

Review

# Research Progress on Up-Conversion Fluorescence Probe for Detection of Perfluorooctanoic Acid in Water Treatment

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**Abstract:** Perfluorooctanoic acid (PFOA) is a new type of organic pollutant in wastewater that is persistent, toxic, and accumulates in living organisms. The development of rapid and sensitive analytical methods to detect PFOA in environmental media is of great importance. Fluorescence detection has the advantages of high efficiency and low cost, in which fluorescent probes have excellent fluorescence properties, excellent bio-solubility, and remarkable photostability. It is necessary to review the fluorescence detection routes for PFOA. In addition, the up-conversion of fluorescent materials (UCNPs), as fluorescent materials to prepare fluorescent probes with, has significant advantages and also attracts the attention of researchers, however, reviews related to their application in detecting PFOA and comparing them with other routes are rare. Furthermore, there are many strategies to improve the performance of up-conversion fluorescent probes including SiO<sub>2</sub> modification and amino modification. These strategies can enhance the detection effect of PFOA. Thus, this work reviews the types of fluorescence detection, the design, and synthesis of UCNPs, their recognition mechanism, properties, and their application progress. Moreover, the development trend and prospects of these detection probes are given.

**Keywords:** PFOA; fluorescence detection; fluorescent probe; rare earth up-conversion fluorescent materials



**Citation:** Mao, T.; Shi, X.; Lin, L.; Cheng, Y.; Luo, X.; Fang, C. Research Progress on Up-Conversion Fluorescence Probe for Detection of Perfluorooctanoic Acid in Water Treatment. *Polymers* **2023**, *15*, 605. <https://doi.org/10.3390/polym15030605>

Academic Editors: Nahum Medellín-Castillo, Juan Carlos Moreno-Piraján and Liliana Giraldo

Received: 28 November 2022

Revised: 7 January 2023

Accepted: 16 January 2023

Published: 24 January 2023

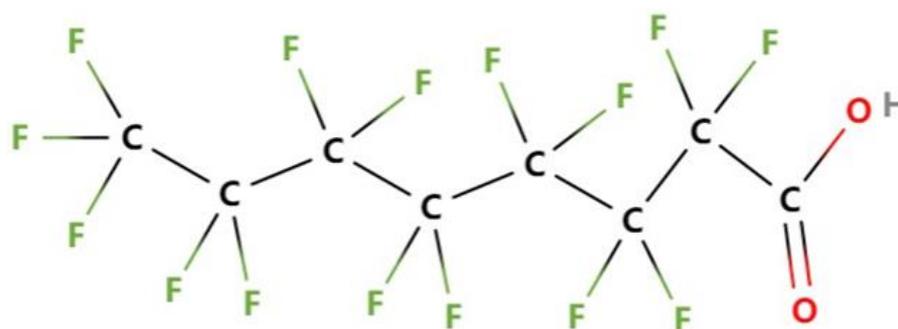


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## 1. Introduction

The chemical formula of PFOA is C<sub>8</sub>HF<sub>15</sub>O<sub>2</sub> and its structure is shown in Figure 1. PFOA contains 15 F atoms and 8 C atoms. The C-F bond of PFOA has strong polarity and bond energy. Fluorine surfactants of it has a higher surface activity, stronger hydrophobicity, and oleophobicity than traditional surfactants. They are widely used in the chemical, textile, leather, paper making, and cosmetics industries. The fluorinated surfactants of PFOA have the strongest surface activity and the most extensive application. PFOA is hard to degrade because of its strong chemical stability and biological inertness, and can withstand strong light, heat, chemistry, microbial action, and has a high biological metabolism. Therefore, PFOA has bioaccumulation and toxic effect, which will threaten the ecosystem and human health.

PFOA is the main active ingredient of waterproof, oil-proof, and stain-proof finishing agents, which were used in the coatings of various special garments, fabrics, and carpets. Adding PFOA into the coating can improve the wettability, the dispersibility, the uniformity of the color carrier, and prevent caking. The most familiar one is the “Non-stick pan” with a polytetrafluoroethylene (PTFE) surface coating. PFOA is one of the processing aids of PTFE [1]. Due to the applications of PFOA in many fields, PFOA enters the rivers, lakes, and even into drinking water. Li's group [2] found that PFOA levels in many sewage discharges in Chongqing seriously exceeded the standard and was extremely difficult to deal with. Wang's group [3] found that the contents of PFOA and PFOS in drinking water in some areas were too high. PFOA is accumulative and transferable, so it can be found in the air, soil, and water [4–7]. This will increase the possibility of PFOA entering the body.



**Figure 1.** The chemical structure of PFOA.

The damage of PFOA to the human body is enormous. PFOA can enter the human body through the skin surface to cause PFOA to accumulate in the body, which may cause cancer. Sun's group [8] found that exposure to PFOA leads to the accumulation of ROS in BRL-3A cells, which ultimately leads to the death of the experimental subject. In addition, PFOA has an impact on the human stomach, liver, and nervous system, even leading to cognitive abnormalities [9]. It also affects human sperm [10].

In 2006, the Environmental Protection Agency (EPA) of the United States proposed a reduction plan for PFOA [11]. In 2014, the Norwegian Environmental Protection Agency issued a national ban on the use of PFOA products. In 2016, the United States Toxic Substances Control Act (USTSCA) included PFOA on the list of toxic chemicals [12]. In 2015, China generated large quantities of fluoropolymers, and the consumption of PFOA reached 200 tons [13,14]. Frederick Pontius [15] analyzed in detail the requirements of PFOA and PFOS of various international organizations in recent years, and the regulatory agencies of more than 12 countries formulated the consultation of PFOA in drinking water or groundwater. PFOA was included on the third drinking water contaminant candidate list (CCL). US-EPA restricted the content of PFOA in healthy drinking water at 70 ng/L. PFOA has attracted worldwide attention since 2016. The requirements for PFOA content in drinking water in China are still improving [16–18].

The detection of PFOA has a great social significance to people's lives, but the effectiveness of the detection methods is different. Therefore, the simplest, quickest, and most sensitive methods need to be explored [19,20].

## 2. Method of Detecting PFOA

Traditional methods of detecting PFOA include chromatography, mass spectrometry, and chromatography–mass spectrometry. These methods often need to use expensive instruments to detect due to the unique properties of PFOA [21–25]. The fluorescence detection method is a new way to detect trace elements, and PFOA as a trace element can be detected by it.

### 2.1. Liquid Chromatography–Mass Spectrometry

#### 2.1.1. High Performance Liquid Chromatography–Tandem Mass Spectrometry (HPLC–MS/MS)

High performance liquid chromatography (HPLC) has the advantages of high accuracy, a wide separation range, and low destruction to the material structure. Figure 2 shows the detection flow chart of HPLC. The sensitivity of mass spectrometry (MS) is better than any other analytical method, it is more accurate in characterizing the structure of unknown compounds. Mass separation–mass spectra Characterization (MS–MS) allows further cleavage of the parent atom to obtain information on the cleavage process and molecular structure, often called as tandem mass spectrometry. HPLC–MS/MS combines the advantages of both. The chromatography can be used as a sampling system for the mass spectrometry, and the mass spectrometry as an identifier for the chromatography. HPLC–MS/MS has certain advantages in terms of selectivity and sensitivity [26,27].

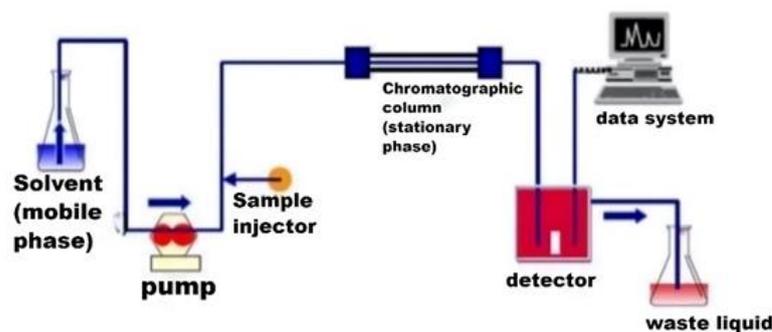


Figure 2. Detection flow chart of HPLC.

### 2.1.2. High Performance Liquid Chromatography–Quadrupole–Time of Flight–Mass Spectrometry (HPLC–Q–TOF–MS)

The mass analyzer used for HPLC–MS/MS is a low-resolution mass spectrometer, it cannot effectively distinguish the interference when analyzing complex samples. Time of flight-mass spectrometry (TOF/MS) has certain advantages in terms of mass range and resolution, and it has a high mass accuracy and a fast analysis speed. Unlike low-resolution mass spectrometry, TOF/MS greatly improves the anti-interference ability in complex backgrounds and makes the detection results more accurate and reliable [28]. Therefore, HPLCQ–TOF–MS has a certain optimization to analyze samples when in a complex environment, increasing the correctness of detection [29,30].

In 2009, USEPA issued Method 537 to analyze 14 PFAS in drinking water. This method needs to use solid-phase extraction and liquid chromatography/tandem mass spectrometry (LC/MS/MS) [15]. However, this method needs skilled analysts and expensive instruments to get effective results.

## 2.2. Gas Chromatography–Mass Spectrometry

### 2.2.1. GC–MS

Because the partition coefficient of the sample in the chromatographic is different when it is in the gas or solid phase state. Therefore, GC–MS uses this principle to complete detection by multiple distributions of the sample. In GC–MS, the gas chromatography is used as the sampling system of mass spectrometry, while the mass spectrometer is used for gas chromatography detection. However, the GC–MS detection process is more cumbersome and therefore less used in applied methods [31].

### 2.2.2. Pre-Column Derivatization–Gas Chromatography

Pre-column Derivatization–Gas chromatography is an improvement on the shortcomings of GC–MS. The derivatization technology can reduce the temperature of the target analyte and improve the signal required to detect the target, which reduces the cost of the detection instrument and can effectively detect PFOA [32], such as silanization, esterification, and acylation [33,34].

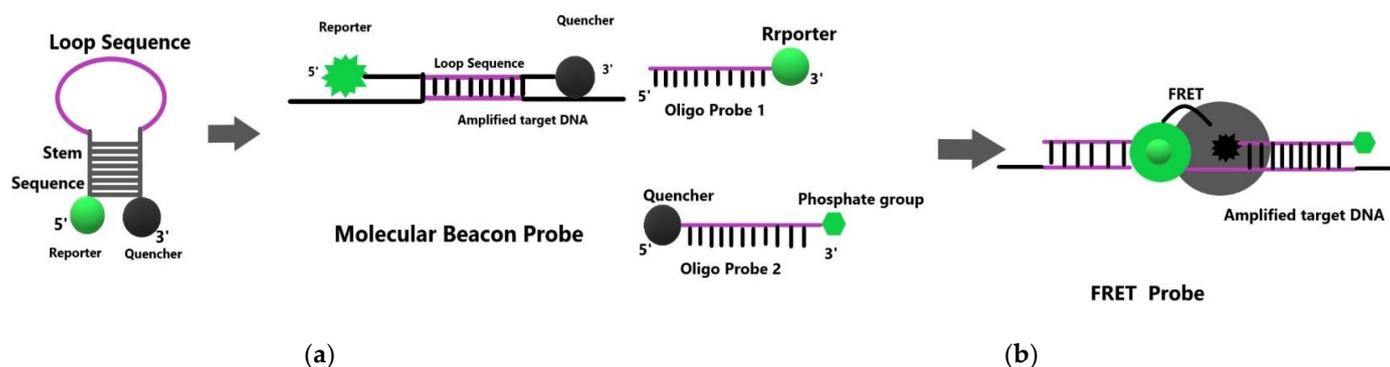
## 2.3. Fluorescence Detection

Traditional methods of detecting PFOA have certain advantages, but still have the shortcomings of high cost and low universality. Fluorescence detection is the latest in current detection methods, which can detect target substances conveniently and quickly. Zheng’s group [35] sensitively detected perfluorinated pollutants through fluorescence detection.

The fluorescent probe uses fluorescent substances as indicators, and under the excitation of a certain wavelength of light, the indicator produces fluorescence. Then, the change of fluorescent strength is detected to achieve a qualitative or quantitative analysis of the detected substances. The signal emitted by the reporter fluorophore is absorbed by the quenching fluorophore when the probe is intact, but the two are separated when the

probe detects the substance. In other words, the luminescent substance at the top of the fluorescent probe will specifically bind to the detection target when the fluorescent probe reacts with the detection substance, which can cause a fluorescence quenching reaction.

