One pot three component synthesis of 9-arylpolyhydroacridine derivatives in an ionic liquid medium

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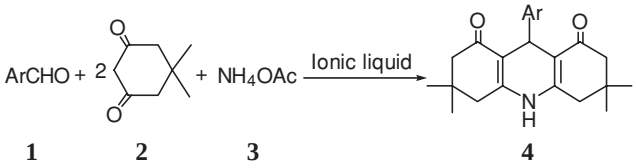
In this paper the preparation of 3,3,6,6-tetramethyl-9-aryl-1,2,3,4,5,6,7,8,9,10- decahydroacridin-1,8-dione derivatives from aromatic aldehydes, 5,5-dimethyl-1,3- cyclohexanedione and ammonium acetate in ionic liquids [bmim$^+$][BF_4^-] is described. This new method has the advantages of easier work-up, milder reaction conditions, high yields and an environmentally benign procedure compared with other methods.

Keywords: acridin-1,8-dione derivatives, ionic liquids, ammonium acetate

1,4-Dihydropyridines (1,4-DHPS) are well-known compounds because of their pharmacological profile as calcium channel modulators.[1] Chemical modifications carried on the DHP ring, such as the presence of different substituents[2] or heteroatoms,[3] have allowed the expansion of structure activity relationships and afforded some insight into molecular interactions at the receptor level. The usual method of 1,4-DHPS synthesis is from Meldrum's acid and dimedone in the presence of different aldehydes catalysed by ammonium acetate[4] or NH_3H_2O.[5] Recently there have been many methods reported for the synthesis of tricyclic compounds containing 1,4-dihydropyridines, such as acridine derivatives, from aldehydes, dimedone and ammonium acetate by traditional heating in organic solvents,[6] or in water catalysed by TEBA,[7] or improved under microwave irradiation.[8] However, they were reacted in organic solvents or had low solubility in water, which inspired us to explore new reaction media for the synthesis of 1,4-dihydropyridines. Room temperature ionic liquids, especially those based on the 1-*N*-alkyl-3-methylimidazolium cation, have shown great promise as attractive alternatives to conventional solvents.[9] Due to the potential of room temperature ionic liquids as alternative reaction media for catalytic processes, much attention has been focused on organic reactions promoted by them.[10] The unique property of room temperature ionic liquids is that they have essentially no vapour pressure, which makes them optimal replacements for the volatile organic solvents traditionally used as industrial solvents. A nice feature of reactions in ionic liquid is that yields can be optimised by changing the anions or properties of the cation. In addition, several ionic liquids show enhancement in reaction rates and selectivity, compared to organic solvents with the added benefit of the ease of recovery and reuse. Because of these advantages, ionic liquids can make a significant contribution to green chemistry and are used widely as reaction media in organic chemistry.[11]



1 **2** **3** **4**

Scheme 1

In view of this we now wish to report the synthesis of 9-arylpolyhydroacridine derivatives in an ionic liquid medium. When the three components of aromatic aldehyde **1**, 5,5-dimethyl-1,3-cyclohexanedione **2** and ammonium acetate **3** were treated in an ionic liquid [bmim$^+$][BF_4^-] at 80°C for a few hours (Scheme 1), the desired 3,3,6,6-tetramethyl-9-aryl-1,2,3,4,5,6,7,8,9,10-decahydroarcidin-1,8-dione derivatives **4** were obtained in high yields (89–98%) (Table 1).

We began our study of the reaction shown in Scheme 1 by optimising the reaction conditions for preparation of 3,3,6,6-tetramethyl-9-aryl-1,2,3,4,5,6,7,8,9,10-deca-hydroarcidin-1,8-dione (**4**). A summary of the optimisation experiments is provided in Table 1. It turned out that at room temperature, no reaction took even when the ratio of aldehyde to ammonium acetate is 1 : 4 (Table 1, entries 1 and 2). Moreover, different ionic liquids were studied and from Table 1, we could conclude that the ionic liquid [bmim$^+$][BF_4^-] was the best reaction medium for this reaction. On the other hand, while **4a** could be obtained at 80°C with 1 equivalent of ammonium acetate, the yield was disappointingly low (Table 1, entry 6). The effects of ammonium acetate on this reaction were then investigated. To our delight, the yield could indeed be

