Accurate detection of β-hCG in women serum and cervical secretions for predicting early pregnancy viability based on time-resolved luminescent lanthanide nanoprobes

- Supporting Information

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Table S1. Assay precision and analytical recovery of β-hCG added to women serum samples based on $LiLuF_4$:Ce,Tb TRPL nanoprobes.

Added ($ng\ mL^{-1}$)	Found ($ng\ mL^{-1}$)	CV (%) (n = 4)[a]	Recovery (%)
Sample 1	1.26	8.85	—
0.5	1.61	6.17	91.48
5	6.76	6.48	107.99
20	20.91	8.91	98.35
Sample 2	8.63	7.65	—
0.5	8.98	5.77	98.36
5	13.97	6.02	102.49
20	27.91	6.18	97.49
Sample 3	35.21	4.95	—
0.5	35.09	5.17	98.26
5	41.11	7.28	102.24
20	57.19	6.58	103.59

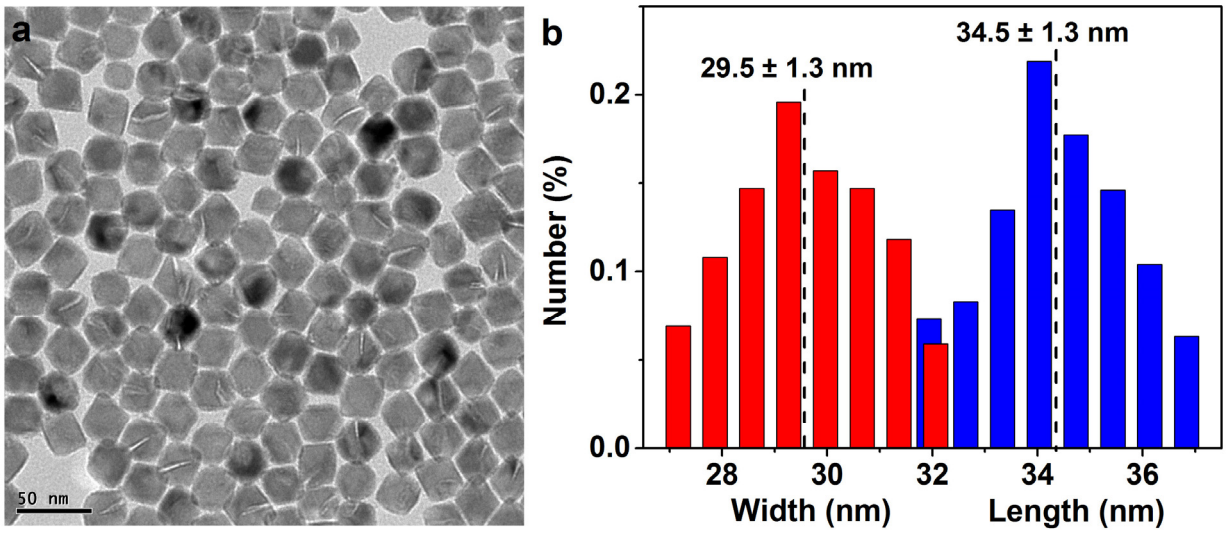

Figure S1. (a) TEM image and (b) size distribution of oleate-capped $LiLuF_4$:5%Ce, 5%Tb NPs. The size distribution of the NPs was obtained by randomly calculating 200 particles in the TEM image.

