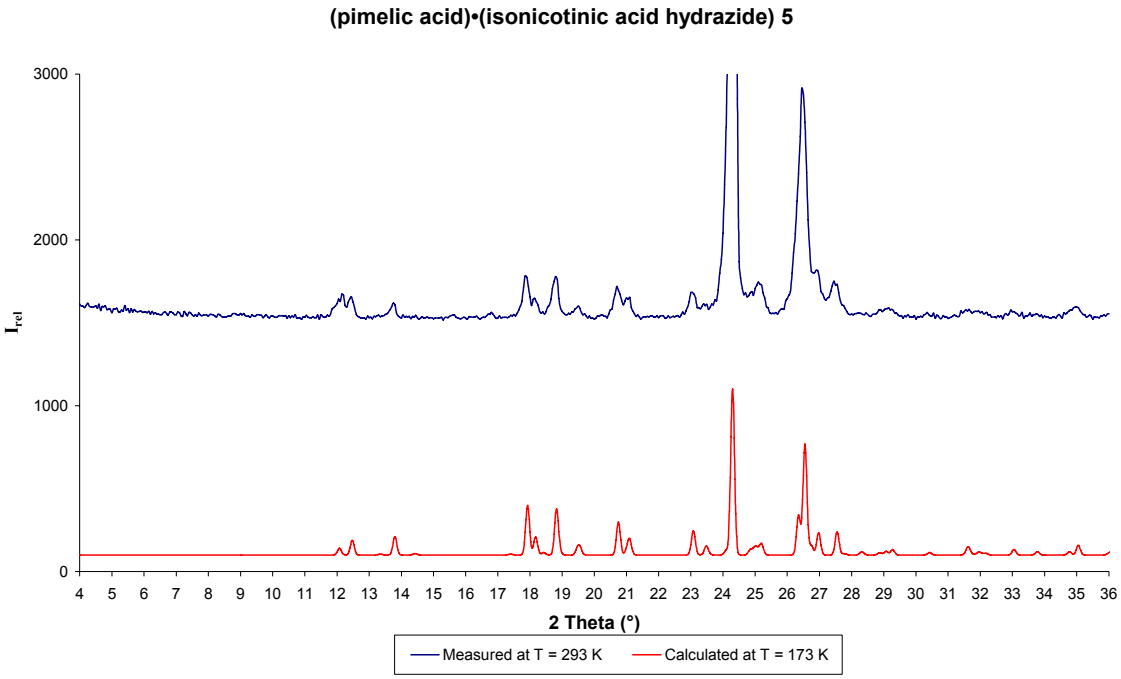



**Figure S14**. Comparative calculated and measured PXRD pattern for **5**.

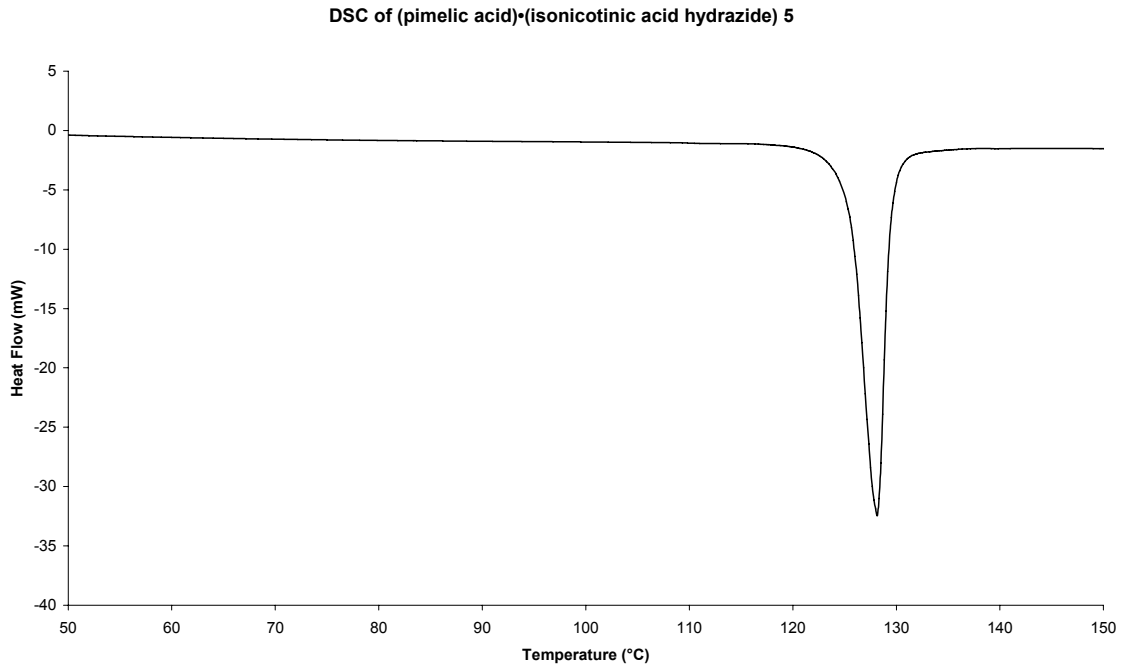



**Figure S15**. DSC trace for **5**.

Supplementary Data for **6**

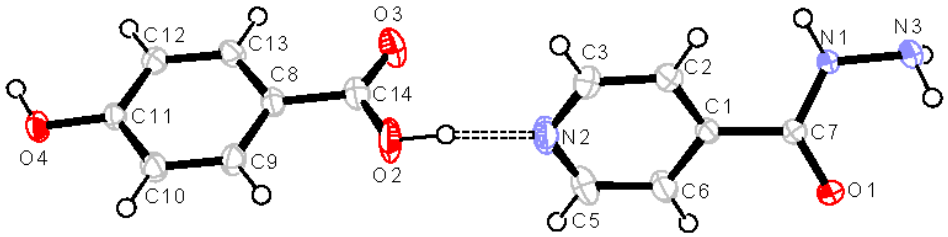