Table 1 Synthesis of **4a** in ionic liquid under different reaction conditions[a]

Entry	Temperature/°C	NH_4OAc/equivalents	ionic liquid	Time/h	Yield[b]/(%)
1	r.t.	1	[bmim$^+$][BF_4^-]	3	0
2	r.t.	4	[bmim$^+$][BF_4^-]	6	0
3	80	4	[bmim$^+$]Br$^-$	3	67
4	80	4	[emim$^+$]Br$^-$	3	56
5	80	4	[emim$^+$][BF_4^-]	3	87
6	80	1	[bmim$^+$][BF_4^-]	3	58
7	80	2	[bmim$^+$][BF_4^-]	3	87
8	**80**	**4**	**[bmim$^+$][BF_4^-]**	**3**	**94**
9	80	6	[bmim$^+$][BF_4^-]	3	94

[a]Reaction condition: 10 ml ionic liquid, 2 mmol 4-chlorobenzaldehyde and 4 mmol dimedone.
[b]Isolated yields.

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Table 2 Synthesis of **4** in ionic liquid [bmim$^+$][BF_4^-][a]

Entry	Ar	Time	Products	Yields/%[b]
1	4-ClC_6H_4	3	4a	94
2	4-HOC_6H_4	5	4b	89
3	4-$CH_3OC_6H_4$	4	4c	92
4	4-$CH_3C_6H_4$	4	4d	94
5	4-BrC_6H_4	3	4e	95
6	4-$(CH_3)_2NC_6H_4$	4	4f	90
7	3,4-$OCH_2OC_6H_3$	4	4g	92
8	3,4-$(CH_3O)_2C_6H_3$	4	4h	92
9	3-CH_3O-4-HOC_6H_3	5	4i	93
10	C_6H_5	3	4j	92
11	3-$NO_2C_6H_4$	3	4k	96
12	2,4-$Cl_2C_6H_3$	3	4l	98
13	2-ClC_6H_4	3	4m	98

[a]Reaction condition: 10 ml ionic liquid, 2 mmol aromatic aldehyde and 4 mmol dimedone, 8 mmol ammonium acetate, 80 °C.
[b]Isolated yields.

improved remarkably by adding the ammonium acetate to 4 equiv (Table 1, entry 8).

In order to demonstrate the efficiency and scope of the present method, we applied this ionic liquid to the reaction of a variety of aromatic aldehydes with dimedone in the presence of an excess of ammonium acetate. The results are summarised in Table 2. Data from Table 2 demonstrated that the reactions proceeded smoothly to give **4** in high yields under the optimised conditions. All the products were characterised by their melting points, ^{1}H NMR and IR spectra.

Finally the recovery and reuse of the ionic liquid were studied. At completion monitored by TLC, the reaction mixture was allowed to cool to room temperature. The solid product was isolated by filtration and the filtrate of the ionic liquid [bmim$^+$][BF_4^-] together with ammonium acetate could be recovered easily by drying at 80 °C *in vacuo* for several hours. Investigations using 4-chlorobenzaldehyde, dimedone and 2 equivalents of ammonium acetate a model system showed the effect of successive reuse of the recovered ionic liquid. A summary is shown in Table 3. Even in the fourth round the yield of the product **4** is fairly good.

In conclusion, with high yields and mild conditions, we think that the present work provides a useful method for the preparation of 3,3,6,6-tetramethyl-9-aryl-1,2,3,4,5,6,7,8,9, 10-decahydroarcidin-1,8-dione derivatives. Compared with other methods, this new method has the advantages of easier work-up, milder reaction conditions, high yields and an environmentally benign procedure.

Experimental

Melting points were determined in open capillaries and are uncorrected. IR spectra were recorded on a Tensor 27 spectrometer in KBr. ^{1}H NMR spectra were obtained for solutions in $CDCl_3$ or DMSO-d_6 with Me_4Si as internal standard using a Bruker-400 spectrometer.

