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Non-covalent synthesis of ionic and molecular complexes of benzoic acid and substituted 2-aminopyrimidines by varying aryl/alkyl *substituents* and their supramolecular chemistry

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The non-covalent synthesis of ionic and molecular complexes of substituted 2-aminopyrimidines with benzoic acid in crystalline solid phase is reported. The nature of supramolecular architecture varied with the substituents in the 2-aminopyrimidine ring. Two complexes have been synthesised from two differently substituted 2-aminopyrimidines and benzoic acid. In one case, proton transfer takes place and an ionic organic salt is formed, whereas in the other, there is no proton transfer and a hydrogen-bonded molecular complex is formed.

Keywords: Molecular recognition; Supramolecular chemistry; Crystal engineering; Host-guest

Recognition of different guest substrates in the solid phase by synthetic receptors or commercially available molecules containing a complementary donor–acceptor array has drawn much attention from the scientific community in last few years [1]. This process of interaction leads to the design of new materials [2] and new carriers for drug delivery which has a great impact in pharmaceutical science [3]. Solid-phase recognition of monocarboxylic acid towards different receptors is very important as many drug molecules contain the carboxylic acid group [4]. Recently, we have observed that the recognition process between the same pair of host–guest is not identical in solution and in solid phases [5]. This difference is mainly due to the various non-bonding weak interactions [6], which play an important role in generating the overall supramolecular architecture of the complexes. Substituted 2-aminopyrimidine (2AP) or simple 2-aminopyrimidine [7] is a very important host for recognising both monocarboxylic [8] and dicarboxylic [9] acids leading to well-defined supramolecular networks in the field of crystal engineering. It also plays a significant role in coordination in metal-containing supramolecular assemblies [10]. 2-Aminopyrimidine has two sets of donor–acceptor array (Fig. 1(a)), which is present in an angular fashion [11]. When molecules containing this unit recognise the carboxylic acid, it forms different and interesting supramolecular networks. Moreover, substituents into the pyrimidine moiety have a great influence towards the whole architecture.

We disclose here the recognition patterns of monocarboxylic acid by differently substituted 2-aminopyrimidines and show how the substituents can change the whole solid-phase arrangement. For this purpose, complexes **1** (Fig. 1(b)) and **2** (Fig. 1(c))

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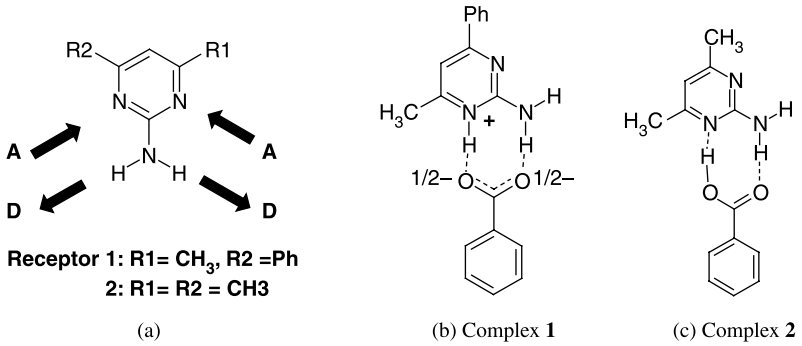
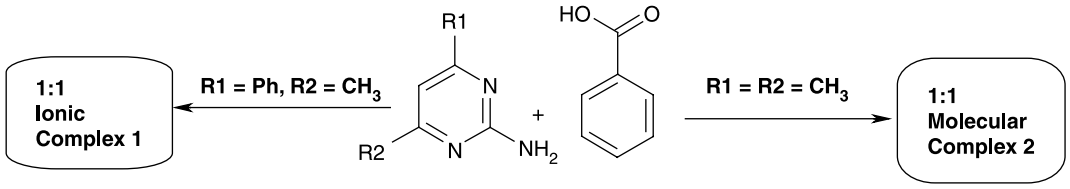
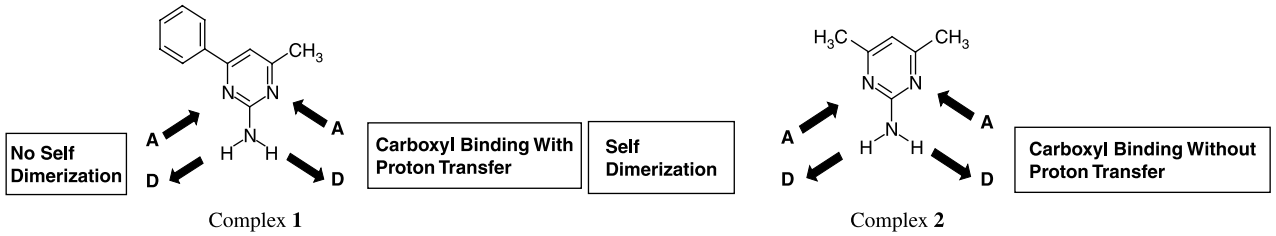