The probe can be divided into a Beacon probe and a FRET (fluorescence resonance energy transfer) probe. Beacon mode depends on the molecular beacon accumulating fluorescence in the presence of enzymatic digestion and cannot show the detection status in time, as shown in Figure 3a. However, the detection signal of the FRET mode is a real-time signal, which can display the detection status in time, as in Figure 3b. Therefore, the FRET mode will be selected when detecting trace elements. The FRET probe includes fluorescein probes, inorganic ion fluorescent probes, fluorescent quantum dots, and molecular beacons [36].



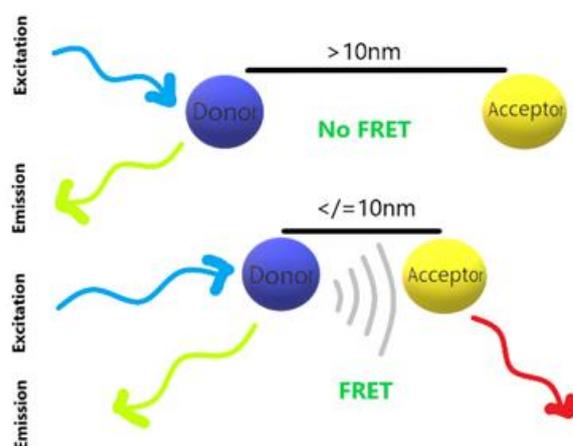
**Figure 3.** (a) Detection principle of Beacon probe (b) Detection principle of FRET probe.

Fluorescence quenching can lower the intensity of emitted light from fluorescent molecules. Fluorescence quenching can cause some changes including fluorescence intensity, related excitation peak changes, and fluorescence peak position changes. The substances react with fluorescent materials causing these changes and these processes are called fluorescence quenchers. For example, electron acceptors are fluorescence quenchers in photosynthesis.

Fluorescence quenching can be divided into dynamic quenching and static quenching. Dynamic quenching occurs when the fluorescence quenching between the excited state fluorescent molecule and the quencher will cause an energy transfer or physical collision. However, for the excited state molecule, it is not necessary to complete the direct contact between the two acting molecules. They can create optical collision effects directly. Static quenching refers to the formation of a compound between the ground state fluorescent molecule and the quencher through weak binding, and the compound will completely quench the fluorescence.

The principle of fluorescence quenching is the result of fluorescence resonance energy transfer (FRET) [37–39]. As shown in Figure 4, FRET is a non-radiative process formed by the interaction between dipoles and dipoles, and the energy will be generated when donor and acceptor close to each other.

A small molecule fluorescent probe is widely used to detect trace elements. It has the advantages of low cost, simplicity of handling, and a high detection sensitivity. A fluorescent probe made of fluorescent materials has the advantages of good hydrophilicity and multifunctional integration. It plays a significant role in detecting trace elements. A detailed comparison can be seen in Table 1.



**Figure 4.** Fluorescence resonance energy transfer (FRET).

**Table 1.** Comparison table of fluorescent probes.

Category	Advantages	Detection Limit	References
Small molecule fluorescent probes	Small molecule fluorescent probes have significant advantages such as high sensitivity, good membrane permeability, real-time in situ analysis, minimal biological damage, and easy handling.	$1.05 \times 10^{-8} \sim 5.8 \times 10^{-7}$ mol/L	[40,41]
Polymeric fluorescent probes	It has a long fluorescence lifetime, good biocompatibility, and high quantum yield. It can also enhance intramolecular electron transport and increase the sensitivity of the probe.	0.17~2.3 $\mu\text{g/mL}$	[42,43]
Fluorescent probes based on nanomaterials	Compared with traditional fluorescent dyes, fluorescent nanomaterials not only have high fluorescence intensity and good light stability, but also have the characteristics of small size effect, quantum effect, and surface effect that are unique to nanomaterials, which can make up for the shortcomings of traditional fluorescent dyes.	$6.4 \times 10^{-10} \sim 1.225 \times 10^{-8}$ mol/L	[44,45]

Fluorescent sensors use materials that have a fluorescence effect to label the detectors and detect substances through changes in the fluorescence signal. Fluorescent sensors have the advantages of high sensitivity and specificity. Kelsey L. Rodriguez's group [46] analyzed in detail the various drawbacks of the conventional detection of PFASs such as GS-MS and so on, and analyzed the ability of various sensors to detect PFAs. Table 2 is a summary of the sensors involved in it. The advantages and disadvantages of HPLC-MS/MS, HPLC-Q-TOF-MS, GC-MS, and Pre-column Derivatization GC are also compared in Table 2.

After comparing the detection methods, it was finally found that the fluorescence detection method was the most convenient and efficient, and the fluorescence probe method was more suitable to detect PFOA in wastewater.

As can be seen from Table 2, the detection effect of HPLC-MS/MS, HPLC-Q-TOF-MS, and GC-MS are not ideal and the cost is high. The detection effect of fluorescent sensors and other methods is good, in which the optical sensor is simple to operate, but the detection performance of rare earth up-conversion fluorescent probes is better.

**Table 2.** Advantages and disadvantages of detection methods.

Category	Advantages	Detection Limit	References
Electrochemical Sensors	Electrochemical sensors can be used to detect chemical and biological compounds. Molecularly imprinted polymers (MIPs) functionalized sensors are effective in detecting PFASs, but may have the disadvantage of long detection times and high costs, as well as the inability to differentiate between similar molecules.	0.07~1.0 µg/L	[47–49]
Fluorescence Sensors	Fluorescent sensors occurred fluorescence quenching because of electron transfer between fluorescent dyes and PFASs, which can easily be detected using fluorescence spectrophotometers. However, this method can increase costs and cause environmental hazards	2.5 ppt~120 ppb	[50,51]
Optical Sensors/ Colorimetric sensors	Optical sensors/ Colorimetric sensors usually use organic dyes and can be detected by the naked eye based on visible color changes. Although it is not overly dependent on fluorescence spectrophotometry, co-existing atoms can interfere with the sensor. In addition, they require a good platform to improve sensor availability.	1 ppb~5 ppt	[52,53]
HPLC–MS/MS	HPLC–MS/MS is based on the principle of using liquid chromatography monopole mass spectrometry or multistage mass spectrometry as the separation and detection system. It has the advantages of low detection limits, high resistance to matrix effect interference, convenience, and rapid detection. The detection limit is approximately 0.07~0.15 µg/kg.	The method is expensive, costly, and highly specialized and requires a high level of operator expertise	[54–57]
HPLC–Q-TOF-MS	HPLC–Q-TOF-MS has a certain optimization in the ability to analyze samples when in a complex environment, which increase the correctness of detection. The detection limit is approximately 20~30 ng/L.	The method is too costly to be suitable for universal detection.	[28]
GC–MS	GC–MS is a tandem gas chromatograph and mass spectrometer method that offers the advantages of simplicity and convenience. The detection limit is approximately 15~40 ng/L.	PFOA is less volatile and needs to be derivatized before it can be detected by injection. The application of GC–MS is narrow, and the experimental process is accompanied by toxic substances.	[58–60]
Pre-column Derivatization GC	It is a method that improves on the shortcomings of GC–MS. The derivatization technique reduces the temperature of the target, and improves the signal required to detect the target. The detection limit is approximately 0.01~0.05 mg/kg.	The formation of by-products may cause greater difficulties in chromatographic separation. Impurities or interfering peaks can easily be introduced or samples lost during derivatization.	[34]
Rare earth up-conversion fluorescent probe	Fluorescence burst reaction of the probe with the detected substance, offering the advantage of simple, efficient, and highly sensitive detection. The detection limit is approximately 0.01~1.2 nmol/L.	PFOA is a perfluorinated compound. The method is not selectively detectable for PFOA	[61]

### 3. Fluorescent Materials

#### 3.1. Fluorescent Polymer Materials

Fluorescent polymers are prepared by bringing small molecular fluorescent compounds into the side chains and segments of polymers, or using the polymerization of fluorescent functional monomers. Since 1960, the research on fluorescent polymers has included materials science, medicine, and chemistry.

Fluorescent polymers can be used as conversion materials to convert chemical signals into visible electrical or optical signals, making it easier to detect trace elements. In the detection of organic pollutants or small organic molecules, fluorescent polymers can detect trace polymers by FRET. Chen's [62] group established a two-in-one platform based on conjugated polymer, which can effectively detect perfluoroalkyl substances. The polymer has the advantages of easy preparation and modification. The sensor made of fluorescent polymer has a superior sensing ability. For example, Vinh Van Tran [63] launched a conjugated polymer-based biosensor to solve the challenge of rapid virus detection based on COVID-19, but there are shortcomings. Akhtar Hussain Malik's group [64] prepared conjugated polymer nanoparticles that can detect nitro-explosive picric acid on multiple platforms. Chen's group [65] reviewed smart conjugated polymers for optical detection. However, there are shortcomings in the separation and analysis method.

Fluorescent polymers can be divided into water-insoluble fluorescent polymers, water-soluble fluorescent polymers, and amphiphilic fluorescent polymers. Water-insoluble fluorescent polymers have properties of high strength, easy film formation, solvent resistance, heat resistance, and easy processing. In the fluorescence detection of molecules or atoms which need to be performed in a biological environment, water-soluble fluorescent polymers show advantages in this respect [66,67]. The amphiphilic fluorescent polymers have the characteristic that the lipophilic part is incompatible with the hydrophilic part in the structure, which makes microphase separation easy. In addition, amphiphilic fluorescent polymers have fixed fluorescence emissions [68,69].

#### 3.2. Rare Earth Up-Conversion Nanomaterials

Rare earth up-conversion nanomaterials (UCNPs) have rich 4f energy levels, in which electrons can transition, so they have rich optical properties and are widely used in optical, electrical, and magnetic fields [70,71]. Up-conversion luminescence is the absorption of low-energy (long-wavelength) photons and the emission of high-energy (short-wavelength) photons, which can also be called a phenomenon of photoluminescence. The famous Stokes theorem shows that materials can only be excited by high-energy light (short wavelength) and emit low-energy light (long wavelength). However, up-conversion luminescence reverses the knowledge of Stokes theorem, so it is called anti-Stokes luminescence [72].

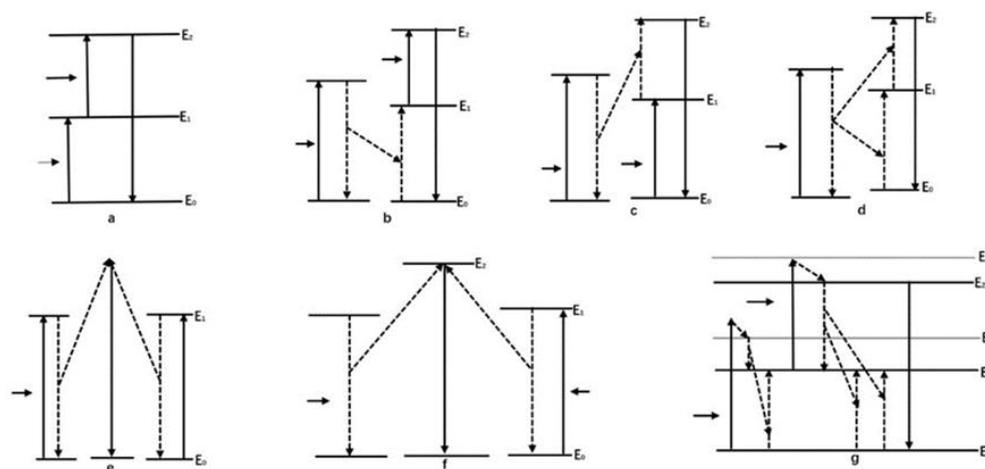
In the mid-1960s, Auzel's group [73,74] discovered up-conversion luminescence. The study found that adding activators and sensitizers to up-conversion materials could greatly improve the luminescence efficiency of UCNPs. The fluorescent probe prepared by UCNPs has better luminescence effect and stable chemical properties [75,76], and does not undergo photochemical changes resulting in photobleaching. Therefore, the up-conversion fluorescent probe is a new way to study.