General procedure for preparation of ionic liquids

1-Butyl-3-methylimidazolium Bromide [bmim$^+$]Br$^-$: In a three-necked, 500 ml round-bottomed flask equipped with reflux condenser, 100-ml dropping funnel, and magnetic stirrer, 23.22 g (0.28 mol) of 1-methylimidazole was diluted under Ar in 250 ml of absolute ethanol and 39.00 g (0.29 mmol) of bromobutane. The solution was then refluxed for 30 min. and the mixture was concentrated at 80 °C under reduced pressure. The product was obtained by drying the residual liquid at 80 °C *in vacuo*.

1-Butyl-3-methylimidazolium tetrafluorate [bmim$^+$][BF_4^-]: A solution of [bmim$^+$]Br$^-$ (26.30 g, 0.12 mmol) in acetone (50 ml), was added dropwise to a rapidly stirring solution of $NaBF_4$(13.20 g, 0.12 mmol) in acetone (50 ml). The mixture was stirred at room temperature for 96 h. The solid was then filtered off and washed with acetone. The filtrate was concentrated at 50 °C under reduced pressure. The product [bmim$^+$][BF_4^-] was obtained by drying the residual liquid at 80 °C *in vacuo*.

Table 3 Study on the reuse of ionic liquid [bmim$^+$][BF_4^-][a]

Round	Temperature/°C	Reaction time/h	Yield/%[b]
1	80	3	94
2	80	3	92
3	80	3	90
4	80	3	89

[a]Reaction condition: 10 ml ionic liquid, 2 mmol aromatic aldehyde and 4 mmol dimedone. 4 mmol ammonium acetate
[b]Isolated yields.

General procedure for preparation of **4**

A dry 50 ml flask was charged with aromatic aldehyde **1** (2 mmol), 5,5-dimethyl-1,3-cyclohexanedione **2** (4 mmol), ammonium acetate **3** (8 mmol) and ionic liquid [bmim$^+$][BF_4^-] (10 ml). The mixture was stirred at 80°C for 3–5 h to complete the reaction (monitored by TLC), then cooled to room temperature. The yellow solid was filtered off and washed with water. The filtrate of ionic liquid [bmim$^+$][BF_4^-] was then recovered for reuse by drying at 80 °C several hours in a vacuum. The crude product was purified by recrystallisation from 95% EtOH to give **4**.

In the ^{1}H NMR the AA'XX' systems of the *para*-substituted benzene rings appeared as pairs of doublets.

3,3,6,6-tetramethyl-9-(4-chlorophenyl)-1,2,3,4,5,6,7,8,9, 10-decahydroacridin-1,8-dione (**4a**): M.p. 296–298 °C (lit.[8] 298–300 °C); ^{1}H NMR (DMSO-d_6, 400 MHz) δ : 0.91 (s, 6H, 2 × CH_3), 1.06 (s, 6H, 2 × CH_3), 2.08(d, J = 16.4 Hz, 2H, CH_2), 2.26 (d, J = 16.4 Hz, 2H, CH_2), 2.50 (d, J = 17.2 Hz, 2H, CH_2), 2.57 (d, J = 17.2 Hz, 2H, CH_2), 5.10 (s, 1H, CH), 6.50 (s, 1H, NH), 7.22 (d, J = 8.0 Hz, 2H, ArH), 7.31 (d, J = 8.0 Hz, 2H, ArH); IR (KBr) ν: 3285, 3049, 2956, 2871, 1644, 1612, 1475, 1394, 1364, 1222, 1170, 1142, 1088, 846 cm^{-1}.

3,3,6,6-tetramethyl-9-(4-hydroxyphenyl)-1,2,3,4,5,6,7,8,9, 10-decahydroacridin-1,8-dione (**4b**): M.p. >300 °C (lit.[7] >300 °C); ^{1}H NMR (DMSO-d_6, 400 MHz) δ : 0.88 (s, 6H, 2 × CH_3), 1.00 (s, 6H, 2 × CH_3), 1.98 (d, J = 15.6 Hz, 2H, CH_2), 2.15 (d, J = 15.6 Hz, 2H, CH_2), 2.30 (d, J = 17.2 Hz, 2H, CH_2), 2.42 (d, J = 17.2 Hz, 2H, CH_2), 4.70 (s, 1H, CH), 6.52 (d, J = 8.4 Hz, 2H, ArH), 6.92 (d, J = 8.4 Hz, 2H, ArH), 8.94 (s, 1H, NH), 9.16 (s, 1H, OH); IR (KBr) ν: 3276, 3186, 2954, 1645, 1613, 1510, 1475, 1372, 1254, 1221, 1169, 1142, 840 cm^{-1}.

