Supporting Information

Using alcohols as alkylation reagents for 4-cyanopyridinium and N,N'-dialkyl-4,4'-bipyridinium and their one-dimensional iodoplumbates

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Contents

Scheme S1  The possible mechanisms on generating complexes 1–8 ........................................ S3

Fig. S1  3D packing diagrams of complexes 2 and 4 .............................................................. S4

Fig. S2  3D packing diagram of complex 5 ........................................................................... S4

Fig. S3  3D packing diagram of complex 7 ........................................................................... S4

Fig. S4  Frequency dependence of permittivity (imaginary part) of complexes 1–8 ............... S5
\[
\text{H}_2\text{O} + \text{I}_2 \xrightarrow{(a)} \text{HI} \xrightarrow{(b)} \text{R-I} \\
\]

\[
\text{R-OH} \\
\]

\[
\text{N} \quad \text{or} \quad \text{N} \\
\]

\[
\text{R-N}^+ \text{CN} \quad \text{or} \quad \text{R-N}^+ \text{CN} \quad \text{I}^- \\
\]

\[
\text{R-N}^+ \text{CN} \quad \text{or} \quad \text{R-N}^+ \text{CN} \quad \text{I}^- \\
\]

\[
\text{PbI}_2 \\
\]

\[
\text{R-N}^+ \text{CN} \quad \text{R-N}^+ \text{CN} \quad 2\text{I}^- \\
\]

R = Ethyl, Propyl, Isopropyl, Benzyl for 4-cypy
R = Ethyl, Propyl, Isopropyl for 4,4'-bipy

**Scheme S1** The possible mechanisms on generating complexes 1–8 with C\(^+\)/C\(^{2+}\) and V\(^{2+}\) cations.
**Fig. S1**  3D packing diagrams of 2 (a) and 4 (b) looking down along $a$-axis. Atom color codes: C, gray; N, cyan; Pb, blue; I, pink (the same hereinafter). H atoms are omitted for clarity.

**Fig. S2**  3D packing diagram of 5 looking down along $a$-axis. H atoms are omitted for clarity.

**Fig. S3**  3D packing diagram of 7 looking down along $a$-axis. H atoms are omitted for clarity.
**Fig. S4** Frequency dependence of permittivity (imaginary part) of 1–8 at 298 K.