##### 3.2.1. Up-Conversion Fluorescence

The luminescence principle of UCNPs can be divided into four situations: the excited state absorption up-conversion process, the energy transfer up-conversion process, the cooperative up-conversion process, and the photon avalanche up-conversion process.

The excited state absorption up-conversion process refers to how an ion in the ground state  $E_0$  absorbs a low-energy photon and is excited to a metastable  $E_1$ . Then, it absorbs a low-energy photon again and is excited to a higher excited state  $E_2$ . Finally, it transitions from  $E_2$  to  $E_0$  and radiates high-energy photons. (Figure 5a). In other words, the excited state absorption process is a continuous multiphoton absorption process of a single ion. The same rare earth ion transits from  $E_0$  to  $E_2$  through absorbing two-photons or multi-photon

absorption, and then releases the energy back to  $E_0$  in the form of light radiation [77]. The excited state absorption up-conversion process is the most basic process of up-conversion fluorescence. However, the process requires a small concentration of doped atoms to reduce the energy loss of energy transfer between atoms.



**Figure 5.** Principle diagram of up-conversion fluorescence. (a) The excited state absorption up-conversion process. (b) The energy transfer up-conversion process. (c) The relaxation up-conversion process. (d) The continuous energy transfer up-conversion process. (e) The cooperative luminescence up-conversion process. (f) The cooperative sensitization up-conversion process. (g) The photon avalanche up-conversion process.

The energy transfer up-conversion process uses the excitation light to excite the sensitizer atoms, prompting them to transition from the ground state to the excited state. The electrons in the excited state have a transition down to the lower-energy excited state or back to the ground state, transferring the released energy to the activated ion in the form of a non-radiative FRET, which excites the activated ion to a higher energy state and finally transitions to the ground state producing a radiative transition. Therefore, this process is in line with the luminescence principle of up-conversion fluorescent probes. The energy transfer up-conversion process can be divided into the energy transfer up-conversion process (Figure 5b), the relaxation up-conversion process (Figure 5c), and the continuous energy transfer up-conversion process (Figure 5d). The process requires that the concentration of rare earth atoms doped is high enough to ensure the effectiveness of energy transfer.

The cooperative up-conversion process can get to the high-energy state directly, and then radiate short-wavelength photons. It can be divided into the cooperative luminescence up-conversion process (Figure 5e) and the cooperative sensitization up-conversion process (Figure 5f). The photon avalanche up-conversion process is a process of using ground state and excited state absorption to rapidly increase the number of intermediate sub-stable atoms. Then, the activator achieves a high-energy state particle number layout, and finally has a transition to the ground state radiation photon process, as shown in Figure 5g [78].

UCNPs are mainly composed of activator, sensitizer, and substrate. UCNPs have the advantages of strong photostability, low toxicity, and no damage to biological tissues during use. They can accurately, stably, and safely detect target substances [79].

### 3.2.2. Composition of UCNPs

#### (1) substrate

In the energy level transition of the up-conversion luminescence, the substrate material mainly provides the appropriate reaction environment for the activator and sensitizer. The up-conversion luminescence intensity will be improved accordingly when using the substrate material which has better properties because rare earth atoms with 4f level

spacing are smaller than other elements. Therefore, the phonon energy of the substrate material must be lower, therefore fluoride and oxide are commonly used. Up-conversion luminescence has a large anti-Stokes displacement. The material needs an intermediate or metastable energy level, which has a long lifetime. At the same time, it also needs a strong excitation light source to realize up-conversion luminescence. The transition between energy levels of rare earth ions to F-F forbidden resistance makes the metastable energy levels of the luminescence center have a long energy level lifetime. This can realize the two-photon or multi-photon effect. In other words, up-conversion materials need more metastable energy levels and stronger excitation light sources for luminescence. The substrate of up-conversion luminescent materials is relatively special, which can reduce the phonon energy. Finally, it can form a large number of metastable energy levels to improve the fluorescence intensity of up-conversion luminescent materials. Therefore, fluoride is usually chosen as the substrate material [80]. NaYF<sub>4</sub> is one of the highest luminescence efficiency substrates used as up-conversion nanomaterials [81]. NaGdF<sub>4</sub> not only meets the luminescence efficiency and has paramagnetic properties, but also can be used as a multifunctional contrast agent [82,83].

#### (2) activator and sensitizer

The activator with less content in UCNPs can make the basic non-luminescent material emit strong light, and can also change the luminescent color and luminescent efficiency of UCNPs. The selection of activators should meet the conditions of more energy level transitions, the full use of infrared light, and the energy exchange with other atoms. The most commonly used activators are Er<sup>3+</sup>, Tm<sup>3+</sup>, and Ho<sup>3+</sup>. Their unique energy level structure makes UCNPs emit emission wavelength, while the sensitizer usually uses Yb<sup>3+</sup>. Yb<sup>3+</sup> can enhance the absorption intensity of rare earth atoms to excited light due to its simple energy level structure, and it can enhance the absorption intensity of rare earth atoms for excitation light. The sensitizer absorbs the energy of photons and transfers the energy to the activator [84,85].

Nie's group [86] used first principles to study and calculate the luminescence rate of UCNPs prepared by different fluoride substrate materials and found that β-NaGdF<sub>4</sub> has the best luminescence rate among the three substrates by comparing β-NaYF<sub>4</sub>, β-NaGdF<sub>4</sub>, and β-NaLuF<sub>4</sub>, as shown in Figure 6. Fan's group [87] carried out further research on β-NaGdF<sub>4</sub>. It was doped with three kinds of systems to make three types of photonic crystals, and their structural properties and optical properties were simulated and compared. They finally found that the optimal preparation was β-NaGdF<sub>4</sub>: Er<sup>3+</sup>/Yb<sup>3+</sup>. Therefore, the luminescent properties of UCNPs obtained from different substrates and activators are also different.

PFOA has rich F atoms, which can make the up-conversion fluorescent probe occur fluorescence quenching by F-F interaction, so as to achieve the purpose of detection. In the detection of Perfluorochemicals, it is found that Ho<sup>3+</sup> can better match Yb<sup>3+</sup> to enhance the luminescence intensity of UCNPs [88–90].

Under 980 nm excitation, the probe produces three emission peaks, in which 541 nm emission from the <sup>5</sup>S<sub>2</sub>-<sup>5</sup>I<sub>8</sub> transition of Ho<sup>3+</sup>, 649 nm emission from the <sup>5</sup>F<sub>5</sub>-<sup>5</sup>I<sub>8</sub> transition of Ho<sup>3+</sup>, and 750 nm emission from the <sup>5</sup>S<sub>2</sub>-<sup>5</sup>I<sub>7</sub> transition of Ho<sup>3+</sup>, as shown in Figure 7. The emission yield of rare-earth up-conversion fluorescent probes can reach 90% and fluorescence quantum yields can reach 5.6%, while core-shell fluorescent probes can reach 95%. The emission yield of rare-earth up-conversion fluorescent probes can be enhanced using functionalized modifications. Table 3 shows a comparison of the fluorescence quantum yields of the different fluorescent probes.

Comparing several fluorescent materials, UCNPs are found to have the best luminescence and do not cause secondary pollution to the environment during the detection process.

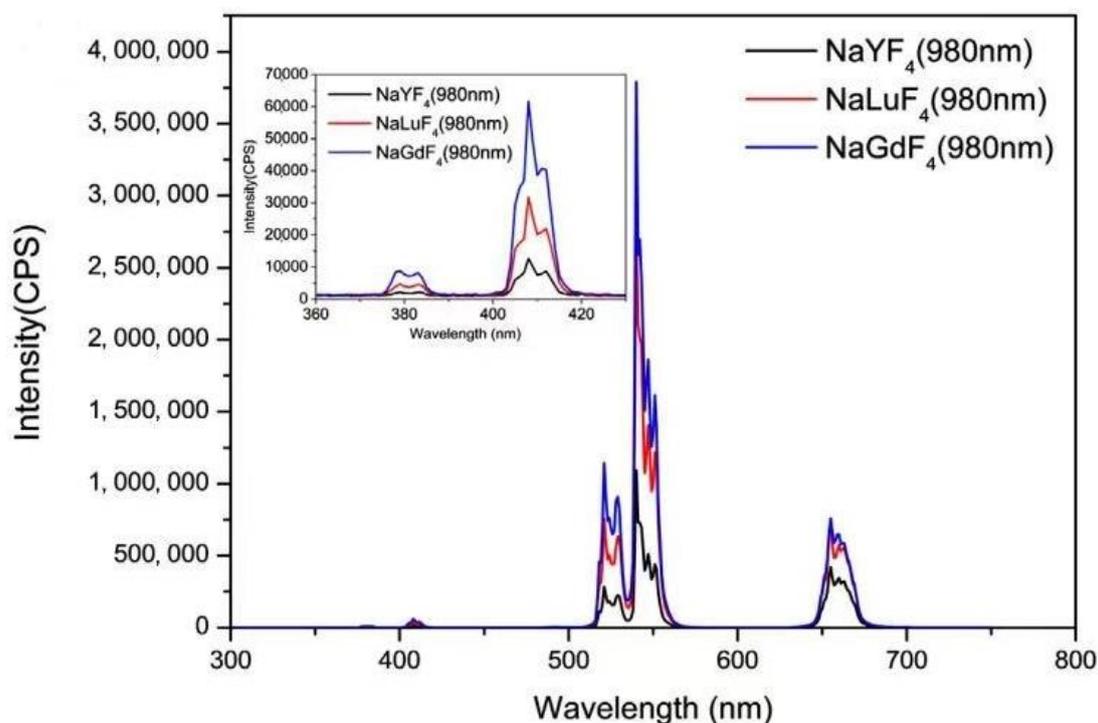


Figure 6. Fluorescence peak of different substrates. Adapted with permission from Nie J et al. [86].

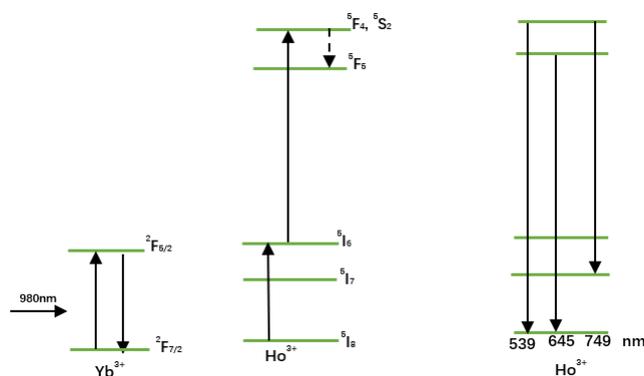


Figure 7. Energy level diagram and energy transfer mechanism of  $\text{Yb}^{3+}$  and  $\text{Ho}^{3+}$ .

Table 3. Fluorescence quantum yields of fluorescent probes.

Fluorescent Probe	Quantum Yields	Reference
A highly selective $\text{Al}^{3+}$ fluorescent probe	5.7% (366 nm)	[91,92]
Near-infrared $\text{H}_2\text{S}$ fluorescent probes	1.2% (530 nm)	[93]
Enhanced fluorescent probes for BSA	1.5% (645 nm)	[94]
Visualization of iron ion fluorescent probes	2.1% (610 nm)	[95]
Rare-earth up-conversion fluorescent probes	4.7% (980 nm)	[96]

## 4. Preparation and Optimization of UCNPs

### 4.1. Method of Preparing UCNPs

#### (1) Hydrothermal method

The hydrothermal method can be used in a special sealed high-pressure vessel, such as a vacuum reactor, as shown in Figure 8. This method makes compounds that are difficult to dissolve or cannot be dissolved, and finally generates synthetic nanomaterials with water

as the reaction system under high temperature and high pressure. The preparation process of the hydrothermal method is easy to control. The crystallized grains are good and the size of the nanoparticles is uniform [97,98]. Zhang's group [99] proposed a general strategy for synthesizing nanocrystals in hydrothermal processes using liquid–solid–solution phase transfer and separation, which can also be considered in the preparation process. However, the reaction time of hydrothermal method is too long. It can be slightly improved by increasing the ion concentration correspondingly [100,101].