3,3,6,6-tetramethyl-9-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9, 10-decahydroacridin-1,8-dione (**4c**): M.p. 273–275 °C (lit.[6] 270–272 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.96 (s, 6H, 2 × CH_3), 1.07 (s, 6H, 2 × CH_3), 2.15 (d, J = 16.0 Hz, 2H, CH_2), 2.21 (d, J = 16.8 Hz, 2H, CH_2), 2.24 (d, J = 16.0 Hz, 2H, CH_2), 2.31 (d, J = 16.8 Hz, 2H, CH_2), 3.69 (s, 3H, CH_3O), 5.02 (s, 1H, CH), 6.72 (d, J = 8.0 Hz, 2H, ArH), 7.20 (s, 1H, NH), 7.24 (d, J = 8.0 Hz, 2H, ArH); IR (KBr) ν: 3204, 3071, 2958, 2839, 1646, 1607, 1486, 1423, 1396, 1367, 1300, 1263, 1223, 1171, 1145, 1032, 1006, 835 cm^{-1}.

3,3,6,6-tetramethyl-9-(4-methylphenyl)-1,2,3,4,5,6,7,8,9, 10-decahydroacridin-1,8-dione (**4d**): M.p. >300 °C (lit.[7] >300 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.98 (s, 6H, 2 × CH_3), 1.09 (s, 6H, 2 × CH_3), 2.16 (d, J = 16.0 Hz, 2H, CH_2), 2.19–2.28 (m, 7H, 2 × CH_2+ CH_3), 2.38 (d, J = 16.0 Hz, 2H, CH_2), 5.03 (s, 1H, CH), 6.11 (s, 1H, NH), 7.00 (d, J = 6.8 Hz, 2H, ArH), 7.22 (d, J = 6.8 Hz, 2H, ArH); IR (KBr) ν: 3181, 3067, 2959, 2867, 1650, 1612, 1492, 1398, 1366, 1267, 1221, 1172, 1146, 1119, 1094, 1020, 837 cm^{-1}.

3,3,6,6-tetramethyl-9-(4-bromophenyl)-1,2,3,4,5,6,7,8,9, 10-decahydroacridin-1,8-dione (**4e**): M.p. >300 °C (lit.[7] >300 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.97 (s, 6H, 2 × CH_3), 1.09 (s, 6H, 2 × CH_3), 2.16 (d, J = 16.0 Hz, 2H, CH_2), 2.25 (d, J = 16.8 Hz, 4H, 2 × CH_2), 2.37 (d, J = 16.8 Hz, 2H, CH_2), 5.03 (s, 1H, CH), 6.56 (s, 1H, NH), 7.22 (d, J = 8.0 Hz, 2H, ArH), 7.31 (d, J = 8.0 Hz, 2H, ArH); IR (KBr) ν: 3175, 3056, 2954, 1647, 1609, 1487, 1397, 1366, 1222, 1145, 1011, 841 cm^{-1}.

3,3,6,6-tetramethyl-9-(4-dimethylaminophenyl)-1,2,3,4,5,6, 7,8,9,10-decahydroacridin-1,8-dione (**4f**): M.p. 270–272 °C (lit.[6] 264–266 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.97 (s, 6H, 2 × CH_3), 1.07 (s, 6H, 2 × CH_3), 2.15 (d, J = 16.0 Hz, 2H, CH_2), 2.22 (d, J = 16.0 Hz, 4H, 2 × CH_2), 2.33 (d, J = 16.0 Hz, 2H, CH_2), 2.86 (s, 6H, 2 × NCH_3), 4.99 (s, 1H, CH), 6.41 (s, 1H, NH), 6.67 (d, J = 8.0 Hz, 2H, ArH), 7.21 (d, J = 8.0 Hz, 2H, ArH); IR (KBr) ν: 3178, 3062, 2954, 2802, 1649, 1606, 1580, 1449, 1395, 1364, 1258, 1222, 1188, 1169, 1144, 1124, 1040, 828 cm^{-1}.