(a) (b) Complex **1** (c) Complex **2**

FIGURE 1 (a) Array of donor–acceptor in 2AP moiety, (b) complex **1** (2-amino-4-methyl-6-phenylpyrimidine and benzoic acid) and (c) complex **2** (2-amino-4,6-dimethylpyrimidine and benzoic acid).



SCHEME 1 Non-covalent synthesis of complexes **1** and **2** using hydrogen bonding.



Complex **1** Complex **2**

SCHEME 2 Design of aryl/alkyl- and dialkyl-substituted pyrimidines for proton transfer and non-proton-transfer in carboxyl acid recognition.

have been synthesised[1] (Scheme 1) from the synthesised substituted pyrimidines [12] in our laboratory to react with the commercially available benzoic acid.

The crystalline 1:1 complex of 2-amino-4-methyl-6-phenylpyrimidine and benzoic acid (complex **1**) in space group *P*-1 has one molecule each in the asymmetric unit. Each 1:1 complex is hydrogen bonded with another complex with an opposite arrangement to form a 2:2 complex. The crystalline complex of 2-amino-4,6-dimethylpyrimidine and benzoic acid (complex **2**) in space group $P2(1)/c$ also has one molecule each in the asymmetric unit. In case of complex **2**, each aminopyrimidine moiety is simultaneously hydrogen bonded with one carboxyl acid moiety of benzoic acid and another amino pyrimidine moiety (self-dimer). In the case of complex **1**, the proton transfer causes the breaking of the self-dimeric hydrogen bonds present in the host 2-aminopyrimidine, but in complex **2**, the self-dimeric pyrimidine hydrogen bonds persist and form dimeric complexes with the guest carboxylic acid (Scheme 2).

[1]Receptor **1**: Mp. 169–71°C; FT-IR (KBr): 3325, 3196, 3059, 2919, 1868, 1636, 1597,1557, 1541, 1352, 1249, 764, 712, 549 cm^{-1}; ^{1}H NMR ($CDCl_3$, 500 MHz): δ (ppm): 7.99–7.96(m, 2H), 7.59(t, 3H, $J = 7.4$ Hz), 6.94(s, 1H), 5.02(bs, 2H), 2.42(s, 3H). Complex **1**: Mp. 106–08°C; FT-IR (KBr): 3365, 3325, 3059, 2919, 1918, 1636, 1597,1557, 1541, 1352, 1249, 764, 712, 549 cm^{-1}; ^{1}H NMR ($CDCl_3$, 500 MHz): δ (ppm): 8.11(d, 2H, $J = 7.8$ Hz), 7.99–7.96(m, 2H), 7.59(t, 1H, $J = 7.4$), 7.48–7.45(m, 5H), 6.93(s, 1H), 5.73(bs, 2H), 2.45(s, 3H); Crystal data are as follows (**CCDC No. 634315**): $C_{18}H_{17}N_3O_2$, *Mr* = 307.35, 100.0(1) K, Triclinic, *P*-1, $a = 8.88970(10)$ Å, $b = 10.13780(10)$ Å, $c = 10.39980(10)$ Å, $\alpha = 61.3550(10)°$, $\beta = 66.5820(10)°$, $\gamma = 71.2800(10)°$, V = 744.822(13) $Å^3$, Z = 2, μ(Mo K_α) = 0.092 mm^{-1}, Reflections collected/unique 34395/7789 [R(int) = 0.0364], R1($I > 2\sigma(I)$) = 0.0479, wR2 = 0.1269, R1(All data) = 0.0613, wR2 = 0.1373. Receptor **2**: Mp. 150–52°C; FT-IR (KBr): 3405, 3312, 3184, 1638, 1597, 1466, 1389, 1241, 794, 552 cm^{-1}; ^{1}H NMR ($CDCl_3$, 500 MHz): δ (ppm): 6.40(s, 1H), 4.86(bs, 2H), 2.29(s, 6H). Complex **2**: Mp.118–20°C; FT-IR (KBr): 3335, 3180, 1662, 1599, 1317, 1298, 804, 714, 574 cm^{-1}; ^{1}H NMR ($CDCl_3$, 500 MHz): δ (ppm): 8.11(d, 2H, $J = 8$ Hz), 7.57(t, 1H, $J = 7.5$ Hz), 7.46(t, 2H, $J = 7.5$ Hz), 6.39(s, 1H), 5.85(bs, 1H), 5.70(bs, 1H), 2.33(s, 6H); Crystal data are as follows (**CCDC No. 634316**): $C_{13}H_{15}N_3O_2$, *Mr* = 245.28, 100.0(1) K, Monoclinic, $P2(1)/c$, $a = 6.7626(2)$ Å, $b = 7.3958(2)$ Å, $c = 25.0054(7)$ Å, $\alpha = 90°$, $\beta = 92.1460(10)°$, $\gamma = 90°$, V = 1249.76(6) $Å^3$, Z = 4, μ(Mo K_α) = 0.090 mm^{-1}, Reflections collected/unique 26058/3699 [R(int) = 0.0545], R1($I > 2\sigma(I)$) = 0.0525, wR2 = 0.1325, R1 = 0.0714 (All data), wR2 = 0.1506.

