

Figure S1. Upper panel: Simultaneous cyclic voltammogram (left ordinate) and $\Delta R / R$ vs. $\mathrm{E}\left(\mathrm{E}_{\text {ref }}=0.2 \mathrm{~V}\right.$, right ordinate) for a $25 \mu \mathrm{~m}$ diameter Pt microelectrode in CO-saturated $0.5 \mathrm{M} \mathrm{HClO}_{4}$ at $200 \mathrm{~V} / \mathrm{s}$ for CO adsorption potential $\mathrm{E}_{\text {ads }}=-0.26 \mathrm{~V}$. A time of 2.0 s at $\mathrm{E}_{\text {ads }}$ was allowed for full CO re-adsorption between every cycle (see text). Lower panel: Expanded section of the cyclic voltammogram (i vs E, thick line, left ordinate) shown in the Upper Panel, over the potential region $0.80<\mathrm{E}<1.05 \mathrm{~V}$ and $-1 / \mathrm{R}(\partial \mathrm{R} / \partial \mathrm{E})$ vs E curve (thin line, right ordinate) over the same potential region obtained by taking the derivative with respect to E of the fitted optical data in the Upper Panel in this figure.


Figure S2. Upper panel: Simultaneous cyclic voltammogram (left ordinate) and $\Delta R / R$ vs. E (right ordinate) for a $25 \mu \mathrm{~m}$ diameter Pt microelectrode in CO-saturated 0.5 M $\mathrm{HClO}_{4}$ at $100 \mathrm{~V} / \mathrm{s}$ (left panels) and $1000 \mathrm{~V} / \mathrm{s}$ (right panels) for CO adsorption potential $\mathrm{E}_{\text {ads }}=-0.26 \mathrm{~V}$. Only the potential region $0.7<\mathrm{E}<1.2 \mathrm{~V}$ is shown. A time of 2.0 s at $\mathrm{E}_{\text {ads }}$ was allowed for full CO re-adsorption between every cycle (see text). Lower panel: Expanded section of the cyclic voltammogram (i vs E, thick line, left ordinate) shown in the Upper Panel, over the potential region $0.75<\mathrm{E}<1.05 \mathrm{~V}$ and $-1 / \mathrm{R}(\partial \mathrm{R} / \partial \mathrm{E})$ vs E curve (thin line, right ordinate) over the same potential region obtained by taking the derivative with respect to E of the fitted optical data in the Upper Panel in this figure.

