Supporting Information

Multiplexed nucleic acid assay of SARS-CoV-2 via a lanthanide nanoparticles tagging strategy

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Apparatus

The detection of stable isotopes was performed by ICPMS using glass concentric atomizer, with collision ball chamber (NexION 350, PerkinElmer, Inc.). The working conditions of ICPMS were optimized as Table S1. An ELEMENT XR HR-ICPMS was used to quantify $^{31}$P (Thermofisher, Inc.). Hydration particle size characterization and zeta-potential of lanthanide nanoparticles and magnetic beads was implemented using a Malvern Zetasizer Nano ZS90 (Malvern PANalytical Ltd., Shanghai, China). The energy-dispersive spectrum (EDS) was processed by scanning electron microscopy (SEM, Hitachi, S3400). Transmission electron microscopy (TEM) with high-resolution TEM (HR-TEM) images was carried out by a JEM-2010 microscope.
(JEOL Co., Japan) at an accelerating voltage of 200 kV. The crystal structures of NaLnF₄ (Ln=Tb, Ho, Eu) were carried out utilizing an X’Pert pro X-ray diffractometer (XRD, Philips) equipped with Cu Kα₁ (λ = 1.5406 Å) radiation. Step scan mode was employed in the 2θ range of 10~80° with a step size of 0.02° and counting time of 0.12 s for each step (scan rate was 10° per min). The FT-IR spectrometer was done using IRTracer-100, Shimadzu Co.Ltd, China. A Mettler-Toledo ME104 microbalance was used for reagents weighing.

**Materials and reagents**

Sodium fluoride (NaF), sodium chloride (NaCl), sodium hydroxide (NaOH), nitric acid (HNO₃) were bought in Chengdu Kelong Chemical Reagent Company (China). Ethylene glycol (EG), N-ethyl-N’-(3-(dimethyl-amino)propyl) carbodiimide (EDC), N-hydroxy sulfo-succinimide (Sulfo-NHS) and polyacrylic acid (PAA) were purchased from Adamas Reagent, Ltd. (Shanghai, China). Bovine serum albumin (BSA), tris (hydroxymethyl) aminomethane (Tris-HCl), hydroxyethyl piperazine ethane sulfonic acid (HEPES), 2-morpholinoethanesulfonic acid (MES) and tris-EDTA buffer (TE buffer) were purchased from Shanghai Sangon Biotechnology Co. Ltd. (Shanghai, China). Terbium chloride hexahydrate (TbCl₃·6H₂O), holmium chloride hexahydrate (HoCl₃·6H₂O) and lutetium chloride hexahydrate (LuCl₃·6H₂O) were purchased from Aladdin Reagent Inc. (Shanghai, China). The enhancer solution containing containing 15 μM β-NTA, 50 μM TOPO, 1% acetic acid and 0.1% Triton X-100 was purchased from Wuxi Jiangyuan Industrial Technology and Trade Corporation. Dynabeads M-280 Streptavidin (SA-MBs) was purchased from Thermo Fisher Scientific Inc. Ultrapure water with 18.24 MΩ cm⁻¹ was obtained from a UPURE Sichuan water purification system. All the water used in the RNA hybridization process was nuclease-free under DEPC treatment.

All oligonucleotides used in this study (DNA and RNA oligo) were HPLC-purified and synthesized by Sangon Biotech Co., Ltd. (Shanghai, China). Their sequences are listed in Table S2.
RNA viral sample collection tubes (inactivated) and EZ-10 spin column viral total RNA extraction kit were purchased in Sangon Biotech Co., Ltd. (Shanghai, China).