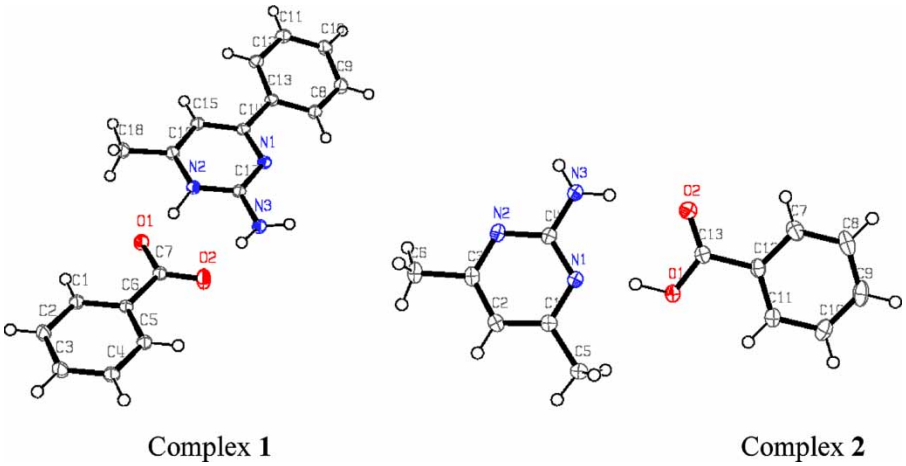


FIGURE 2 ORTEP (50% probability) diagram of complexes of substituted pyrimidines and benzoic acid.