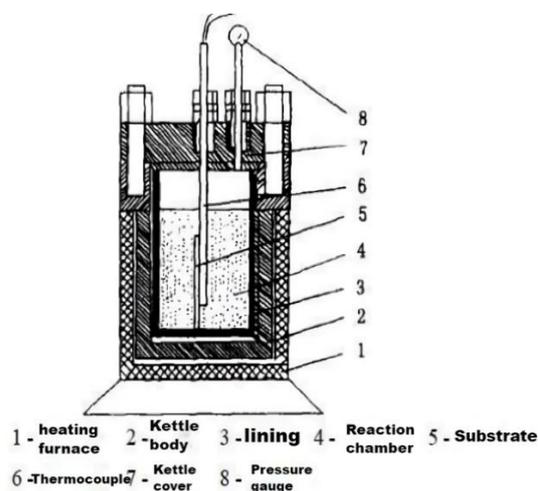


Figure 8. Hydrothermal reactor.

## (2) Solvothermal method

The solvent method is one of the most commonly used methods to prepare UCNPs, which can be used to synthesize lanthanide tetrafluoride UCNPs, such as  $\text{NaYF}_4$ ,  $\text{NaGdF}_4$ , and  $\text{NaLuF}_4$  [102–104]. The solvothermal method is similar to the hydrothermal method, but the difference lies in their precursor solution. The solvothermal method uses non-aqueous organic solvents, which can greatly improve the activity of reactants under the set pressure. UCNPs prepared by it have good crystallization, good particle dispersion, dispersion, and small particle size [105]. Figure 9 is an SEM image of  $\text{Bi}_2\text{O}_3$  prepared by the solvothermal method [106,107]. It can be shown that the crystallinity of nanoparticles produced by the solvothermal method is good, but this method has some experimental risks.

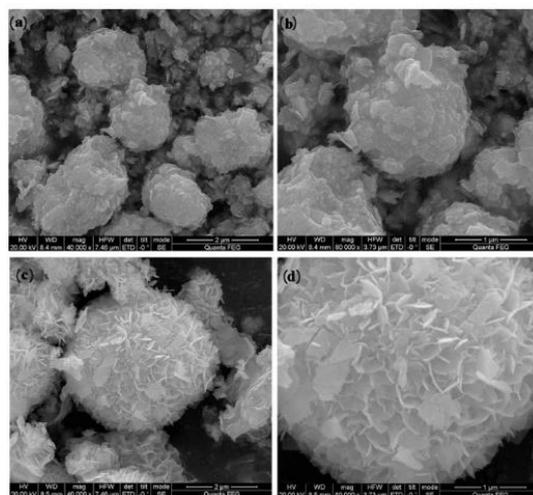


Figure 9. SEM images of  $\text{Bi}_2\text{O}_3$  (a,b); 1:1-Bi/BiOBr (c,d). Adapted with permission from Yuan XY et al. [106].

### (3) Coprecipitation method

The coprecipitation method refers to the adding of the corresponding precipitant into the solution containing two or more cations and finally producing the target substance through a precipitation reaction. Figure 10 shows the coprecipitation device. NaF,  $\text{NH}_4\text{F}$ , and other precipitating agents can be added to the preparation of up-conversion nanoparticles. Because the  $\alpha$ -phase up-conversion nanomaterials have low luminous efficiency, they need to be calcined and annealed to become  $\beta$ -phase up-conversion nanomaterials which is a way to obtain high-quality nanocrystals [108]. At the same time, sealing ligands such as PVPk30 need to be added to improve their stability [109,110]. The equipment required by this method is easy to obtain and easy to operate, but it has the disadvantage of low crystallinity synthetic and agglomerate [111,112].

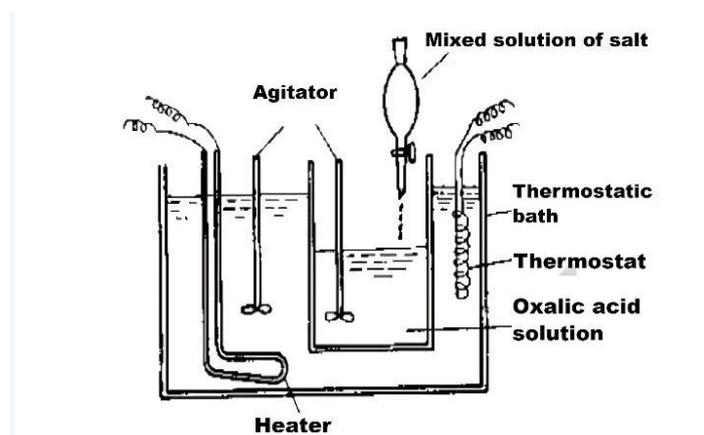


Figure 10. Diagram of compound precipitation synthesis device.

### (4) Sol-gel method

The sol-gel method involves the use of chemicals to prepare the precursors of metal water-soluble inorganic salts. It will produce a uniformly distributed liquid when the precursors are placed in a solvent, and the dispersion liquid can be reasonably added to it. An appropriate amount of coagulant can be added to make the salt hydrolyze produce a coagulation process and generate a uniform organic sol. Finally, the uniform organic sol is dried to obtain the application of UCNPs, as can be seen in Figure 11. However, it is difficult to control the morphology and particle size of the final product when applying this method. The method has the advantages of simple operation and low cost, but it has the disadvantages of poor dispersion, long preparation cycles, and the need for specific aging. Even so, the method has the possibility of agglomeration [113].

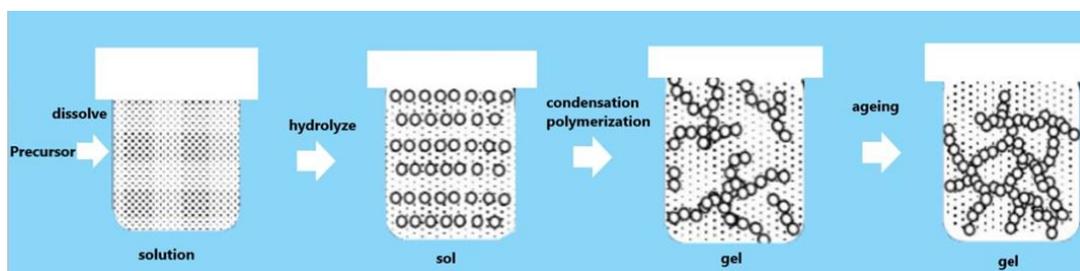


Figure 11. Flow chart of sol-gel method.

### (5) Microemulsion method

The microemulsion method can obtain a new reaction substance by adding certain emulsifiers to two immiscible liquids, which makes the aqueous phase effectively dispersed in the oil phase through the reaction. UCNPs prepared by this method have controllable particle size and a good appearance. For example, Zhou's group [114] used this method

to prepare the nano-SiO<sub>2</sub> with a narrow particle size distribution and good dispersion. However, the operation is complex and costly, and the required reaction conditions are harsh. The flow chart is shown in Figure 12.

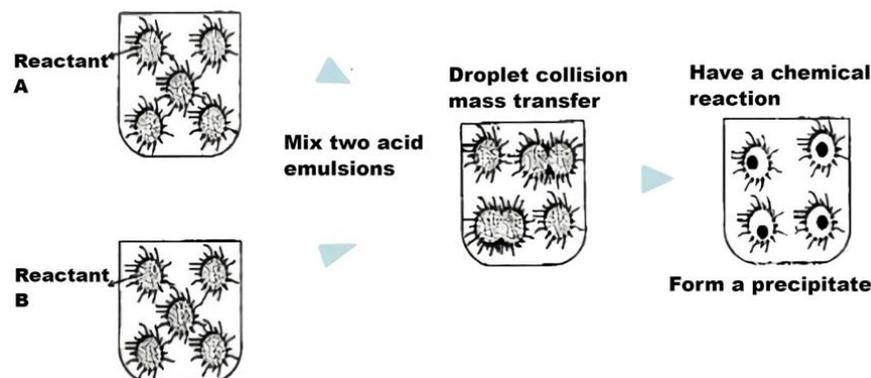


Figure 12. Flow chart of microemulsion method.

Among the five methods, the hydrothermal method and solvothermal method are simple and fast, which will be used in the practical application of the preparation of UCNPs in the future. Table 4 shows a comparison of the advantages of the common methods used to prepare UCNPs.

Table 4. Advantages and disadvantages of UCNPs preparation methods. Adapted with permission from Qin YK et al. [115].

Method	Advantages	Disadvantages
Thermal decomposition method	The product has high quality, is monodisperse, and has excellent morphology	Reaction requires high temperature, no oxygen, and no water, easy to produce toxic gases
Coprecipitation method	Simple process, low cost, relative environmental protection, high yield	The reaction requires high-temperature calcination, and the particle size of the product is not uniform
Hydrothermal method	The synthesis method is efficient, simple, and morphologically controllable	The size of the product is commonly large
Sol-gel method	The product has good dispersion and uniform morphology	The product agglomerates easily
Microemulsion method	Particle size controllable, good morphology	The operation is complicated, the cost is high and the reaction condition is harsh

#### 4.2. Optimization of Luminescence Efficiency of Up-Conversion Nanomaterials

There are three ways to optimize the up-conversion luminescence efficiency. The first way is to modify the surface of up-conversion nanoparticles. Using the atom resonance effect enhances the probability of radiative transition between the energy levels of rare earth atoms.

The second way is to dope some other atoms in the substrate [116,117], such as Gd<sup>3+</sup> and Mn<sup>3+</sup> [118]. The third way is to improve the luminous efficiency of materials by using the core-shell cladding and this way is more commonly used. The up-conversion luminescence nanocrystalline is coated with a similar lattice structure of passivated layer to form a large nanocrystalline. In this way, the activator can be effectively confined to the center of the nanocrystalline. This limits its aimless migration to cause quenching. The core-shell structure also greatly modifies the surface defects of nanocrystals, improving the luminescence optimization of up-conversion [119,120].

#### 4.3. Surface Functionalization of Up-Conversion Nanomaterials

##### (1) Hydrophilic modification

Up-conversion nanomaterials are oil-based, and their surfaces are often coated with hydrophobic ligands (OA or OM), making them difficult to be directly used for the detection of pollutants in water treatment. Combined with the water-insoluble property of PFOA, the detection of PFOA is more difficult. The hydrophilic modification of UCNPs can greatly improve their practical application performance. The hydrophilic modification methods include ligand exchange, surface silanization, ligand oxidation, and layer by layer assembly.

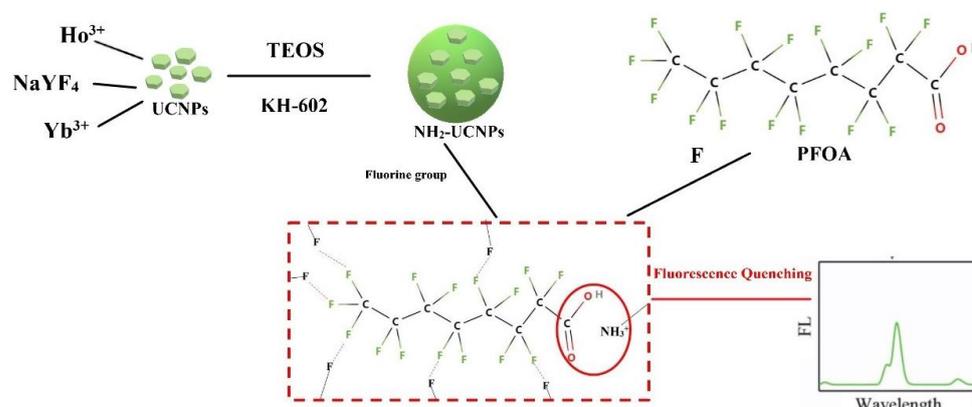
Surface silanization can prevent quenching by blocking the contact between nanoparticles and peripheral water, and can make the active groups carry out the next biomolecular coupling. For example, SiO<sub>2</sub> modification can make the material multi-pore size [121,122]. Ligand oxidation can couple UCNPs to biomolecules without interfering with the morphology and luminescence properties of UCNPs. However, this method is only applicable to the surface of nanoparticles without oxidation, and the modification time is long. In addition, it is easy to aggregate after modification [123]. The ligand removal method is simple and portable, and has little effect on the particle size of UCNPs. UCNPs are easy to aggregate after OA removal [124]. The ligand exchange method can greatly improve hydrophilicity and biocompatibility, but UCNPs will lose part of the luminous intensity after the exchange [125]. The surface-coated amphiphilic polymer method makes UCNPs have a good stability in an aqueous solution, but it will increase the diameter of UCNPs. The method is also difficult and costly [126].

