Supporting Information.

AFM images of [Pt(en)$_2$][Pt(en)$_2$(Cl)$_2$](3)$_4$ (n = 16) cast on mica surface (a) and its height profile (b). Scanned area: 1001 x 1001 nm. Cross-sectional view of the AFM image was taken along the line in (a).

AFM observation (Instrument, Topometrix, TMX-2100 Explorer, non-contact mode) of a cast [Pt(en)$_2$][PtCl$_2$(en)$_2$](3)$_4$ sample. A chloroform solution of [Pt(en)$_2$][PtCl$_2$(en)$_2$](3)$_4$ was once heated to 60 °C and then cooled to room temperature. Two drops of this indigo-colored solution (0.1 mM) were let flow on the mica surface, which was tilted by ca. 45° against the normal and dried in air. Rectangular crystalline islands (lengths, ca. 800 nm, widths, ca. 100 nm) with a height of 6-7 nm were abundantly observed. The observed height is consistent with the diameter or thickness of the structure models of Figure 3, and these islands must be formed by crystallization of the polyion complex [Pt(en)$_2$][PtCl$_2$(en)$_2$](3)$_4$, that has been dispersed in chloroform as nanowires.