3,3,6,6-tetramethyl-9-(3,4-methylenedioxylphenyl)-1,2,3,4, 5,6,7,8,9,10-decahydroacridin-1,8-dione (**4g**): M.p. >300 °C (lit.[8]

324–326 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.99 (s, 6H, 2 × CH_3), 1.09 (s, 6H, 2 × CH_3), 2.19 (d, *J* = 16.8 Hz, 2H, CH_2), 2.25 (d, *J* = 16.0 Hz, 2H, CH_2), 2.28 (d, *J* = 16.0 Hz, 2H, CH_2), 2.39 (d, *J* = 16.8 Hz, 2H, CH_2), 4.99 (s, 1H, CH), 5.85 (s, 2H, OCH_2O), 6.21 (s, 1H, NH), 6.64 (d, *J* = 8.0 Hz, 1H, ArH), 6.80 (d, *J* = 8.0 Hz, 1H, ArH), 6.84 (s, 1H, ArH); IR (KBr) ν: 3202, 3070, 2959, 2787, 1652, 1607, 1488, 1396, 1368, 1292, 1225, 1170, 1144, 1038, 938, 920, 865, 815, 679 cm^{-1}.

3,3,6,6-tetramethyl-9-(3,4-dimethoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridin-1,8-dione (**4h**): M.p. 261–263 °C (lit.[8] 260–262°C); ^{1}H NMR ($CDCl_3$) δ : 0.97 (s, 3H, CH_3), 0.99 (s, 3H, CH_3), 1.06 (s, 3H, CH_3), 1.09 (s, 3H, CH_3), 2.14–2.42 (m, 8H, 4 × CH_2), 3.77 (d, *J* = 11.6 Hz, 3H, OCH_3), 3.82 (d, *J* = 11.6 Hz, 3H, OCH_3), 5.04 (s, 1H, CH), 6.66 (s, 1H, NH), 6.69 (d, *J* = 7.6 Hz, 1H, ArH), 6.81 (d, *J* = 7.6 Hz, 1H, ArH), 6.92 (s, 1H, ArH); IR (KBr) ν: 3199, 3070, 2954, 2836, 1642, 1611, 1513, 1485, 1421, 1396, 1367, 1264, 1223, 1172, 1138, 1023, 889, 857, 813, 750, 679 cm^{-1}.

3,3,6,6-tetramethyl-9-(3-methoxyl-4-hydroxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridin-1,8-dione (**4i**): M.p. >300 °C (lit.[7] >300 °C); ^{1}H NMR ($CDCl_3$) δ : 0.99 (s, 6H, 2 × CH_3), 1.09 (s, 6H, 2 × CH_3), 2.19 (d, *J* = 16.4 Hz, 2H, CH_2), 2.25 (d, *J* = 16.8 Hz, 2H, CH_2), 2.28 (d, *J* = 16.8 Hz, 2H, CH_2), 2.40 (d, *J* = 16.4 Hz, 2H, CH_2), 3.88 (s, 3H, OCH_3), 4.98 (s, 1H, CH), 6.22 (s, 1H, NH), 6.62 (d, *J* = 7.6 Hz, 1H, ArH), 6.71 (d, *J* = 7.6 Hz, 1H, ArH), 7.04 (s, 1H, ArH), 7.53 (s, 1H, OH); IR (KBr) ν: 3228, 3168, 3048, 2955, 1648 1625, 1489, 1427, 1399, 1271, 1172, 1143, 1037, 1006, 884, 845, 748, 677 cm^{-1}.