##### (2) Modification of functionalization

UCNPs should be further functionalized to enhance practical performance when after hydrophilic modification. For example, it can be modified by amino group [76] or carboxyl group modification [127]. These ways can enhance the ability of detection.

#### 4.4. Principle of Up-Conversion Fluorescent Probe Detecting of PFOA

PFOA has rich F atoms, and F atoms have a huge electronegativity which makes the carbon-fluorine bond highly polar. Therefore, we can use this characteristic to detect PFOA. If the substrate of the UCNPs is fluoride, the up-conversion fluorescent probe prepared by it has fluorine groups on the surface, as shown in Figure 13. In addition, the amino modification of UCNPs can enhance luminescence. PFOA can bind to up-conversion fluorescent probes through F-F and electrostatic interactions, resulting in fluorescence quenching. Then, the content of PFOA in the aqueous solution can be known by comparing the fluorescence intensity of this process.



**Figure 13.** Schematic diagram of detecting PFOA.

Li's group [96] prepared a mesoporous structure-based up-conversion molecularly imprinted fluorescent probe ( $\text{NH}_2$ -UCNPs @MIPs) to detect PFOS. The chemical structure of PFOS is similar to PFOA. They both have 15 F atoms. The difference is that PFOS contains  $\text{SO}_3^-$  and PFOA contains  $\text{OH}^-$ .

PFOS can combine with  $\text{NH}_2$ -UCNPs @MIPs using the interaction of F-F and electricity, resulting in its fluorescence quenching. Based on this, they establish an efficient identification and highly sensitive detection technology to detect PFOS which a concentration range of PFOS is 0.01~15 nmol/L. This principle also applies to PFOA detection, because they have similar chemical structures and properties.

## 5. Application Progress and Recovery

### 5.1. Applications of Up-Conversion Fluorescent Probes in Detection

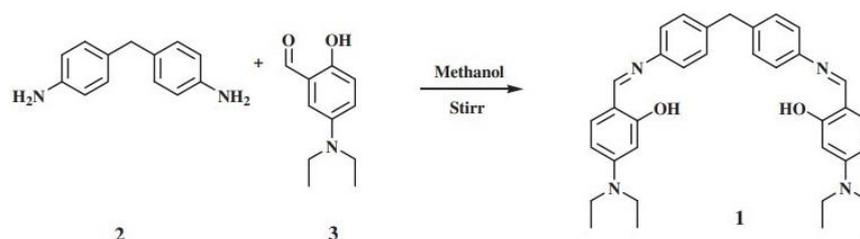
In the aspects of sewage treatment, food safety, and blood detection, detection technology is constantly being improved and innovated, such as through colorimetric analysis [128], enzyme inhibition [129], biochip technology [130], and UCL technology. Up-conversion fluorescent probes have excellent applications in detection.

In sewage treatment, it is necessary to face the detection of multiple sources of pollution, such as pesticides and veterinary drugs which easily flow into the water during use. It will cause clean water to be polluted into dirty water. Wang's group [131] found that  $\text{Cu}^{3+}$  can generate a fluorescence quenching reaction with UCNPs, so as to develop a highly sensitive fluorescence probe. Liu's group [132] found that UCNPs and gold nanoparticles can generate resonance energy transfer to detect cyanogen in sewage. Au nanoparticles will be adsorbed on the surface of UCNPs through electrostatic reaction, which reduces the fluorescence intensity of UCNPs. When up-conversion fluorescent probe detects an aqueous solution containing cyanide, the fluorescence intensity of UCNPs is strengthened. The content of cyanogen is displayed according to the comparison of fluorescence intensity. Up-conversion fluorescent probes have a great role in promoting the field of sewage treatment, which is suitable for its commercial generation and promotion [133–136].

### 5.2. Recovery and Challenges of Up-Conversion Fluorescent Probes

Up-conversion fluorescent probes have been fully used in many fields, but there are still some key problems to be solved when applying UCNPs. The sewage treatment detection needs to face a complex and variable detection environment. Therefore, the challenges of up-conversion fluorescent probes include selectively detecting target substances, improving the luminescence rate, and recycling. In addition, there is a lack of functional modification methods for UCNPs, and methods should be enriched which can make detection more convenient.

The recycling of fluorescent probes is the regeneration process of it. After the probe has selectively detected the target substance, using the principle of ionic competition between the intermediate product that was generated in situ and the specific ions to make the probe de-complexed to recover, it finally realizes the recycling of the probe [137]. Singh's group [138] prepared a multiple response fluorescent probe, using  $\text{Zn}^{2+}$  to form a complex to detect metal atoms. Finally,  $\text{PO}_4^{3-}$  was added to realize the regeneration of the fluorescent probe based on the principle of ionic competition, as shown in Figure 14. Tang's group [139] also used a similar principle to add  $\text{S}^{2-}$  realizing the regeneration of fluorescent probes.



**Figure 14.** Structure diagram of multiple responsive fluorescent probes. Adapted with permission from K Kaur et al. [138]. 1:  $\text{Zn}^{2+}$  complex 2:  $\text{NH}_2$  ions 3: N ion and OH ion.

PFOA is an organic pollutant. In the existing research, HPLC–MS/MS is used to detect it. However, it has high cost and great damage to the environment. This work focuses on the application of UCNPs. On the one hand, UCNPs have the unique advantages of good photostability, adjustable emission wavelength, low toxicity, and good biocompatibility. The fluorescent probes prepared by UCNPs can effectively detect PFOA. On the other hand, UCNPs not only have low cost but also cause little secondary pollution to the environment. Therefore, this work has contributed to the detection of PFOA to a certain extent.

## 6. Conclusions and Outlook

The effective and simple detection of PFOA has importance to people's lives. UCNPs are a good fluorescent material to detect PFOA, and  $\text{NaYF}_4: \text{Ho}^{3+}, \text{Yb}^{3+}$  is one of the better ones. Because  $\text{NaYF}_4$ , as fluoride, is more suitable as the substrate of UCNPs to detect PFOA, and  $\text{Ho}^{3+}$  as the activator can better fit the characteristics of  $\text{Yb}^{3+}$ . The up-conversion fluorescent probe prepared with UCNPs can effectively detect PFOA. The  $\text{SiO}_2$  modification and amino modification of the probe can make the probe dissolve in water faster and enhance luminous efficiency. Finally, the up-conversion fluorescent probe uses F-F interaction and electrostatic interaction to occur fluorescence quenching to detect PFOA.

The detecting method can be established in the laboratory, however, the preparation on a large scale is still a problem. In addition, it still has big obstacles to complete the experiments of recycling up-conversion fluorescent probes and selectively detecting PFOA. This method will have greatly positive influences on economics and the environment, because of the simplicity, ease, and efficiency of detecting PFOA in sewage and drinking water.

**Author Contributions:** Writing—original draft preparation, T.M. and X.S.; investigation, L.L.; writing—review and editing, Y.C. and X.L.; funding acquisition, C.F. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was funded by the National Key Research and Development Program of China (No.2021YFD1600402), the Key Research and Development Plan of Shaanxi Province (Grant No. 2020GXLH-Z-031), the Key Scientific Research Project of Weinan City (Grant No. 2020ZDYF-GYCX-107), the Key Scientific Research Project of Education Department of Shaanxi Province (Grant No. 20JS106), Central Guidance on Local Science and Technology Development Fund of Shaanxi Province (NO.2020-ZYYD-NCC-9), the Shaanxi Provincial Department of Education Collaborative Innovation Center Project (20JY052), the National Natural Science Foundation of China (NO.52072075), the Science and Technology planning project of Qindu District, Xianyang City (NO.2021QKJ-001), and the Outstanding Youth Science Fund of Shaanxi Province (No.2018JC-028).

**Institutional Review Board Statement:** The study did not require ethical approval.

**Data Availability Statement:** No data were created or analyzed in this study. Data sharing is not applicable to this article.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. Valsecchi, S.; Rusconi, M.; Mazzoni, M.; Viviano, G.; Pagnotta, R.; Zaghi, C.; Serrini, G.; Polesello, S. Occurrence and Sources of Perfluoroalkyl Acids in Italian River Basins. *Chemosphere* **2015**, *129*, 126–134. [[CrossRef](#)] [[PubMed](#)]
2. Li, M.; Cai, F.S.; Qin, R.X.; Zhuo, L.; Hu, F.Q.; Shi, Y.G.; Zheng, J. Pollution Characteristics of 16 Kinds of Perfluorinated Compounds in Wastewater from Typical Industries in Chongqing. *Acta Ecotoxicol.* **2021**, *16*, 44–58.
3. Wang, K.M.; Wu, X.Y.; Zhang, R.Z.; Hu, P. Determination of Perfluorooctanoate and Perfluorooctanesulfonate in Drinking Water by Solid Phase Extraction and High-Performance Liquid Chromatography Tandem Mass Spectrometry. *Guangdong Chem. Ind.* **2022**, *49*, 102–103.
4. Rushing, B.R.; Hu, Q.; Franklin, J.N.; McMahan, R.; Dagnino, S.; Higgins, C.P.; Strynar, M.J.; DeWitt, J.C. Evaluation of the Immunomodulatory Effects of 2,3,3,3-Tetrafluoro-2-(heptafluoropropoxy)-Propanoate in C57BL/6 mice. *Toxicol. Sci.* **2017**, *156*, 179–189. [[CrossRef](#)] [[PubMed](#)]
5. Guo, H.; Wang, J.H.; Yao, J.Z.; Sun, S.J.; Sheng, N.; Zhang, X.W.; Guo, X.J.; Guo, Y.; Sun, Y.; Dai, J.Y. Comparative Hepatotoxicity of Novel PFOA Alternatives(perfluoropolyether carboxylic acids)on Male Mice. *Environ. Sci. Technol.* **2019**, *53*, 3929–3937. [[CrossRef](#)]
6. Sheng, N.; Pan, Y.T.; Guo, Y.; Sun, Y.; Dai, J.Y. Hepatotoxic Effects of Hexafluoropropylene Oxide Trimer Acid(HFPO-TA),a Novel Perfluorooctanoic Acid (PFOA) Alternative on Mice. *Environ. Sci. Technol.* **2018**, *52*, 8005–8015. [[CrossRef](#)]