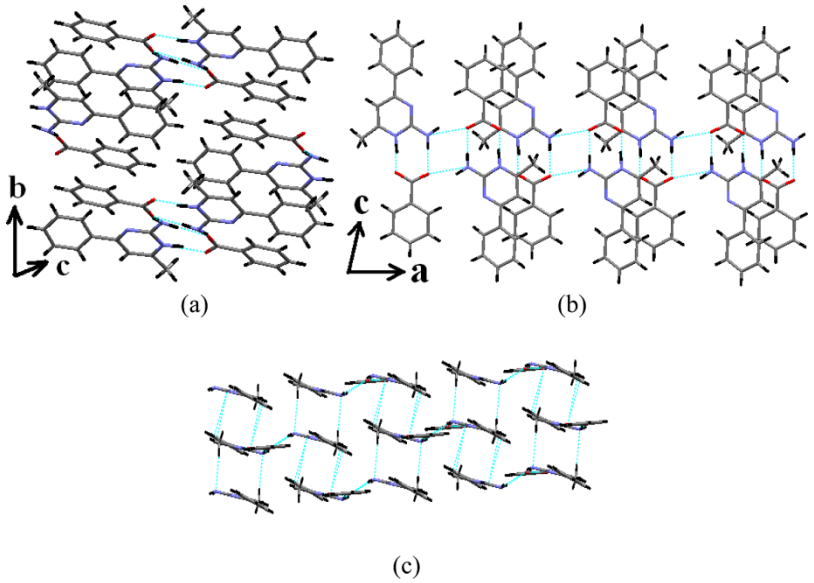


FIGURE 3 Illustrations for the crystal structure of complex **1**: (a) one-dimensional layer, viewed along the crystallographic *a*-axis, (b) viewed along the crystallographic *b*-axis and (c) elongation of wave-like layer. Hydrogen bonds are shown as dashed lines.

Analysis of the X-ray results [13] of the complex **1** reveals that an eight-membered hydrogen-bonded ring is formed between the benzoate anion and the pyrimidinium ion, which is generated in solid phase by the transfer of the acidic hydrogen of the benzoic acid to the pyrimidine ring 'N2' (Fig. 2). This complex is further hydrogen bonded with another complex through [N3—H2 . . . O2, 2.093(18) Å] to form another eight-membered cyclic hydrogen-bonded ring. This 2:2 ionic complex further interacts with another 2:2 complex unit through [C18—H18B . . . N3, 2.5647 Å]. By this way, wave-like polymeric chains are formed (Fig. 3(c)) which further interact with one another to form a three-dimensional network. Another important fact is that the pyrimidine ring 'N1' is not involved in any type of interaction (Table I).

In the case of complex **2**, the donor–acceptor array is highly similar with the reported results. Here, both

TABLE I Hydrogen bond parameters (Å, °) of complex **1**

D—H . . . A	D—H	H . . . A	D . . . A	D—H . . . A
N2—H1 . . . O1	1.128(18)	1.465(18)	2.5847(8)	170.8(18)
N3—H1 . . . O2	0.927(14)	1.874(14)	2.7995(8)	176.4(12)
N3—H2 . . . O2$^{(i)}$	0.884(17)	2.093(18)	2.8478(11)	142.7(14)
C8—H8A . . . N1	0.9296	2.4182	2.7492(9)	100.85
C18—H18B . . . N3$^{(ii)}$	0.9601	2.5647	3.5131(12)	169.55

Symmetry codes: (i) $2-x, -y, 1-z$; (ii) $1-x, -y, 1-z$.

TABLE II Hydrogen bond parameters (Å, °) of complex **2**

D—H...A	D—H	H...A	D...A	D—H...A
O1—H1...N1	1.01(3)	1.59(3)	2.5931(15)	170(2)
N3—H1...N2$^{(i)}$	0.87(2)	2.20(2)	3.0688(17)	174(2)
N3—H2...O2	0.90(2)	2.13(2)	3.0201(16)	170.1(19)
C8—H8A...O2$^{(ii)}$	0.9301	2.5858	3.4236(19)	150.11

Symmetry codes: (i) $-1-x, 1-y, 1-z$; (ii) $-x, 1/2+y, 1/2-z$.

homo and hetero interactions play a major role in the formation of the wave-like one-dimensional supramolecular synthon. Between two sets of donor–acceptor units, one set is being involved in the hetero interaction with the carboxylic acid moiety of benzoic acid and the other set is being involved in the homo interaction with the other substituted pyrimidine molecules. Each 2:2 unit interacts with another such unit through C—H...π[C6—H6B...*Cg*2($-x$, $2-y$, $1-z$), 2.99 Å; *Cg*2 is the centroid of the ring C7—C12] interactions [14] and forms a layer. The whole supramolecular network is formed by the wave-like arrangement of neighbour units which interact with each other through the C—H...O [C8—H8A...O2, 2.5858 Å] weak interaction (Table 2; Fig. 4).