**Figure S16**. The asymmetric unit of **7** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

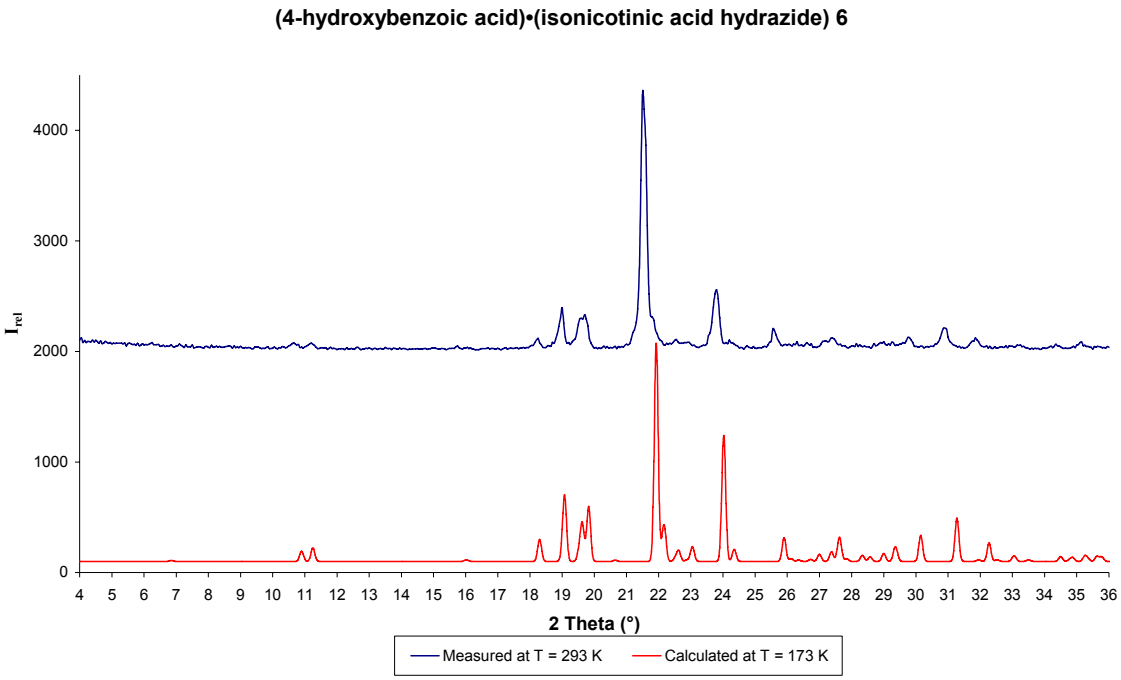



**Figure S17**. Comparative calculated and measured PXRD pattern for **6**.

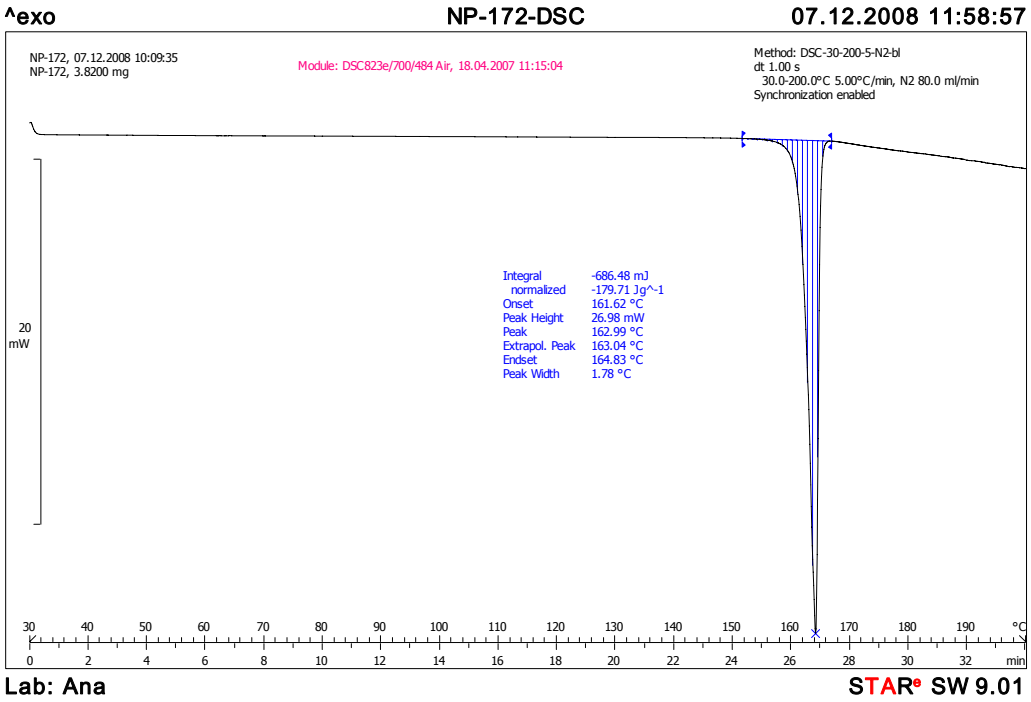