7. Shi, Y.L.; Vestergren, R.; Xu, L.; Zhou, Z.; Li, C.X.; Liang, Y.; Cai, Y.Q. Human Exposure and Elimination Kinetics of Chlorinated Polyfluoroalkyl Ether Sulfonic Acids(Cl-PFESAs). *Environ. Sci. Technol.* **2016**, *50*, 2396–2404. [[CrossRef](#)]
8. Sun, W.Q.; Ma, X.Z.; Zhou, Y.B.; Wang, L.; Liu, H. Effect of Perfluorooctanoic Acid on Lipid Accumulation of Rat Hepatocytes BRL-3A. *Med. Health Sci. Technol.* **2022**, *51*, 107–112.
9. Dong, G.H.; Zhang, Y.H.; Zhen, L.; Liang, Z.F.; He, Q.C. Effects of Perfluorooctane Sulfonic Acid on Proliferation and Cytokine Secretion of Mouse Lymphocytes in Vitro. *J. Toxicol.* **2011**, *25*, 125–128.
10. Zhang, L.M.; Rimal, B.P.; Nichols, R.G.; Tian, Y.; Smith, P.B.; Hatzakis, E.; Chang, S.C.; Butenhoff, J.L.; Peters, J.M.; Patterson, A.D. Perfluorooctane Sulfonate Alters Gut Microbiota-host Metabolic Homeostasis in Mice. *Toxicology* **2020**, *413*, 152365. [[CrossRef](#)]
11. Wei, J.P.; Xia, J.; Wang, X.J. Analysis of Detection Standards for PFOA and PFOS of Perfluorinated Compounds. *Org. Fluoride Ind.* **2014**, *162*, 38–42.
12. Cennamo, N.; Zeni, L.; Tortora, P.; Regonesi, M.E.; Giusti, A.; Staiano, M.; D’Auria, S.; Varriale, A. A High Sensitivity Biosensor to detect the presence of perfluorinated compounds in environment. *Talanta* **2018**, *178*, 955–961. [[CrossRef](#)]
13. Xu, X.L. Production, Processing and Use status of PFOA Substances in China. *Chem. Manag.* **2017**, *459*, 60–61.
14. Chen, H.; Zhang, Y.L. Current Situation and Safety Analysis of Food Packaging Materials in China. *Packag. Food Mach.* **2017**, *35*, 53–57.
15. Pontius, F. Regulation of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonic Acid (PFOS) in Drinking Water: A Comprehensive Review. *Water* **2019**, *11*, 2003. [[CrossRef](#)]
16. Baduel, C.; Paxman, C.J.; Mueller, J.F. Perfluoroalkyl Substances in a FireFigurehting Training Ground (FTG), Distribution and Potential Future Release. *J. Hazard. Mater.* **2015**, *296*, 46–53. [[CrossRef](#)]
17. Tsuda, S. Differential Toxicity between Perfluorooctane Sulfonate(PFOS)and Perfluorooctanoic Acid(PFOA). *J. Toxicol. Sci.* **2016**, *41*, 27–36. [[CrossRef](#)]
18. Lorenzo, M.; Campo, J.; Picó, Y. Analytical Challenges to Determine Emerging Persistent Organic Pollutants in Aquatic Ecosystems. *TrAC Trends Anal. Chem.* **2018**, *103*, 137–155. [[CrossRef](#)]
19. Sher, M.; Javed, M.; Shahid, S.; Hakami, O.; Qamar, M.A.; Iqbal, S.; AL-Anazy, M.M.; Baghdadi, H.B. Designing of highly active g-C<sub>3</sub>N<sub>4</sub>/Sn doped ZnO heterostructure as a photocatalyst for the disinfection and degradation of the organic pollutants under visible light irradiation. *J. Photochem. Photobiol. A Chem.* **2021**, *418*, 113393. [[CrossRef](#)]
20. Qamar, M.A.; Javed, M.; Shahid, S.; Iqbal, S.; Abubshait, S.A.; Abubshait, H.A.; Ramay, S.M.; Mahmood, A.; Ghaithan, H.M. Designing of highly active g-C<sub>3</sub>N<sub>4</sub>/Co@ZnO ternary nanocomposites for the disinfection of pathogens and degradation of the organic pollutants from wastewater under visible light. *J. Environ. Chem. Eng.* **2021**, *9*, 105534. [[CrossRef](#)]
21. Leng, T.H.; Wang, L.; Zheng, Y. Determination of 12 Perfluorinated Compounds in Rice Powder for Infants and Young Children by High Performance Liquid Chromatography-tandem Mass Spectrometry. *Food Saf. Qual.* **2019**, *10*, 8087–8092.
22. Liu, S.Y.; Jin, Q.; Ren, R. Determination of Perfluorocarboxylic Acids in Fish by Ultra Performance Liquid Chromatography Tandem Mass Spectrometry. *Prev. Med.* **2020**, *32*, 1204–1207.
23. Guo, J.; Guo, M.M.; Wu, H.Y.; Zhuo, Y.X.; Mou, H.J.; Lu, L.N.; Tan, Z.J. Simultaneous Determination of Perfluorinatedacids and Their Precursors in Bivalve Shellfish by Double SPE Columns Purification and Ultra Fast Liquid Chromatography-tandem Mass Spectrometry. *Food Sci.* **2017**, *38*, 248–255.
24. He, J.L.; Peng, T.; Xie, J.; Dai, H.H.; Chen, D.D.; Yue, Z.F.; Fan, C.L.; Li, C. Development of a Qu ECh ERs Method for Determination of 20 Perfluorinated Compounds in Animal Liver by HPLC–MS/MS. *Chin. J. Anal. Chem.* **2015**, *43*, 40–48. [[CrossRef](#)]
25. Feng, S.; Lan, F.; Wu, X.P.; Shen, J.C.; Yue, Z.F.; Xiong, B.B. Determination of 15 Perttuorinated Compounds in Chinese Spirit by Dispersive Solid Phase Extraction and HPLC-MS-MS. *Food Sci.* **2013**, *34*, 143–149.
26. Huang, B.; Liu, W.J.; Fu, J.W.; Su, D.S.; Wu, J.H.; Ai, Y.Z. Simultaneous Detection of 7 Neonicotinoid Insecticides in Camellia Oil by QuEChERS Method Coupled with Ultra High Performance Liquid Chromatography Tandem Mass Spectrometry. *Qual. Saf. Agric. Prod.* **2019**, *97*, 45–48.
27. Yang, W.L.; Guo, J.; Du, W. Determination of Perfluorooctanoic Acid and Perfluorooctanesulfonates in Environmental Water and Soil Matrixes Using ultra High Performance Liquid Chromatography Coupled with State-of-art Tandem Quadrupole Mass Spectrometry. *Environ. Chem.* **2018**, *37*, 2820–2823.
28. Yu, L.; Song, W.; Lv, Y.L.; Zhao, M.Y.; Zhao, M.Y.; Zhou, F.F.; Hu, Y.Y.; Zheng, P. Rapid Determination of 204 Pesticide Residues in Tea by Ultra Performance Liquid Chromatography Coupled with Quadrupole-time of Flight Mass Spectrometry. *Colour Spectr.* **2015**, *33*, 597–612. [[CrossRef](#)]
29. Zabaleta, I.; Negreira, N.; Bizkarguenaga, E. Screening and Identification of Per- and Polyfluoroalkyl Substances in Microwave Popcorn Bags. *Food Chem.* **2017**, *230*, 497. [[CrossRef](#)] [[PubMed](#)]
30. Huang, M.W.; Wu, H.; Yu, W.; Wang, Y.; Wang, F.C.; Zhang, C.C.; Zhou, L.S.; Li, Z.G. Rapid Identification of Chemical Constituents of Qiyu Sanlong Decoction by Ultra Performance Liquid Chromatography-Quadrupole-Time of Flight Mass Spectrometry. *Chromatogr. J.* **2021**, *39*, 730–743.
31. Buszewska-Forajta, M.; Bujak, R.; Yumba-Mpanga, A.; Siluk, D.; Kaliszan, R. GC/MS Technique and AMDIS Software Application in Identification of Hydrophobic Compounds of Grasshoppers’ Abdominal Secretion (*Chorthippus* spp.). *J. Pharm. Biomed. Anal.* **2015**, *102*, 331–339. [[CrossRef](#)] [[PubMed](#)]
32. Wei, L.L. Rapid Determination of Perfluorooctanoic Acid in Food Contact Materials by Pre-column Derivatization and Gas Chromatography. *Packag. Food Mach.* **2019**, *37*, 69–72.

33. Luo, S.P.; Shang, G.Q.; Chen, M. Research Progress of Detection Methods of Perfluorinated Compounds in Food Contact Materials. *J. Food Saf. Qual. Insp.* **2013**, *4*, 993–998.
34. Wang, X.Y.; Shen, W.J.; Wang, H.; Yu, K.Y.; Wu, B.; Hu, G.S.; Yang, G.J. Determination of 10 Perfluorocarboxylic Acid Compounds in Water by Gas Chromatography-negative Chemical Source-mass Spectrometry. *Chromatography* **2019**, *37*, 32–39.
35. Zheng, Z.; Yu, H.J.; Geng, W.C.; Hu, X.Y.; Wang, Y.Y.; Li, Z.H.; Wang, Y.F.; Guo, D.S. Guanidinocalix[5]arene for sensitive fluorescence detection and magnetic removal of perfluorinated pollutants. *Nat. Commun.* **2019**, *10*, 5762. [[CrossRef](#)] [[PubMed](#)]
36. Tian, F. Application of Rhodamine Fluorescent Probes in the Detection of Heavy Metals and Transition Metal atoms. *Petrochem. Technol.* **2019**, *26*, 2693–2699.
37. Chen, Y.Y.; Ding, M.M.; Li, J.Q.; Sheng, W.; Liu, B.; Zhang, Y.; Wang, S. Fluorescence quenching immunoaffinity test column with quantum dots as fluorescence donors for the quick detection of malachite green and crystal violet in aquatic products. *Food Anal. Methods* **2018**, *11*, 3362–3370. [[CrossRef](#)]
38. Hua, G.S.; Sheng, W.; Li, J.M.; Zhang, Y.; Wang, J.P.; Wang, S. Fluorescent quenching immune chromatographic strips with quantum dots and up-conversion nanoparticles as fluorescent donors for visual detection of sulfaquinolone in foods of animal origin. *Anal. Chim. Acta* **2017**, *982*, 185–192. [[CrossRef](#)] [[PubMed](#)]
39. Zhang, Z.H.; Zhou, H.L.; Wu, F.; Zhang, Y. Temperature Sensing Characteristics of Up-conversion Luminescence in  $Tm^{3+}/Yb^{3+}$ -Coped LuYO<sub>3</sub> Phospho. *J. Lumin.* **2021**, *42*, 1872–1881.
40. Chen, J.Y.; Su, W.; Wang, E.J. Highly selective ratiometric fluorescent probe for Hg<sup>2+</sup> based on fluorescence resonance energy transfer between 1,8-naphthalimide and rhodamine B. *Chem. J. Chin. Univ.* **2016**, *37*, 232–238.
41. Li, M.; Jiang, X.; Wu, H.; Jiang, X.J.; Wu, H.H.; Xu, H.; Zang, S.Q. A dual functional probe for “turn-on” fluorescence response of Pb<sup>2+</sup> and colorimetric detection of Cu<sup>2+</sup> based on a rhodamine derivative in aqueous media. *Dalton Trans.* **2015**, *44*, 17326–17334. [[CrossRef](#)] [[PubMed](#)]
42. Balamurugan, A.; Kumar, V.; Jayakannan, M. Triple action polymer probe: Carboxylic distilbene fluorescent polymer chemosensor for temperature, metal-ions and biomolecules. *Chem. Commun.* **2014**, *50*, 842. [[CrossRef](#)] [[PubMed](#)]
43. Duret, D.; Haftek-Terreau, Z.; Carretier, M.; Berki, Z.; Ladavière, C.; Monier, K.; Bouvet, P.; Marvel, J.; Leverrier, Y.; Charreyre, M.T.; et al. Labeling of native proteins with fluorescent RAFT polymer probes: Application to the detection of a cell surface protein using flow cytometry. *Polym. Chem.* **2020**, *9*, 1857. [[CrossRef](#)]
44. Zhang, Y.; Jiang, H.; Wang, X. Cytidine-stabilized gold nanocluster as a fluorescence turn-on and turn-off probe for dual functional detection of Ag<sup>+</sup> and Hg<sup>2+</sup>. *Anal. Chim. Acta* **2015**, *870*, 1–7. [[CrossRef](#)]
45. Peng, Y.; Wang, M.M.; Wu, X.X.; Wang, F.; Liu, L. Methionine-capped gold nanoclusters as a fluorescence-enhanced probe for cadmium(II) sensing. *Sensors* **2018**, *18*, 658–668. [[CrossRef](#)]
46. Kelsey, L.R.; Hwang, J.-H.; Esfahani, A.R.; Esfahani, A.R.; Anwar, S.A.H.M.; Lee, W.H. Recent Developments of PFAS-Detecting Sensors and Future Direction: A Review. *Micromachines* **2020**, *11*, 667.
47. Karimian, N.; Stortini, A.M.; Moretto, L.M.; Costantino, C.; Bogialli, S.; Ugo, P. Electrochemosensor for trace analysis of perfluorooctanesulfonate in water based on a molecularly imprinted poly(o-phenylenediamine) polymer. *ACS Sens.* **2018**, *3*, 1291–1298. [[CrossRef](#)]
48. Cheng, Y.H.; Barpaga, D.; Soltis, J.A.; Shutthanandan, V.; Kargupta, R.; Han, K.S.; McGrail, B.P.; Motkuri, R.K.; Basuray, S.; Chatterjee, S. Metal–Organic Framework-Based Microfluidic Impedance Sensor Platform for Ultrasensitive Detection of Perfluorooctanesulfonate. *ACS Appl. Mater. Interfaces* **2020**, *12*, 10503–10514. [[CrossRef](#)]
49. Feng, H.; Wang, N.Y.; Tran, T.T.T.; Yuan, L.J.; Li, J.Z.; Cai, Q.Y. Surface molecular imprinting on dye-(NH<sub>2</sub>)-SiO<sub>2</sub> NPs for specific recognition and direct fluorescent quantification of perfluorooctane sulfonate. *Sens. Actuators B Chem.* **2014**, *195*, 266–273. [[CrossRef](#)]
50. Cennamo, N.; D’Agostino, G.; Sequeira, F.; Mattiello, F.; Porto, G.; Biasiolo, A.; Nogueira, R.; Billo, L.; Zeni, L. A simple and low-cost optical fiber intensity-based configuration for perfluorinated compounds in water solution. *Sensors* **2018**, *18*, 3009. [[CrossRef](#)]
51. Takayose, M.; Akamatsu, K.; Nawafune, H.; Murashima, T.; Matsui, J. Colorimetric detection of perfluorooctanoic acid (PFOA) utilizing polystyrene-modified gold nanoparticles. *Anal. Lett.* **2012**, *45*, 2856–2864. [[CrossRef](#)]
52. Chen, S.H.; Li, A.M.; Zhang, L.Z.; Gong, J.M. Molecularly imprinted ultrathin graphitic carbon nitridenanosheets-Based electrochemiluminescence sensing probe for sensitive detection of perfluorooctanoic acid. *Anal. Chim. Acta* **2015**, *896*, 68–77. [[CrossRef](#)] [[PubMed](#)]
53. Xia, W.; Wan, Y.J.; Wang, X.L.; Li, Y.Y.; Wang, W.J.; Wang, C.X.; Xu, S.Q. Sensitive bioassay for detection of PPAR $\alpha$  potentially hazardous ligands with gold nanoparticle probe. *Hazard. Mater.* **2011**, *192*, 1148–1154. [[CrossRef](#)] [[PubMed](#)]
54. Yang, H. Monitoring of perfluorinated compounds from the imported fishery products in Korea by liquid chromatography-tandem mass spectrometry (LC-MS/MS). *Environ. Epidemiol.* **2019**, *3*, 452–453.
55. Lertassavakorn, T.; Pholphana, N.; Rangkadilok, N.; Suriyo, T.; Satayavivad, J. Determination of perfluorooctane sulfonate and perfluorooctanoic acid in seafood and water from Map Ta Phut Industrial Estate area, Thailand. *Food Addit. Contam. Part A* **2021**, *38*, 11–16. [[CrossRef](#)] [[PubMed](#)]
56. Gan, C.D.; Gan, Z.W.; Cui, S.F.; Fan, R.J.; Fu, Y.Z.; Peng, M.Y.; Yang, J.Y. Agricultural activities impact on soil and sediment fluorine and perfluorinated compounds in an endemic fluorosis area. *Sci. Total Environ.* **2021**, *771*, 144809. [[CrossRef](#)] [[PubMed](#)]