3,3,6,6-tetramethyl-9-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridin-1,8-dione (**4j**) M.p. 194–196 °C (lit.[6] 190–192 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.96 (s, 6H, 2 × CH_3), 1.08 (s, 6H, 2 × CH_3), 2.16 (d, *J* = 16.8 Hz, 2H, CH_2), 2.20 (d, *J* = 16.8 Hz, 2H, CH_2), 2.24 (d, *J* = 16.8 Hz, 2H, CH_2), 2.34 (d, *J* = 16.8 Hz, 2H, CH_2), 5.08 (s, 1H, CH), 7.01 (s, 1H, NH), 7.07–7.21 (m, 3H, ArH), 7.33 (d, *J* = 7.2 Hz, 2H, ArH); IR (KBr) ν: 3175, 3045, 2961, 2870, 1649, 1619, 1494, 1395, 1369, 1343, 1258, 1222, 1147, 742, 750 cm^{-1}.

3,3,6,6-tetramethyl-9-(3-nitrophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridin-1,8-dione (**4k**): M.p. 286–288 °C (lit.[8] 283–285 °C); ^{1}H NMR ($CDCl_3$) δ : 0.97 (s, 6H, 2 × CH_3), 1.11 (s, 6H, 2 × CH_3), 2.17 (d, *J* = 16.8 Hz, 2H, CH_2), 2.26 (d, *J* = 16.8 Hz, 2H, CH_2), 2.32 (d, *J* = 16.8 Hz, 2H, CH_2), 2.43 (d, *J* = 16.8 Hz, 2H, CH_2), 5.17 (s, 1H, CH), 6.26 (s, 1H, NH), 7.39 (dd, *J* = 8.0 Hz, *J*′ = 7.2 Hz, 1H, ArH), 7.89 (d, *J* = 7.2 Hz, 1H, ArH), 7.97 (d, *J* = 8.0 Hz, 1H, ArH), 8.04 (s, 1H, ArH); IR (KBr) ν: 3183, 3064, 2959, 1647, 1609, 1528, 1496, 1425, 1397, 1367, 1255, 1223, 1144, 1092, 813, 692 cm^{-1}.

3,3,6,6-tetramethyl-9-(2,4-dichlorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridin- 1,8- dione (**4l**): M.p. >300°C; ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.99 (s, 6H, 2 × CH_3), 1.08 (s, 6H, 2 × CH_3), 2.13 (d, *J* = 16.8 Hz, 2H, CH_2), 2.21 (d, *J* = 16.8 Hz, 4H, 2 × CH_2), 2.36 (d, *J* = 16.8 Hz, 2H, CH_2), 5.26 (s, 1H, CH), 6.11 (s, 1H, NH), 7.13 (d, *J* = 8.0 Hz, 1H, ArH), 7.24 (s, 1H, ArH), 7.47 (d, *J* = 8.0 Hz, 1H, ArH); IR (KBr) ν: 3305, 3064, 2958, 2874, 1641,1614, 1584, 1474, 1364, 1223, 1169, 1143, 1100, 1049, 864, 855, 838 cm^{-1}.

3,3,6,6-tetramethyl-9-(2-chlorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridin-1,8-dione (**4m**): M.p. 231–233°C (lit. [8] 226–227 °C); ^{1}H NMR ($CDCl_3$, 400 MHz) δ : 0.95 (s, 6H, 2 × CH_3), 1.05 (s, 6H, 2 × CH_3), 2.12 (d, *J* = 16.8 Hz, 2H, CH_2), 2.16 (d, *J* = 16.8 Hz, 2H, CH_2), 2.21 (d, *J* = 16.8 Hz, 2H, CH_2), 2.27 (d, *J* = 16.8 Hz, 2H, CH_2), 5.36 (s, 1H, CH), 7.01–7.05 (m, 1H, ArH), 7.14–7.17 (m, 1H, ArH), 7.24 (d, *J* = 7.6 Hz, 1H, ArH); 7.50 (d, *J* = 8.0 Hz, 1H, ArH), 7.66 (s, 1H, NH); IR (KBr) ν: 3199, 3071, 2953, 2871, 1635, 1614, 1487, 1422, 1397, 1364, 1256, 1223, 1171, 1146, 1124, 1037, 750 cm^{-1}.

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