**Figure S18**. DSC trace for **6**.

Supplementary Data for **7**

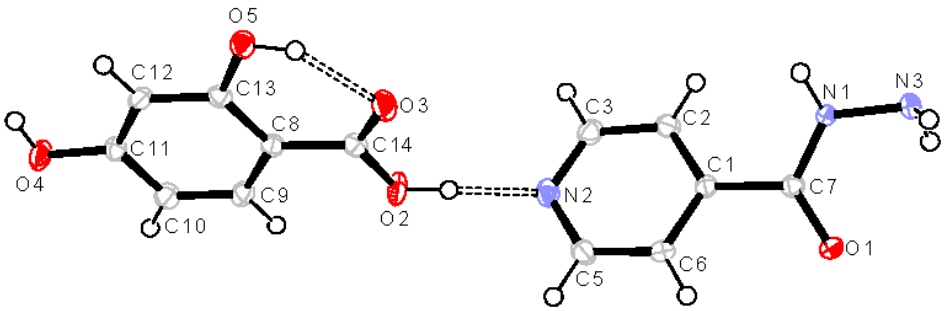



**Figure S19**. The asymmetric unit of **7** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

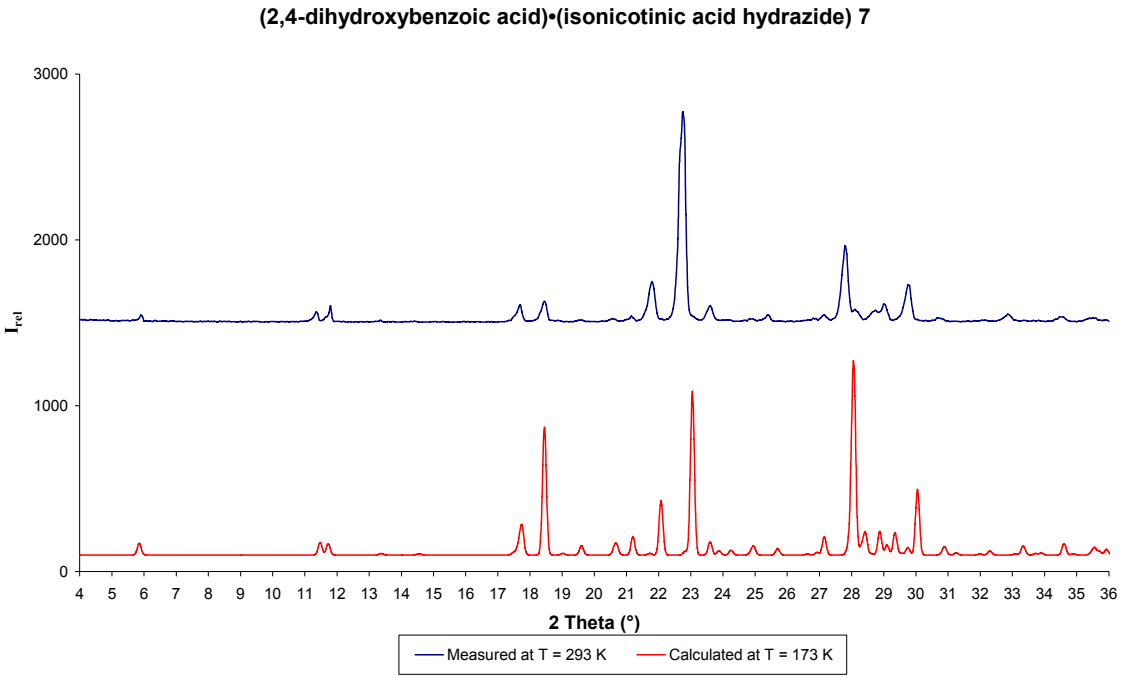