From the above two complexes, it is found that two substituted pyrimidine amines recognise the same acid in different ways and ultimately form two interesting supramolecular architectures. The basic difference in the recognition of the two substituted pyrimidines in solid phase is that in case of complex **1**, proton transfer [11b] takes place and in the other (complex **2**), there is no proton transfer. By this way, in the case of complex **1** we observed the formation of an ionic complex, but in other a molecular complex was found. Though proton transfer in a complex is governed by the ΔpK_a rule [15], in the case of complex **1** proton transfer only occurs when it crystallised from the chloroform solution of the 1:1 mixture. The enhanced stability of the protonated species (complex **1**) when compared with that of complex **2** is due to the extended resonance stability of N2—H$^+$ (the 4-phenyl derivative of pyrimidine when compared with the 4-methyl derivative). To study

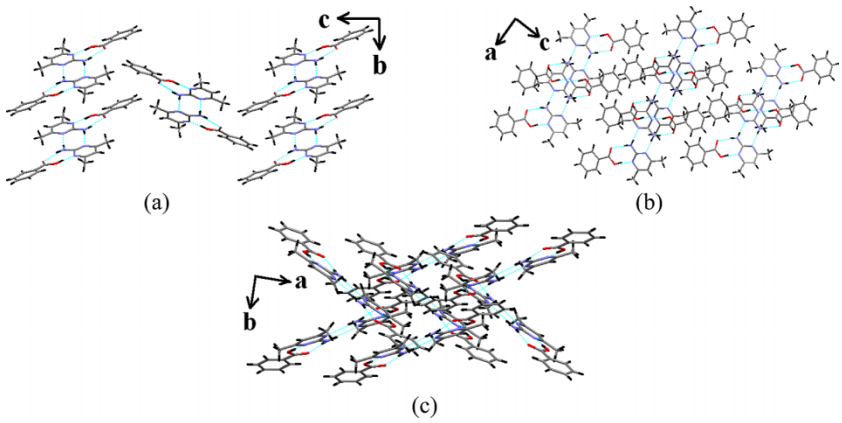


FIGURE 4 Illustrations for the crystal structure of complex **2**: (a) infinite two-dimensional wave-like layer, viewed along the crystallographic *a*-axis, (b) viewed along the crystallographic *b*-axis and (c) viewed along the crystallographic *c*-axis. Hydrogen bonds are shown as dashed lines.

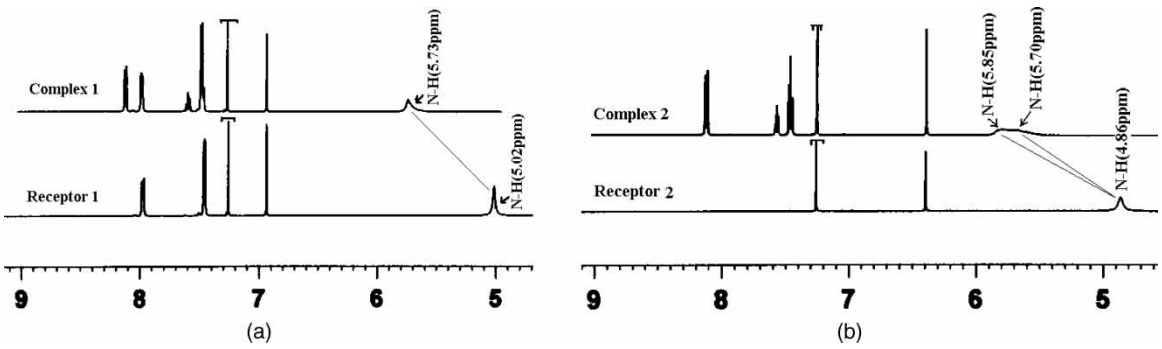


FIGURE 5 Partial ^{1}H NMR ($CDCl_3$, 500 MHz) spectra of receptors and their 1:1 complexes with benzoic acid: (a) receptor **1** and complex **1**, and (b) receptor **2** and complex **2**.