A series of buffer was involved in this work, with details listed as follows:
- MES buffer: 20 mM MES in high-purity deionized water (pH 6.0)
- HEPES buffer: 10mM HEPES in high-purity deionized water (pH 7.4):
- TE buffer: (pH 6.0): 10 mmol L⁻¹ Tris-HCl and 1 mmol L⁻¹ EDTA in DEPC water (pH 8.0).
- TBE buffer (5×TBE): 450 mmol L⁻¹ Tris, 450 mmol L⁻¹ H₃BO₃ and 10 mmol L⁻¹ EDTA in DEPC water (pH 8.3).
- B&W buffer (2×B&W): TE buffer and 2 mol L⁻¹ NaCl in DEPC water (pH 8.0).
- TBS buffer: 25 mmol L⁻¹ Tris-HCl and 150 mmol L⁻¹ NaCl in DEPC water (pH 8.0).
- Tris-HCl buffer: 10 mmol L⁻¹ Tris-HCl and 150 mmol L⁻¹ NaCl in high-purity deionized water (pH 7.4). high-purity deionized water (pH 7.4).
- Blocking buffer 1: Washing buffer with 2% BSA (w/v).
- Blocking buffer 2: TBS buffer with 2% BSA (w/v).
- Solution A: 100 mmol L⁻¹ NaOH and 50 mmol L⁻¹ NaCl in DEPC water.
- Solution B: 100 mmol L⁻¹ NaCl in DEPC water.

**Experimental Details**

**Synthesis of -COOH functionalized NaLnF₄ (Ln=Tb, Ho, Eu) NPs**

To synthesis NaLnF₄ (Ln=Tb, Ho, Eu) nanoparticles, 1 mmol NaF was firstly dissolve in 16 mL (for Tb, Eu) /8 mL (for Ho) H₂O. Meanwhile,1mmol LnCl₃·6H₂O was dissolved in 24 mL (for Tb, Eu) /32 mL (for Ho) ethylene glycol to form lanthanides stock solution in a steel reactor. To this homogeneous stock solution, 1.8 g PAA was added and stir to dissolve. After that the NaF solution was added drop wisely and the transparent solution turned white and turbid. After continuing the reaction for 60 min, the liquid is transferred to a 100 mL autoclave with a filling degree of about 40%. Put the reaction kettle in the oven, 200 ℃ react for 4 hours. After the reaction is over, the reaction kettle is taken out and naturally cooled to room temperature. Then transfer
the solution to tubes for centrifugation. Discard the upper liquid, wash the precipitation with ethanol and water three times each, the precipitate was re-dissolved in 10 mL of water aiding with signification to form homogeneous dispersion and then to facilitate subsequent experimental operations.

**Preparation of Probe DNA-NaLnF\(_4\) NPs (O/R/E-Tb/Ho/Eu NPs).**

For DNA conjugation, 60 µL NaLnF\(_4\) -PAA was incubated with 0.02g EDC and 0.02g NHS in 500 µL MES buffer for 1 h to activate the carboxylic acid groups. Centrifuge and redissolve in 500 µL HEPES buffer solution, the activated NPs were then incubated with 1 OD NH\(_2\) modified probe DNA at RT for 12 h, followed by centrifugation and washing to obtain DNA functionalized NaLnF\(_4\) NPs.

**Preparation of Capture DNA-MBs (Capture O/R/E-MB).**

Firstly, RNase inactivation treatment was performed on SA-MBs: for 100 µL of 10 mg mL\(^{-1}\) SA-MBs, wash twice with Solution A and once with Solution B, then rinse with B&W buffer and then redispersed in 100 µL of B&W buffer. Subsequently, 100 µL of SA-MBs was mixed with 330 µL of 2 µmol L\(^{-1}\) biotin modified Capture O, Capture R and Capture E simultaneously. After the mixture was incubated at 37 °C for 1h with gentle shaking to immobilize DNA on the surface of SA-MBs via a specific binding between SA and biotin, an MBs-capture DNA probe was obtained. The MB-DNA probe was washed with B&W buffer and three times with Tris-HCl buffer to remove excess unreacted DNA.