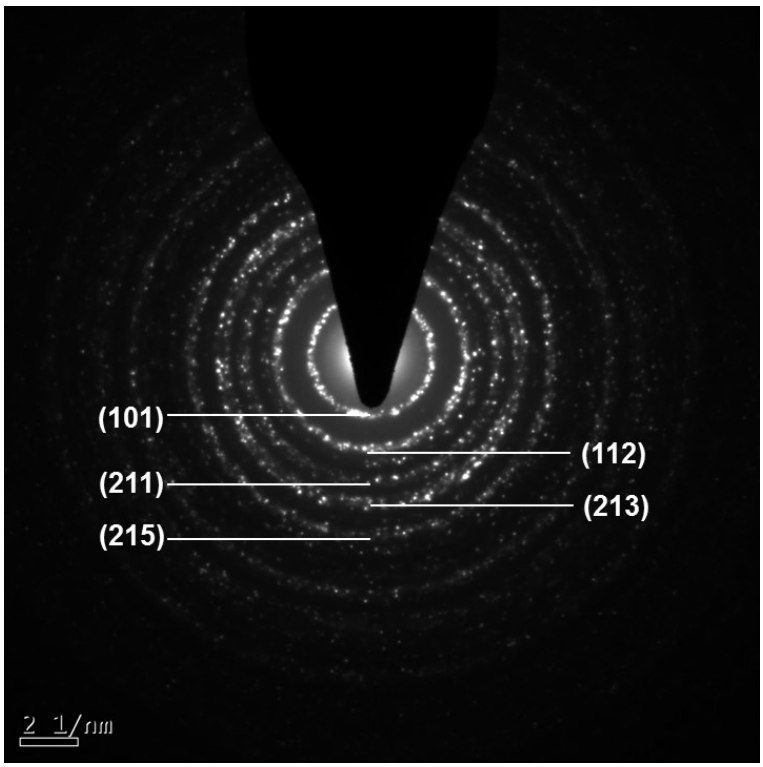

Figure S2. Selected area electron diffraction (SAED) pattern of $LiLuF_4$:5%Ce, 5%Tb NPs, showing high crystallinity of the NPs.

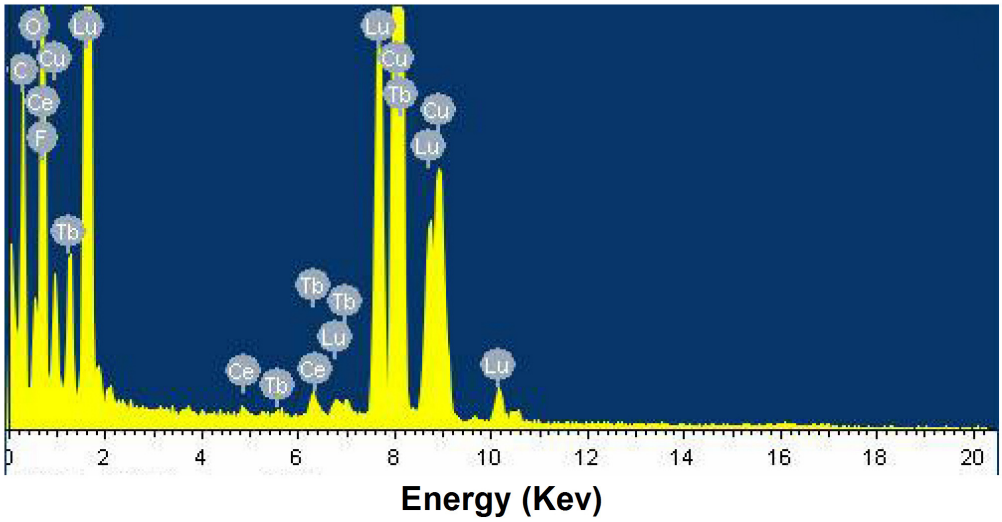

Figure S3. Energy dispersive X-ray spectrum (EDS) of $LiLuF_4$:5%Ce, 5%Tb NPs, showing the elements of Ce, Tb, Lu, and F in the NPs. Note that Li cannot be detected by EDS because of its small atomic number (M = 3).