**Figure S20**. Comparative calculated and measured PXRD pattern for **7**.

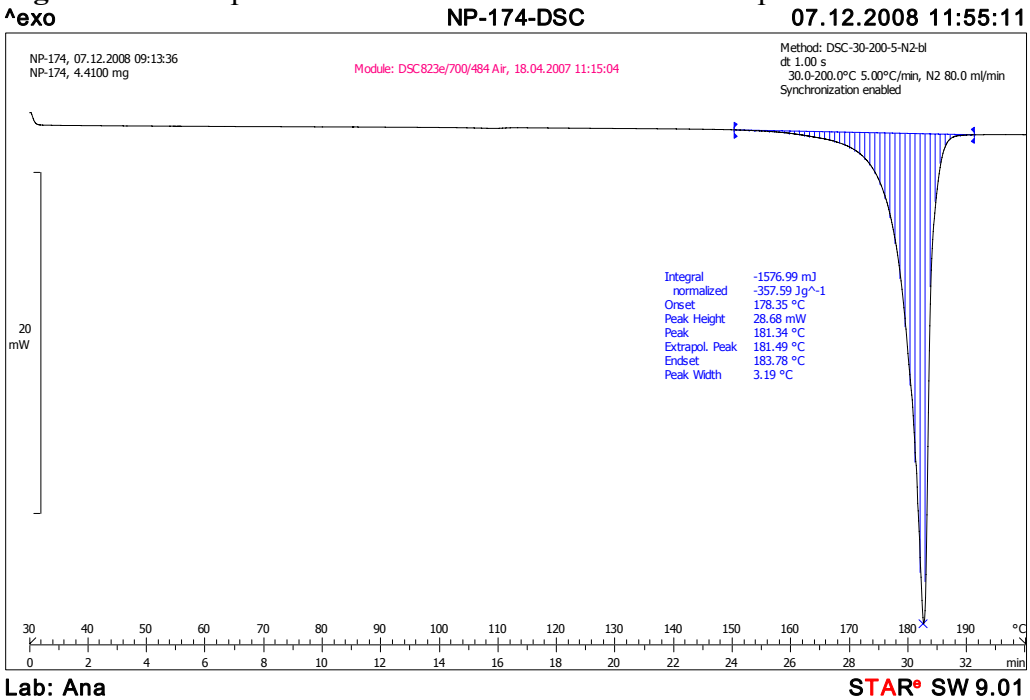



**Figure S21**. DSC trace for **7**.

Supplementary Data for **8**

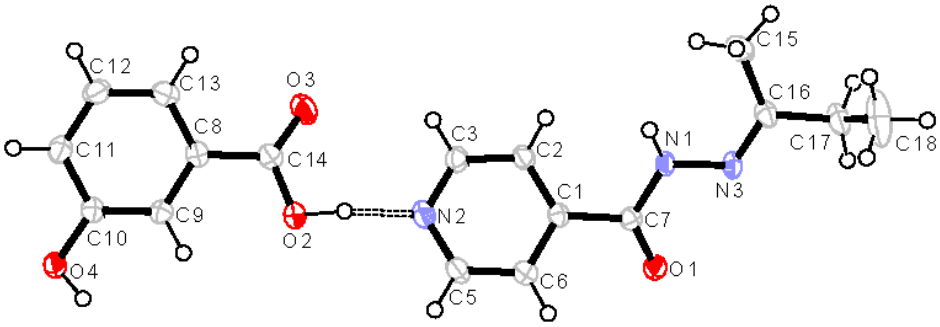