**Recovery of viral RNA from real samples**

The real sample matrix of recovery experiment was obtained by the RNA viral sample collection tubes (inactivated) and EZ-10 spin column viral total RNA extraction kit: Three human throat swab samples were firstly collected. After sampling, these swabs were put into the tube containing preservative solution 1 mL avoiding contact with other parts. Break the swab tip, discard stick, and cap the tube containing samples. Each swab sample preservation solution was fully shaken on a vortex for half a minute, and then 0.2 mL was transferred to a 1.5 mL plastic centrifuge tube for the second step. Add 0.6 mL Buffer Rlysis-VG to the 0.2 mL sample obtained in the first step, shake for 30 s, mix well, and place at room temperature for 10 min. Add 0.6 mL of absolute ethanol,
cap the tube and vortex for 15 sec. After a short centrifugation, transfer 700 μL of the solution to a centrifugal adsorption column and leave it at room temperature for 2 minutes. Centrifuge at 12,000 rpm for 1 min at room temperature, discard the penetrant, and put the adsorption column back into the collection tube. Transfer the remaining solution to the centrifugal adsorption column and leave it at room temperature for 2 minutes. Centrifuge at 12,000 rpm for 1 min at room temperature, discard the penetrant, and put the centrifugal adsorption column back into the collection tube. Add 500 μL RPE solution to the centrifugal adsorption column, centrifuge at 12,000 rpm for 1 min at room temperature, discard the penetrant, and put the centrifugal adsorption column back into the collection tube. Repeat last step once. Centrifuge at 12,000 rpm for 2 minutes at room temperature. Discard the centrifuge tube containing the penetrant. Put the centrifugal adsorption column into a new self-prepared RNase-free 1.5 mL centrifuge tube and add 120 μL to the middle of the filter membrane of the centrifugal adsorption column DEPC-treated H2O, then let it stand at room temperature for 2 minutes. Centrifuge at 12,000 rpm at room temperature for 2 minutes. The sample in the collection tube at last was added certain amount of target RNA fragments. 20 μL spiked sample was added in each reaction system.
Supplementary Tables & Figures

**Table S1.** Operating parameters of ICP-MS

| Parameters                        | Settings         |
|-----------------------------------|------------------|
| RF power (W)                      | 1300             |
| Plasma gas flow (L/min)           | 18               |
| Auxiliary gas flow (L/min)        | 1.20             |
| Nebulizer gas flow (L/min)        | 0.90             |
| Vacuum pressure (Torr)            | $8.7 \times 10^{-7}$ |
| Sweeps per reading                | 120              |
| Sample uptake rate (L/min)        | 0.25             |
| Dwell time (ms)                   | 25               |
| Dead time (ns)                    | 35               |
| Element isotope monitored         | $^{153}\text{Eu}/^{159}\text{Tb}/^{165}\text{Ho}$ |
### Table S2. DNA and RNA sequences in this work

| Name          | Sequence (5’-3’)                                                                 |
|---------------|----------------------------------------------------------------------------------|
| ORF1ab        | CCAUAACCUUUCCACAUACCCGCAGACGG                                                     |
| RdRp          | GCAUCUCCUGAUGAGGGUCCACCUG                                                        |
| E             | CGAAGCGCAGUAAGGAUGGCUAGUGU                                                        |
| Probe O       | TGGAAAGGTTATGGGFFFFFFFFFFFFFFFFFFFFF-(CH₂)₆-NH₂                                 |
| Probe R       | CATCAGGAGATGCFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFH |
| Probe E       | TTACTGCGCTTGGFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFH |
| Capture O     | biotin-FFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFH |
| Capture R     | biotin-FFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFH |
| Capture E     | biotin-FFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFFH |
| SM ORF1ab     | CCAUAUCCUUUCCACAUACCGCAGACGG                                                    |
| TM ORF1ab     | CCAUAUGGUUUUCCACAUACCGCAGACGG                                                    |
| SM RdRp       | GCAUCAACCUGAUGGGUCCACCUG                                                        |
| TM RdRp       | GCAUCAAGUGAUGGGUCCACCUG                                                        |
| SM E          | CGAAGGCCAUCAGGAUGGCUAGUGU                                                      |
| TM E          | CGAAGGCCAGUAGGAUGGCUAGUGU                                                      |
| Random 1      | UUGUACUACACAAAGUACUG                                                           |
| Random 2      | CAA TTG AGG ATC CAG TTT TAG CAA AGA A                                           |
Figure S1. Calibration curve of label elements (a) $^{159}$Tb, (b) $^{165}$Ho and (c) $^{153}$Eu detected by ICPMS.
Figure S2. XRD patterns of NaTbF$_4$, NaHoF$_4$ and NaEuF$_4$ NPs. The vertical lines represent the standard pattern of hexagonal-phase NaTbF$_4$ (PDF#27-0809), hexagonal-phase NaHoF$_4$ (PDF#49-1896), hexagonal-phase NaEuF$_4$ (PDF#49-1897).
Figure S3. XPS spectrum of the as-synthesized NaTbF$_4$, NaHoF$_4$ and NaEuF$_4$ NPs.
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