57. Wang, Z.; Qiao, H.Q.; Yang, J.; Fu, Y.Q.; Wang, H.; Yao, J.H.; Gao, T.Q.; Deng, J.G. Simultaneous determination of fluoroalkanes in aquatic products by ultra tandem high performance liquid chromatography mass spectrometry. *J. Food Saf. Qual.* **2022**, *13*, 1132–1140.
58. Wang, Y.K.; Chen, W.Y.; Wen, S.G.; Meng, K.Q.; Xie, S.Z.; Zhen, J.X.; Qiu, C.S. Headspace gas chromatography detection method for perfluorohydrocarbon tracer. *Fine Spec. Chem.* **2020**, *28*, 10–13.
59. Gebbink, W.A.; Letcher, R.I. Linear and branched perfluorooctane sulfonate isomer patterns in herring gull eggs from colonial sites across the Laurentian Great Lakes. *Environ. Sci. Technol.* **2010**, *44*, 3739–3745. [[CrossRef](#)]
60. Su, C.Y.; Zheng, N.; Li, S.L.; Qu, X.Y.; Zhou, X.W.; Ma, T.; Yang, H.J.; Wang, J.Q. Research progress on detection methods of perfluorinated compounds in milk. *China Dairy Ind.* **2018**, *46*, 29–33.
61. Wang, J.; Chen, H.Y.; Ru, F.; Zhang, Z.; Mao, X.; Shan, D.L.; Chen, J.; Lu, X.Q. Encapsulation of dual-emitting fluorescent magnetic nanoprobe in metal-organic frameworks for ultrasensitive ratiometric detection of Cu<sup>2+</sup>. *Chem. A Eur. J.* **2018**, *24*, 3499–3505. [[CrossRef](#)]
62. Chen, X.Y.; Sameer Hussain Tang, Y.H.; Chen, X.; Zhang, S.J.; Wang, Y.; Zhang, P.; Gao, R.X.; Wang, S.; Hao, Y. Two-in-one platform based on conjugated polymer for ultrasensitive ratiometric detection and efficient removal of perfluoroalkyl substances from environmental water. *Sci. Total Environ.* **2022**, *860*, 160467. [[CrossRef](#)] [[PubMed](#)]
63. Van Tran, V. Conjugated Polymers-Based Biosensors for Virus Detection: Lessons from COVID-19. *Biosensors* **2022**, *12*, 748. [[CrossRef](#)] [[PubMed](#)]
64. Malik, A.H.; Hussain, S.; Kalita, A.; Iyer, P.K. Conjugated Polymer Nanoparticles for the Amplified Detection of Nitro-explosive Picric Acid on Multiple Platforms. *ACS Appl. Mater. Interfaces* **2015**, *7*, 26968–26976. [[CrossRef](#)]
65. Chen, X.; Hussain, S.; Hao, Y.; Tian, X.M.; Gao, R.X. Review—Recent Advances of Signal Amplified Smart Conjugated Polymers for Optical Detection on Solid Support. *ECS J. Solid State Sci. Technol.* **2021**, *10*, 037006. [[CrossRef](#)]
66. Feng, F.D.; Liu, L.B.; Yang, Q.; Wang, S. Water-soluble conjugated polymers for fluorescent-enzyme assays. *Macromol. Rapid Commun.* **2010**, *31*, 1405–1421. [[CrossRef](#)] [[PubMed](#)]
67. Sun, X.; Wang, Y.W.; Peng, Y. A selective and ratiometric bifunctional fluorescent probe for Al<sup>3+</sup> ion and proton. *Org. Lett.* **2012**, *14*, 3420–3423. [[CrossRef](#)]
68. Luo, P.G.; Yang, F.; Yang, S.T.; Sonkar, S.K.; Yang, L.J.; Broglie, J.J.; Liu, Y.; Sun, Y.P. Carbon-based quantum dots for fluorescence imaging of cells and tissues. *RSC Adv.* **2014**, *4*, 10791–10797. [[CrossRef](#)]
69. Yao, J.; Yang, M.; Duan, Y. Chemistry, biology, and medicine of fluorescent nanomaterials and related systems: New insights into biosensing, bioimaging, genomics, diagnostics, and therapy. *Chem. Rev.* **2014**, *114*, 6130–6148. [[CrossRef](#)] [[PubMed](#)]
70. Idris, N.M.; Jayakumar, M.K.G.; Bansal, A.; Zhang, Y. Up-conversion nanoparticles as versatile light nanotransducers for photoactivation applicatatoms. *Chem. Soc. Rev.* **2015**, *44*, 1449. [[CrossRef](#)]
71. Zhou, B.; Shi, B.Y.; Jin, D.Y.; Liu, X.G. Controlling up-conversion nanocrystals for emerging applicatatoms. *Nat. Nanotechnol.* **2015**, *10*, 924. [[CrossRef](#)] [[PubMed](#)]
72. Zhu, X.H.; Zhang, J.; Liu, J.L.; Zhang, Y. Recent progress of rare-earth doped up-conversion nanoparticles: Synthesis, optimization, and applicatatoms. *Adv. Sci.* **2019**, *6*, 30. [[CrossRef](#)] [[PubMed](#)]
73. Auzel, F. Up-conversion and anti-stokes processes with f and d atoms in solids. *Chem. Rev.* **2004**, *104*, 139–174. [[CrossRef](#)] [[PubMed](#)]
74. Shi, Y.L.; Song, X.W.; Jin, Q.; Li, W.H. Tissue distribution and bio-accumulation of a novel polyfluoroalkyl benzenesulfonate in crucian carp. *Environ. Int.* **2020**, *135*, 105418. [[CrossRef](#)] [[PubMed](#)]
75. Ren, Y.; He, S.; Huttad, L. An NIR-II/MR dual modal nanoprobe for liver cancer imaging. *Nanoscale* **2021**, *12*, 11510–11517. [[CrossRef](#)]
76. Chen, H.Q.; Tang, W.; Liu, Y.C.; Wang, L. Quantitative image analysis method for detection of nitrite with cyanine dye-NaYF<sub>4</sub>:Yb,Tm@NaYF<sub>4</sub> up-conversion nanoparticles composite luminescent probe. *Food Chem.* **2021**, *367*, 130660. [[CrossRef](#)]
77. Huang, D. Theoretical research and application of rare earth up-conversion luminescent materials. *J. Chifeng Univ. (Nat. Sci. Ed.)* **2017**, *33*, 7–8.
78. Dan, S.; Wu, H.; Ma, X.J.; Tan, M.Q. Preparation and biological application of rare earth up-conversion fluorescent nanomaterials. *Prog. Biochem. Biophys.* **2013**, *40*, 925–934.
79. Zhao, X.; Zhang, L.; Yan, X.; Zhang, L.; Yang, L.; Pan, J.L.; Zhang, M.L.; Wang, C.G.; Suo, H.; Jia, X.T.; et al. A near-infrared light triggered fluorometric biosensor for sensitive detection of acetylcholinesterase activity based on NaErF<sub>4</sub>: 0.5% Ho<sup>3+</sup>@NaYF<sub>4</sub> up-conversion nano-probe. *Talanta* **2021**, *235*, 122784. [[CrossRef](#)]
80. Tian, J.; Luo, H.F. Research Development of Up-Converting Rare-earth Nanophosphors. *Front. Chem.* **2014**, *35*, 1–4.
81. Xu, J.T.; Gulzar, A.; Yang, P.P.; Bi, H.T.; Yang, D.; Gai, S.L.; He, F.; Lin, J.; Xing, B.G.; Jin, D.Y. Recent advances in near-infrared emitting lanthanide-doped nanoconstructs: Mechanism, design and application for bioimaging. *Coord. Chem. Rev.* **2019**, *381*, 104. [[CrossRef](#)]
82. Yamini, S.; Gunaseelan, M.; Kumar, G.A.; Singh, S.; Dannangoda, G.C.; Martirosyan, K.S.; Sardar, D.K.; Sivakumar, S.; Girigoswami, A.; Senthilselvan, J. NaGdF<sub>4</sub>:Yb,Er-Ag nanowire hybrid nanocomposite for multifunctional up-conversion emission, optical imaging, MRI and CT imaging applicatatoms. *Microchim. Acta* **2020**, *187*, 10. [[CrossRef](#)] [[PubMed](#)]
83. Lin, S.L.; Chang, C.A. Optimising FRET-efficiency of Nd<sup>3+</sup> sensitised up-conversion nanocomposites by shortening the emitter-photosensitizer distance. *Nanoscale* **2020**, *12*, 8742. [[CrossRef](#)]