**Figure S22**. The asymmetric unit of **8** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

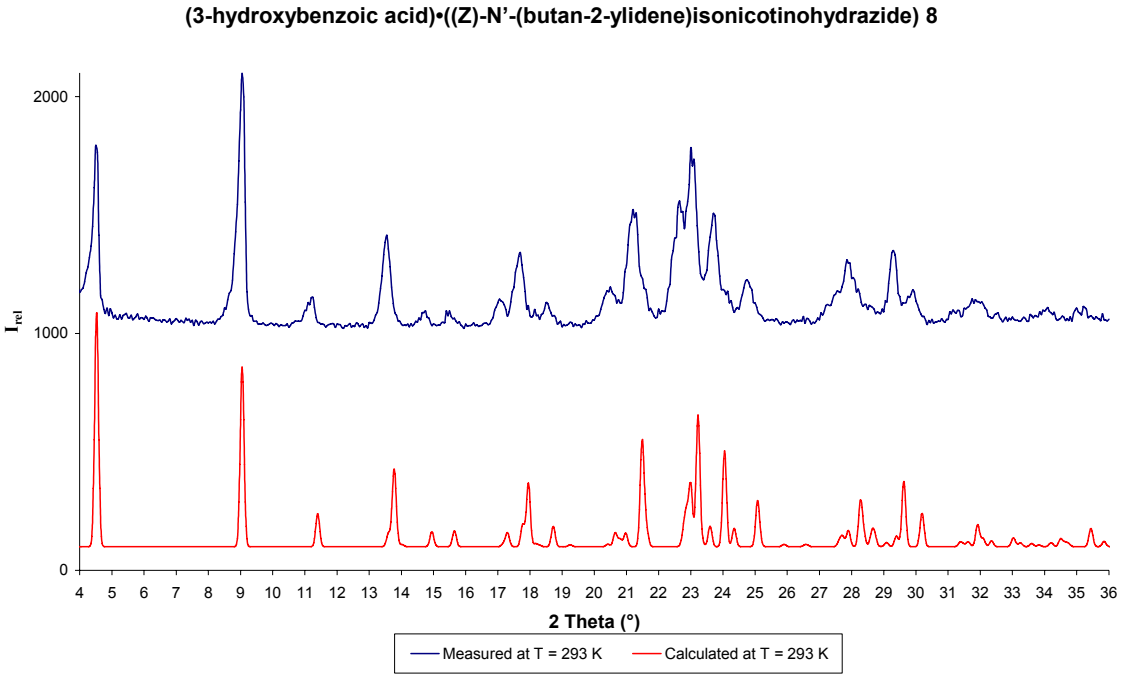



**Figure S23**. Comparative calculated and measured PXRD pattern for **8**.

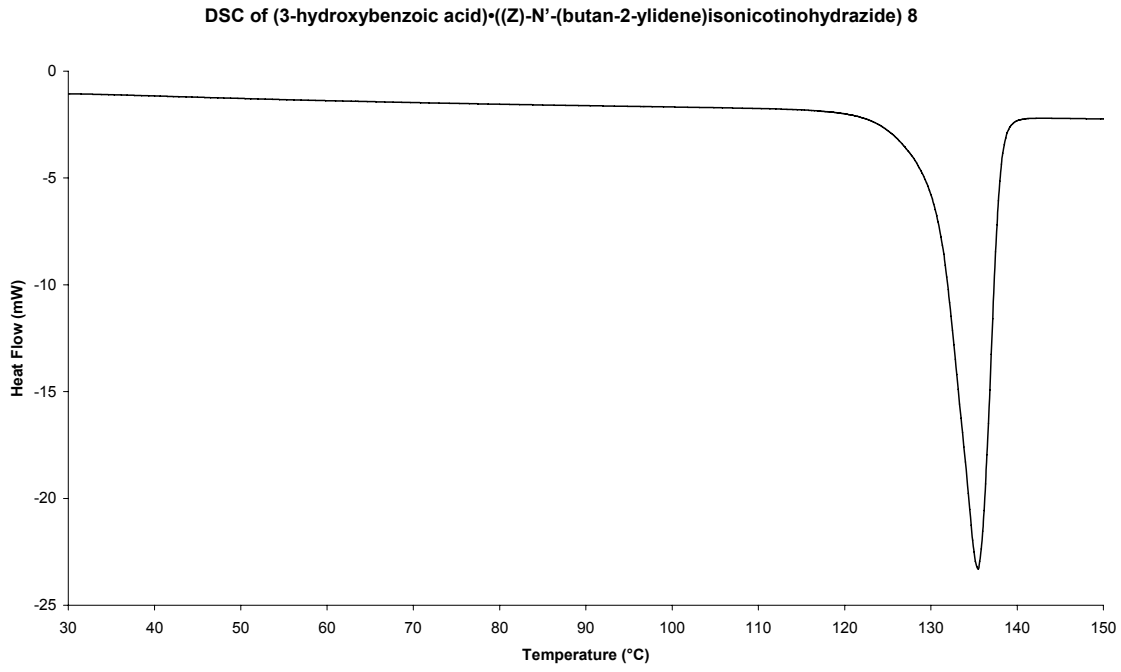



**Figure S24**. DSC trace for **8**.

Supplementary Data for **9**

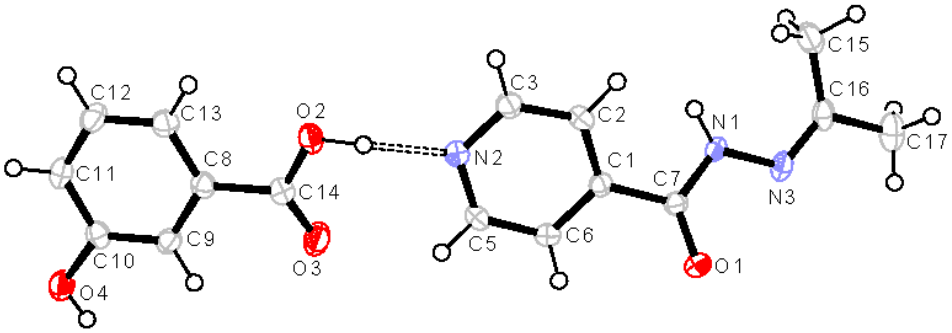



**Figure S25**. The asymmetric unit of **9** showing the atomic numbering scheme and 50% displacement ellipsoids. Only the symmetry independent hydrogen bonds are shown.