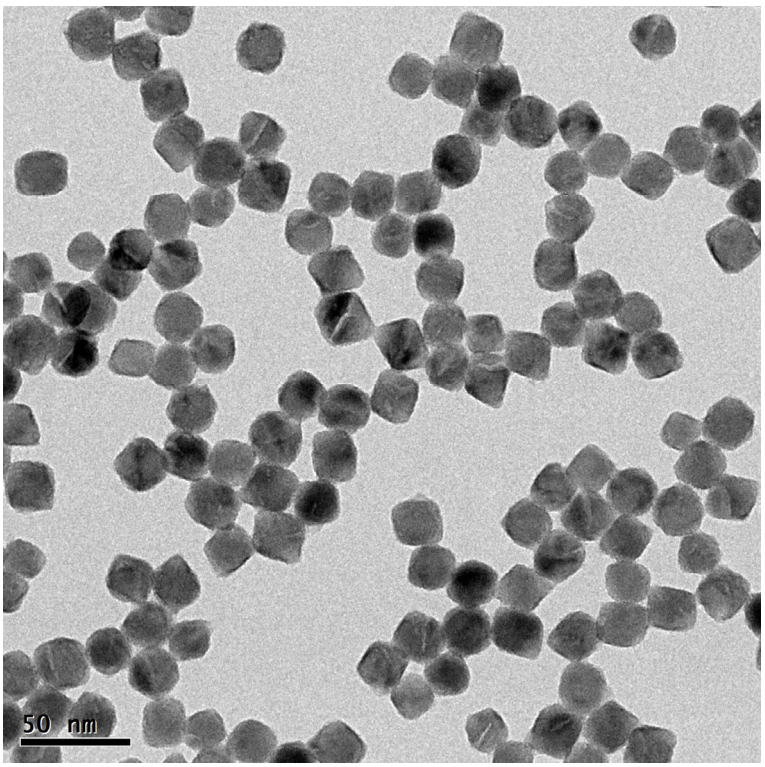

Figure S4. TEM image of ligand-free $LiLuF_4$:5%Ce, 5%Tb NPs, showing nearly identical particle size and morphology to their oleate-capped NPs.

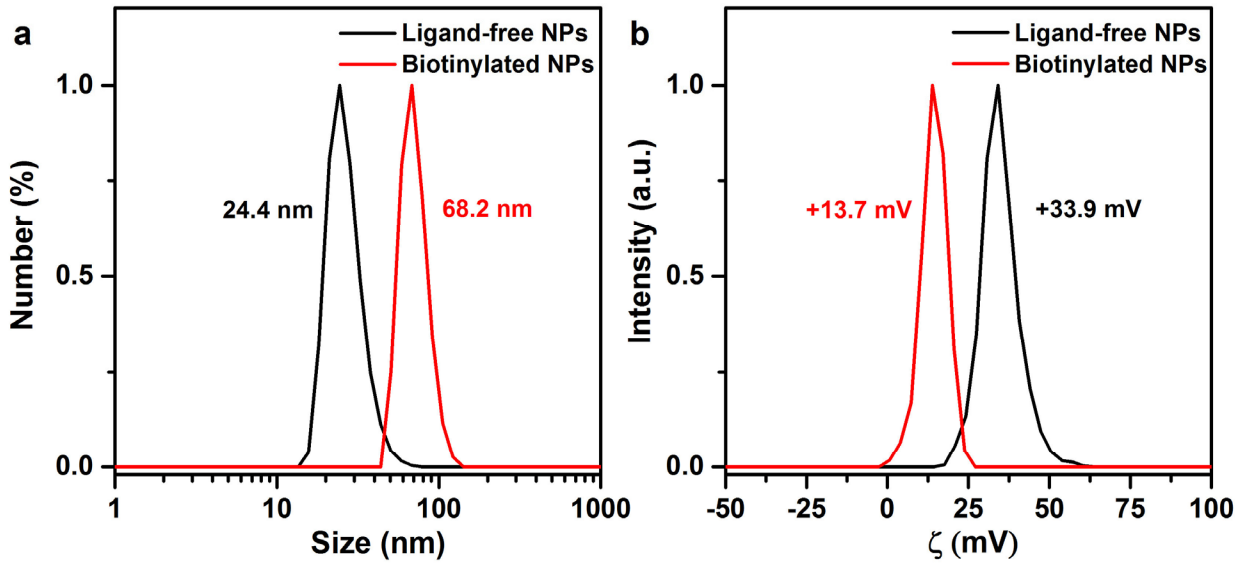

Figure S5. (a) Hydrodynamic diameter distributions and (b) ζ potentials of ligand-free and biotinylated $LiLuF_4$:5%Ce, 5%Tb NPs dispersed in aqueous solution (pH 6.9). The hydrodynamic diameter of the NPs increased from 24.4 nm in ligand-free NPs to 68.2 nm in biotinylated NPs due to the conjugation of biotin to the NP surface that may cause slight aggregation of the NPs. The ζ potential changed from +33.9 mV in ligand-free NPs to +13.7 mV in biotinylated NPs, as a result of reduced positively-charged Lu^{3+} ions exposed on the surface of the NPs.

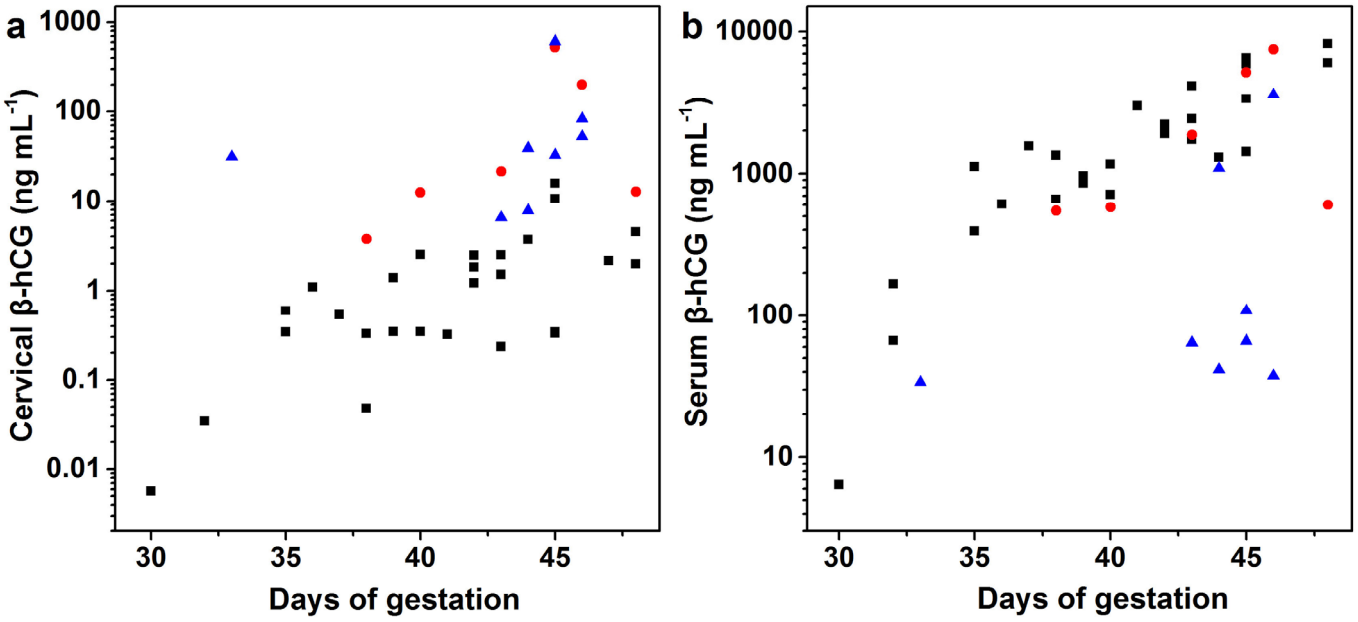

Figure S6. Distributions of the β-hCG levels in (a) cervical secretion and (b) serum samples of 42 pregnant women with days of gestation from 30 to 48 and pregnancy outcomes of viable intrauterine pregnancy (black square), miscarriage of intrauterine pregnancy (red circles), and ectopic pregnancy (blue triangle). The β-hCG levels in cervical secretion and serum samples were determined by the heterogeneous TRPL assay based on biotinylated $LiLuF_4$:Ce,Tb nanoprobes. In the viable group of intrauterine pregnancy, the β-hCG levels in cervical secretion were about 3–5 orders of magnitude lower than those in serum, while in the groups of miscarriage of intrauterine pregnancy and ectopic pregnancy, the β-hCG levels in cervical secretion increased significantly and were 1–2 orders of magnitude lower or even higher than those in serum.