84. Gu, B.; Zhang, Q.C. Recent advances on functionalized up-conversion nanoparticles for detection of small molecules and atoms in biosystems. *Adv. Sci.* **2018**, *5*, 16. [[CrossRef](#)] [[PubMed](#)]
85. Zheng, K.Z.; Loh, K.Y.; Wang, Y.; Chen, Q.S.; Fan, J.Y.; Jung, T.; Nam, S.H.; Suh, Y.D.; Liu, X.G. Recent advances in up-conversion nanocrystals: Expanding the kaleidoscopic toolbox for emerging applications. *Nano Today* **2019**, *29*, 100797. [[CrossRef](#)]
86. Nie, J.H.; Gao, H.; Li, X.; Liu, S.M. Up-conversion luminescence properties of different fluoride substrate materials NaREF<sub>4</sub>(RE: Gd, Lu, Y) doped with Er<sup>3+</sup>/Yb<sup>3+</sup>. *J. Lumin.* **2018**, *204*, 333–340. [[CrossRef](#)]
87. Fan, X.; Huang, W.; Ying, W.T.; Xu, S.Q. High efficient upconversion luminescence of NaGdF<sub>4</sub>: Yb<sup>3+</sup>/Er<sup>3+</sup> nanoparticle: First-principles calculation, dual-wavelength stimuli and logic gate application. *Mater. Technol.* **2021**, *37*, 1–10. [[CrossRef](#)]
88. Wu, X.F.; Chen, Z.H. Preparation of up-conversion fluorescent probe and its plant cell imaging. *Electron. Test.* **2015**, *7*, 43–45.
89. Li, X.F.; Wu, X.M.; Tao, J. Hydrothermal Synthesis of ZnO:Ho, Yb Luminescent Powder and Its Up-conversion Luminescence Characteristics. *J. Synth. Cryst.* **2016**, *45*, 2567–2571.
90. Mettra, B.; Appaix, F.; Olesiak-Banska, J.; Le Bahers, T.; Leung, A.; Matczyszyn, K.; Samoc, M.; van der Sanden, B.; Monnereau, C.; Andraud, C. A fluorescent polymer probe with high selectivity toward vascular endothelial cells for and beyond noninvasive two-photon intravital imaging of brain vasculature. *ACS Appl. Mater. Interfaces* **2016**, *8*, 17047–17059. [[CrossRef](#)]
91. Li, Z.Y.; Liu, K.X.; Yang, Q.Z.; Yang, Y.X.; Yuan, L. Synthesis and properties of a highly selective fluorescent probe for Al<sup>3+</sup>. *Mod. Chem. Ind.* **2022**, *43*, 254–258.
92. Yu, Y.C.; Feng, J.K.; Liu, B.; Feng, J.K.; Liu, B.; You, J.; Wu, W.J. A turn-on type Al<sup>3+</sup> photoprobe and its in vivo bioassay. *China Environ. Sci.* **2020**, *40*, 5422–5427.
93. Zhang, Y.M.; Chen, Y.C.; Bai, Y.; Bai, Y.; Xue, X.L.; He, W.J.; Guo, Z.J. FRET-based fluorescent ratiometric probes for the rapid detection of endogenous hydrogen sulphide in living cells. *Analyst* **2020**, *145*, 4233–4238. [[CrossRef](#)]
94. Hu, W.R.; Song, Q.J.; Dong, M.M. Enhanced fluorescent probe for detection of BSA based on aggregation-induced emission. *Chin. J. Anal. Lab.* **2022**. [[CrossRef](#)]
95. Li, C.J.; Xing, K.Q.; Liu, Y.C.; Xiang, K.Q.; Liu, Y.C.; Zheng, Y.C.; Tian, B.Z.; Zhang, J.L. A novel colorimetric chemosensor for Cu<sup>2+</sup> with high selectivity and sensitivity based on Rhodamine B. *Res. Chem. Intermed.* **2015**, *41*, 10169–10180. [[CrossRef](#)]
96. Li, J.; Guo, H.Q.; Yu, H.; Tian, L.X.; Yan, L.S.; Liu, X.M.; Lin, L.K.; Wang, J.L. Mesoporous Structure-based Up-conversion Molecularly Imprinted Fluorescent Probes for the Detection of Perfluorooctanoic Sulfonic Acid. *Chin. J. Anal. Chem.* **2020**, *48*, 1493–1501.
97. Zhang, T.; Zhao, H.; Lei, A.; Quan, X. Electrochemical Biosensor for Detection of Perfluorooctane Sulfonate Based on Inhibition Biocatalysis of Enzymatic Fuel Cell. *Electrochemistry* **2014**, *82*, 94–99. [[CrossRef](#)]
98. Guo, H.; Li, Z.Q.; Qian, H.S.; Hu, Y.; Muhammad, I.N. Seed-mediated synthesis of NaYF<sub>4</sub>:Yb, Er/NaGdF<sub>4</sub> nanocrystals with improved up-conversion fluorescence and MR relaxivity. *Nanotechnology* **2010**, *21*, 125602. [[CrossRef](#)] [[PubMed](#)]
99. Wang, X.; Zhuang, J.; Peng, Q.; Li, Y.D. A general strategy for nanocrystal synthesis. *Nature* **2005**, *437*, 121–124. [[CrossRef](#)] [[PubMed](#)]
100. Zhang, F.; Li, J.; Shan, J.; Xu, L.; Zhao, D. Shape, Size, and Phase- Controlled Rare- Earth Fluoride Nanocrystals with Optical Up-Conversion Properties. *Chem.-Eur. J.* **2009**, *15*, 11010–11019. [[CrossRef](#)]
101. Wang, F.; Han, Y.; Lim, C.S.; Lu, Y.H.; Wang, J.; Xu, J.; Chen, H.Y.; Zhang, C.; Hong, M.H.; Liu, X.G. Simultaneous phase and size control of up-conversion nanocrystals through lanthanide doping. *Nature* **2010**, *463*, 1061–1065. [[CrossRef](#)]
102. Radunz, S.; Schavkan, A.; Wahl, S.; Wuerth, C.; Tschiche, H.; Krumrey, M.; Resch-Genger, U. Evolution of size and optical properties of upconverting nanoparticles during high-temperature synthesis. *Phys. Chem. C* **2018**, *122*, 28958–28967. [[CrossRef](#)]
103. Skripka, A.; Marin, R.; Benayas, A.; Canton, P.; Hemmer, E.; Vetrone, F. Double rare-earth nanothermometer in aqueous media: Opening the third optical transparency window to temperature sensing. *Phys. Chem. Chem. Phys.* **2017**, *7*, 44531–44536. [[CrossRef](#)]
104. Dong, J.; Zhang, J.; Han, Q.; Zhao, X.; Yan, X.; Liu, J.; Ge, H.; Gao, W. Tuning and enhancing the red upconversion emission of Er<sup>3+</sup> in LiYF<sub>4</sub> nanoparticles. *J. Lumin.* **2019**, *207*, 361–368. [[CrossRef](#)]
105. Wang, H.D.; Wang, L.; Ye, S.Y.; Song, X.L. Construction of Bi<sub>2</sub>WO<sub>6</sub>-TiO<sub>2</sub>/starch nanocomposite films for visible-light catalytic degradation of ethylene. *Food Hydrocoll.* **2019**, *88*, 92–100. [[CrossRef](#)]
106. Yuan, X.Y.; Yu, L.X.; Jia, W. One-step solvothermal preparation of Bi-Bi<sub>2</sub>O<sub>3</sub>-BiOBr ternary complex and its visible light-driven catalytic degradation of methylene blue. *Acta Mater. Compos. Sin.* **2022**, 1000–3851.
107. Hu, X.K.; Shang, X.Y.; Huang, P.; Zheng, W.; Chen, X.Y. Polarization up-conversion luminescence and orientation tracing of NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> single particle nanorods. *J. Chem.* **2022**, *80*, 244–248.
108. Li, Y.T.; Guo, W.; Wu, Z.W.; Jin, L.; Ke, Y.; Guo, Q.; Hu, S. Determination of ultra-trace rare earth elements in high-salt groundwater using aerosol dilution inductively coupled plasma-mass spectrometry (ICP-MS) after iron hydroxide co-precipitation. *Microchem. J.* **2016**, *126*, 194–199. [[CrossRef](#)]
109. Wang, F.; Liu, X. Recent advances in the chemistry of lanthanide doped up-conversion nanocrystals. *Chem. Soc. Rev.* **2009**, *38*, 976–989. [[CrossRef](#)]
110. Du, H.; Zhang, W.; Sun, J. Structure and up-conversion luminescence properties of BaYF<sub>5</sub>:Yb<sup>3+</sup>, Er<sup>3+</sup> nanoparticles prepared by different methods. *J. Alloys Compd.* **2011**, *509*, 3413–3418. [[CrossRef](#)]
111. Zhang, T.; Yue, H.J.; Qiu, H.L.; Wei, Y.J.; Wang, C.Z.; Chen, G.; Zhang, D. Nano-particle assembled porous core-shell ZnMn<sub>2</sub>O<sub>4</sub> microspheres with superb performance for lithium batteries. *Nanotechnology* **2017**, *28*, 105403. [[CrossRef](#)]

112. Wang, J.; Zhao, D.S.; Shen, Y.X.; Li, F.F.; Zhang, M.X. Preparation of silver vanadate photocatalyst by coprecipitation method and the effect of pH on its photocatalytic activity. *Rare Met.* **2022**, *46*, 906–912.
113. Yi, G.S.; Lu, H.C.; Zhao, S.Y.; Yang, W.J.; Chen, D.P.; Guo, L.H. Synthesis, Characterization, and Biological Application of Size-Controlled Nanocrystalline NaYF<sub>4</sub>:Yb,Er Infrared-to-Visible Up-Conversion Phosphors. *Nano Lett.* **2004**, *4*, 2191–2196. [[CrossRef](#)]
114. Zhou, F.R.; Zhang, M.X. Preparation of Nano-SiO<sub>2</sub> by Microemulsion Method and Performance of Its Modified PVA-CS Films. *Anhui Chem. Ind.* **2022**, *48*, 52–54.
115. Qin, Y.K.; Li, S.; Hong, Y.; Wang, Z.G.; Han, D.P.; Peng, Y.; Zhou, H.Y.; Gao, Z.X.; Han, T.; Wang, Z.G.; et al. Research Process of Synthesis Functionalization and Application of Up-conversion Nanoparticles in Food Safety Detection. *Anal. Chem.* **2021**, *49*, 1955–1969.
116. Shao, Q.Q.; Zhang, H.; Dai, J.Y.; Yang, C.; Chen, X.X.; Feng, G.Y.; Zhou, S.H. Preparation, growth mechanism, size manipulation and near-infrared luminescence enhancement of beta-NaYF<sub>4</sub>:Nd<sup>3+</sup> microcrystals via Ca<sup>2+</sup> doping. *Crystemcomm* **2019**, *21*, 741. [[CrossRef](#)]
117. Dibaba, S.T.; Xiao, Q.G.; Ren, W.; Sun, L. Recent progress of energy transfer and luminescence intensity boosting mechanism in Nd<sup>3+</sup> sensitized up-conversion nanoparticles. *J. Rare Earths* **2019**, *37*, 791. [[CrossRef](#)]
118. Tian, G.; Gu, Z.J.; Zhou, L.J.; Yin, W.Y.; Liu, X.X.; Yan, L.; Jin, S.; Ren, W.L.; Xing, G.M.; Li, S.J.; et al. Mn<sup>2+</sup> dopeant-controlled Synthesis of NaYF<sub>4</sub>:Yb/Er Up-conversion Nanoparticles for in Vivo Imaging and Drug Delivery. *Adv. Mater.* **2012**, *24*, 1226. [[CrossRef](#)]
119. Kang, N.; Zhao, J.; Zhou, Y.; Ai, C.C.; Wang, X.M.; Ren, L. Enhanced Up-conversion Luminescence Intensity of Core-shell NaYF<sub>4</sub> Nanocrystals Guided by Morphological Control. *Nanotechnology* **2019**, *30*, 105701. [[CrossRef](#)]
120. Würth, C.; Fischer, S.; Grauel, B.; Alivisatos, A.P.; Resch-Genger, U. Quantum yields, surface quenching, and passivation efficiency for ultrasmall core/shell upconverting nanoparticles. *J. Am. Chem. Soc.* **2018**, *140*, 4922–4928. [[CrossRef](#)]
121. Sasidharan, S.L.; Niagara, M.I.; Li, Z.Q.; Huang, K.; Soo, K.C.; Zhang, Y. Titania coated upconversion nanoparticles for near-infrared light triggered photodynamic therapy. *ACS Nano* **2015**, *9*, 191–205.
122. Rong, J.M.; Li, P.C.; Ge, Y.K.; Chen, H.L.; Wu, J.; Zhang, R.W.; Lao, J.; Lou, D.W.; Zhang, Y.X. Histone H2A-peptide-hybridized upconversion mesoporous silica nanoparticles for bortezomib/p53 delivery and apoptosis induction. *Colloids Surf. B* **2020**, *186*, 110674. [[CrossRef](#)] [[PubMed](#)]
123. Chhetri, B.P.; Karmakar, A.; Ghosh, A. Recent Advancements in Ln-Ion-Based Upconverting Nanomaterials and Their Biological Applications. *Part. Part. Syst. Charact.* **2019**, *36*, 1900153. [[CrossRef](#)]
124. Han, S.Y.; Qin, X.; An, Z.F.; Zhu, Y.H.; Liang, L.L.; Han, Y.; Huang, W.; Liu, X.G. Multicolour synthesis in lanthanide-doped nanocrystals through cation exchange in water. *Nat. Commun.* **2016**, *7*, 13059. [[CrossRef](#)]
125. