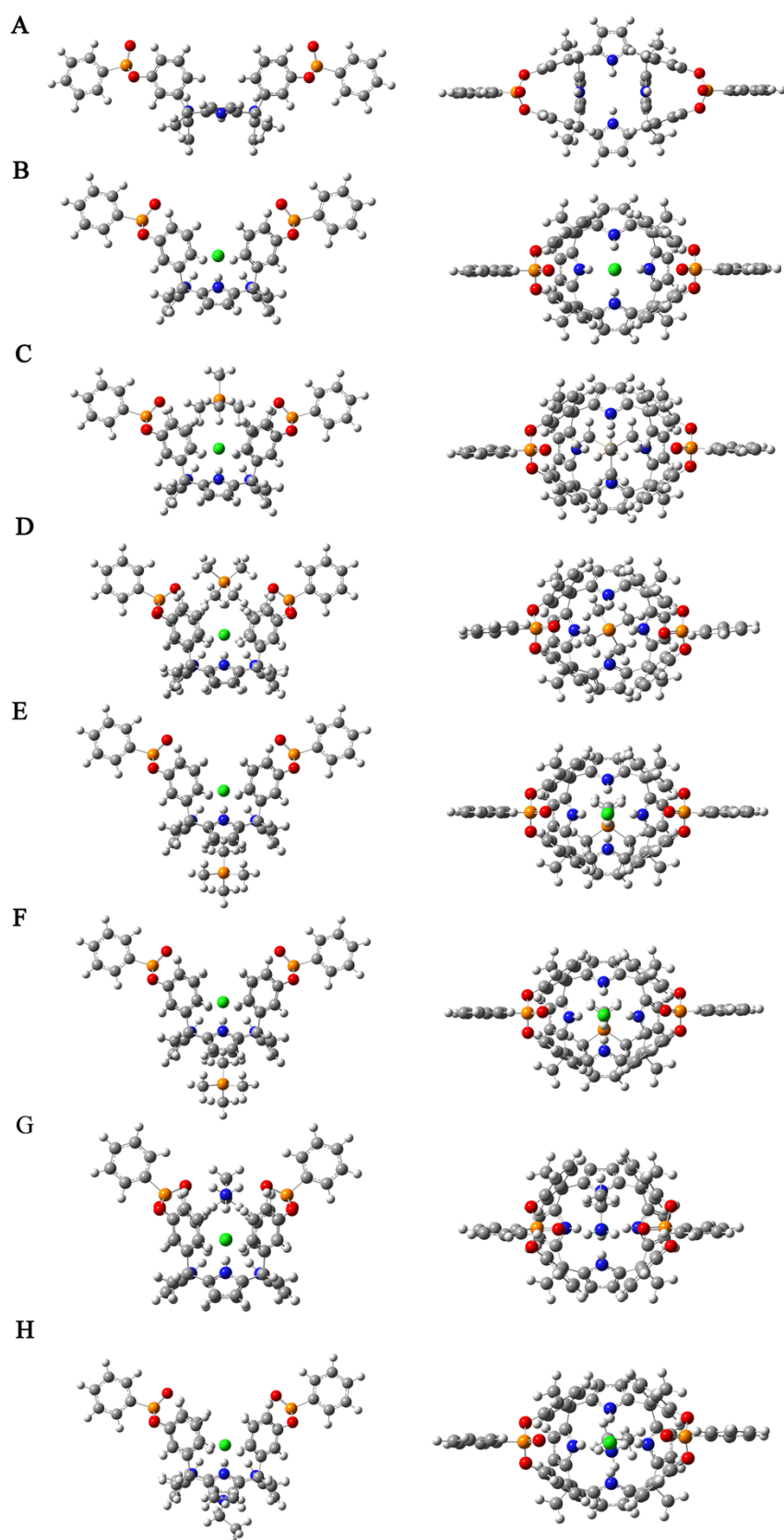




**Fig. S1** Side and top views of optimized structures of the ii and ii complexes.  
(A) ii (B) ii-Cl (C) ii-TMPCl-1a (D) ii-TMPCl-1b (E) ii-TMPCl-2a (F) ii-TMPCl-2b  
(G) ii-EAMCl-1 (H) ii-EAMCl-2.

**Table S1** Selected bond distances (in Å) and bond angles (in degree) for optimized structures of the ii and ii complexes

| parameter   | ii       | ii-Cl    | ii-TMPCl<br>-1a | ii-TMPCl<br>-1b | ii-TMPCl<br>-2a | ii-TMPCl<br>-2b | ii-EAMCl<br>-1 | ii-EAMCl<br>-2 |
|-------------|----------|----------|-----------------|-----------------|-----------------|-----------------|----------------|----------------|
| symmetry    | $C_{2v}$ | $C_{2v}$ | $C_s$           | $C_2$           | $C_s$           | $C_1$           | $C_s$          | $C_1$          |
| N1-H1       | 1.01     | 1.03     | 1.02            | 1.02            | 1.03            | 1.03            | 1.02           | 1.03           |
| N2-H2       | 1.01     | 1.02     | 1.02            | 1.02            | 1.03            | 1.03            | 1.02           | 1.03           |
| N3-H3       | 1.01     | 1.03     | 1.02            | 1.02            | 1.03            | 1.03            | 1.02           | 1.03           |
| N4-H4       | 1.01     | 1.02     | 1.02            | 1.02            | 1.03            | 1.03            | 1.02           | 1.03           |
| H1···Cl     | -        | 2.19     | 2.26            | 2.24            | 2.13            | 2.13            | 2.18           | 2.17           |
| H2···Cl     | -        | 2.24     | 2.30            | 2.26            | 2.21            | 2.21            | 2.22           | 2.20           |
| H3···Cl     | -        | 2.19     | 2.27            | 2.24            | 2.17            | 2.17            | 2.28           | 2.17           |
| H4···Cl     | -        | 2.24     | 2.30            | 2.26            | 2.21            | 2.21            | 2.22           | 2.26           |
| ∠N1-H1···Cl | -        | 175.79   | 171.52          | 178.09          | 175.11          | 175.10          | 168.99         | 160.35         |
| ∠N2-H2···Cl | -        | 163.39   | 161.56          | 169.47          | 163.80          | 163.77          | 178.40         | 160.79         |
| ∠N3-H3···Cl | -        | 175.79   | 174.04          | 178.09          | 170.30          | 170.30          | 162.48         | 169.86         |
| ∠N4-H4···Cl | -        | 163.39   | 161.56          | 169.47          | 163.80          | 163.82          | 178.40         | 155.04         |
| P3···Cl     | -        | -        | 3.65            | 3.99            | 6.07            | 6.07            | -              | -              |
| P1···P2     | 14.01    | 10.81    | 10.89           | 9.81            | 10.37           | 10.37           | 8.15           | 10.15          |
| O1···O2     | 13.46    | 9.00     | 8.90            | 7.44            | 8.34            | 8.34            | 5.43           | 8.03           |
| O1···Ha     | -        | -        | 2.09            | 2.17            | -               | -               | -              | -              |
| O2···Hb     | -        | -        | 2.09            | 2.17            | -               | -               | -              | -              |
| ∠C1-Ha···O1 | -        | -        | 156.14          | 134.38          | -               | -               | -              | -              |
| ∠C2-        | -        | -        | 156.14          | 134.38          | -               | -               | -              | -              |
| N5···Cl     | -        | -        | -               | -               | -               | -               | 3.00           | 4.08           |
| H5···Cl     | -        | -        | -               | -               | -               | -               | 2.08           | -              |
| O1···H6     | -        | -        | -               | -               | -               | -               | 1.88           | -              |
| O2···H7     | -        | -        | -               | -               | -               | -               | 1.88           | -              |
| ∠N5-H5···Cl | -        | -        | -               | -               | -               | -               | 147.13         | -              |
| ∠N5-        | -        | -        | -               | -               | -               | -               | 143.72         | -              |
| ∠N5-        | -        | -        | -               | -               | -               | -               | 143.72         | -              |
| H5···N1     | -        | -        | -               | -               | -               | -               | -              | 2.57           |
| H6···N4     | -        | -        | -               | -               | -               | -               | -              | 2.01           |
| H7···N2     | -        | -        | -               | -               | -               | -               | -              | 2.04           |
| ∠N5-        | -        | -        | -               | -               | -               | -               | -              | 103.46         |
| ∠N5-        | -        | -        | -               | -               | -               | -               | -              | 176.50         |
| ∠N5-        | -        | -        | -               | -               | -               | -               | -              | 157.67         |

The atom labels in Fig. S1 are the same as those in Fig. 6 and Fig. 7 of the paper published in *RSC Adv* (2014, 4, 1864–1873).

**Table S2** NBO charges of ions and hosts and dipole moments (in Debye) in ii complexes

|               | ii-Cl  | ii-TMPCl | ii-TMPCl | ii-TMPCl | ii-TMPCl | ii-EAMCl | ii-EAMCl |
|---------------|--------|----------|----------|----------|----------|----------|----------|
|               |        | -1a      | -1b      | -2a      | -2b      | -1       | -2       |
| Cl            | -0.757 | -0.811   | -0.804   | -0.724   | -0.724   | -0.760   | -0.733   |
| TMP           | —      | 0.943    | 0.966    | 0.958    | 0.958    | —        | —        |
| EAM           | —      | —        | —        | —        | —        | 0.874    | 0.925    |
| ii            | -0.243 | -0.132   | -0.162   | -0.234   | -0.234   | -0.114   | -0.192   |
| dipole moment | 2.370  | 12.373   | 14.821   | 15.424   | 15.424   | 12.105   | 8.203    |