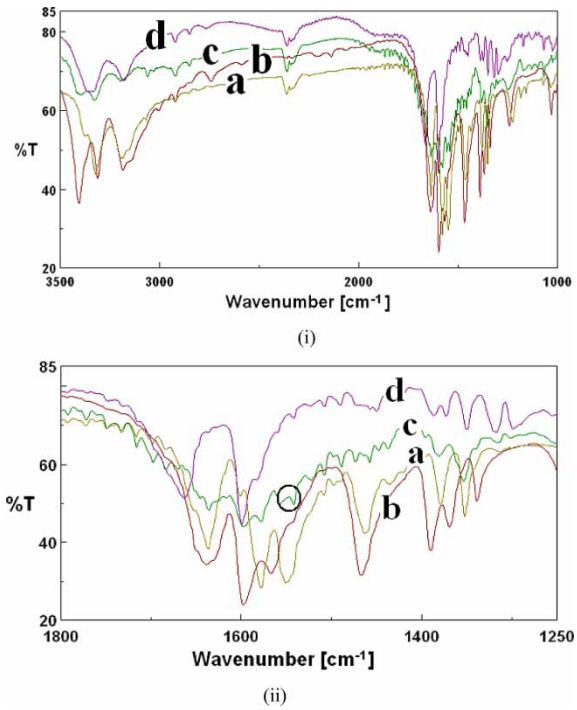


FIGURE 6 IR spectrum: (i) full spectrum of [a] receptor **1**, [b] receptor **2**, [c] complex **1**, and [d] complex **2** and (ii) enlarged portion (1800–1250 cm^{-1}).

the case of proton transfer, we have investigated the binding behaviour in solution phase by UV–Vis method [16] and we have also checked the ^1H NMR of both the 1:1 complexes in chloroform. In case of the UV–Vis method, the binding constants ($K_a = 3.6657 \times 10^3$ M^{-1} for complex **1** and $K_a = 7.8903 \times 10^3$ M^{-1} for complex **2**) are not much different. In other words, proton transfer does not take place in solution phase.

In case of ^1H NMR, no extra peak is found in case of complex **1** (Fig. 5). The NH protons are only shifted downfield (Δδ for complexes **1** and **2** are 0.61 and 0.99–0.84 ppm, respectively). Another interesting fact is that in case of complex **2**, the two amine protons are slightly differentiated (Δδ = 0.15 ppm) in the 1:1 complexation during the ^1H NMR study but this differentiation is not observed in complex **1**. Therefore, proton transfer did not take place in the solution phase. In the solid phase, it is probably the electronic effect due to very close proximity (during crystallisation), which leads to host–guest interactions, resulting in the proton transfer.

We have also studied the IR spectra (Fig. 6) [15f, 17] of the complexes with respect to the substituted pyrimidines and the results are interesting for the case of proton transfer in complex **1**. In the IR spectrum of complex **1**, a peak is observed at 1541 cm^{-1} due to the presence of the carboxylate anion (encircled in Fig. 6(ii)); however, this is not found in the IR spectrum of complex **2**, which also proved the case of proton transfer of complex **1** in the solid phase. From the structural viewpoint of complex **1**, since two hydrogen atoms of the pyrimidine ring are replaced by a methyl and a phenyl group (unsymmetric groups), the ring π-electron is not uniformly delocalised; rather an excess of the electron cloud is localised at the ring 'N' adjacent to the methyl group. It is probably for that reason that when this molecule crystallises from the solution, the proton transfer takes place resulting the formation of an organic salt which generates the whole supramolecular assembly. In the case of complex **2**, two hydrogen atoms of the pyrimidine ring are replaced by two methyl groups (symmetric groups) and the ring π-electron of the pyrimidine ring is uniformly distributed between the two ring 'N' atoms and hence no protonation occurs and a simple molecular complex has been formed. These two supramolecular arrays demonstrate beautifully the structural diversity that governs the crystal engineering of these complexes (Fig. 7).

CONCLUSION

From this study, it is observed that, either an ionic or a molecular complex can be formed by changing the substituents to the host containing the 2-aminopyrimidine moiety. For complex **1**, proton transfer takes place in the much higher concentrated solution from which single crystals are grown because of an electronic effect of the delocalised π-electron density of the unsymmetrically substituted 2-aminopyrimidine-based host. However, for the symmetrically substituted 2-aminopyrimidine-based host in complex **2**, a molecular complex is formed. In conclusion, different supramolecular networks can be engineered by changing the host 2-aminopyrimidine backbone.

FIGURE 7 Space filled model of the polymeric chain: (a) complex **1** and (b) complex **2**.

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