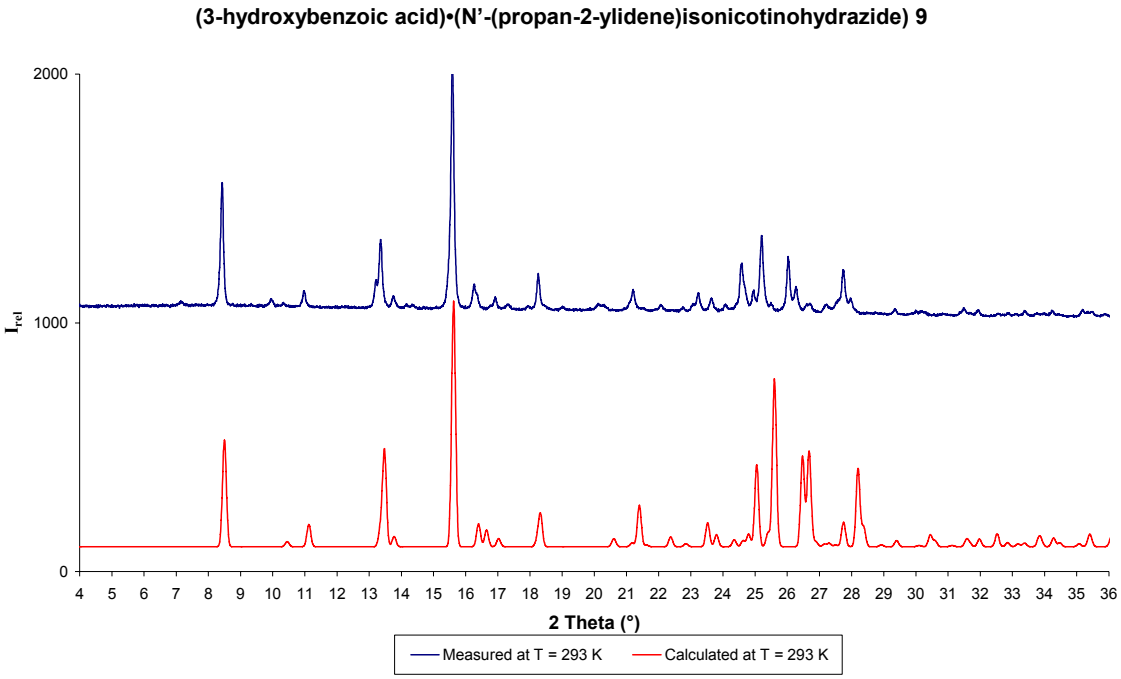



**Figure S26**. Comparative calculated and measured PXRD pattern for **9**.

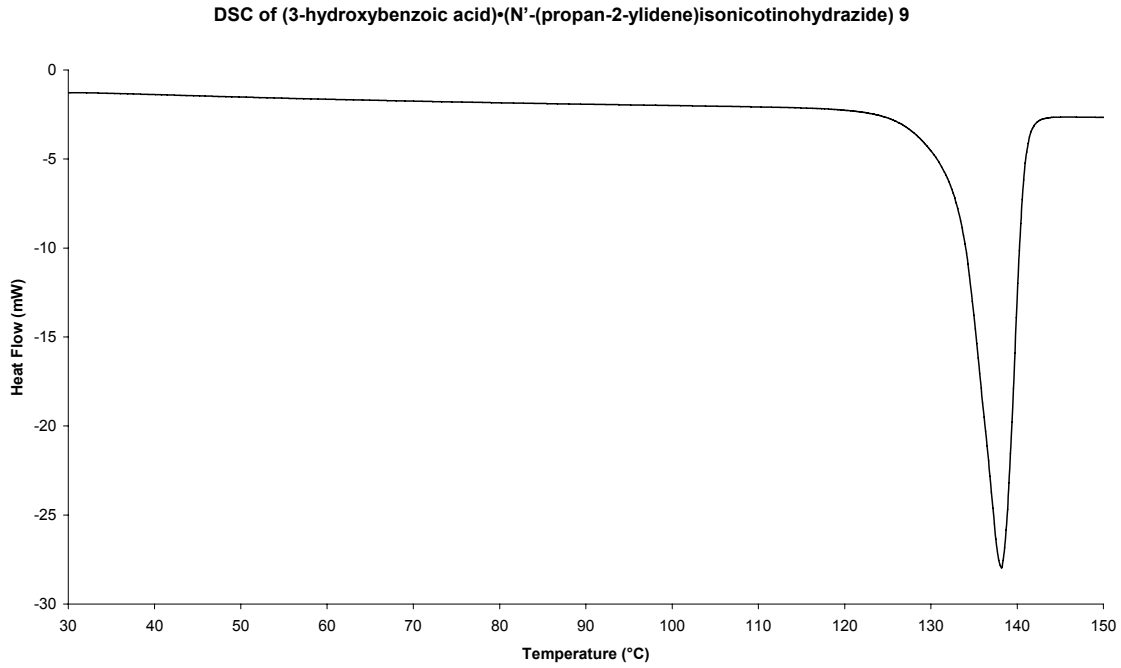



**Figure S27**. DSC trace for **9.**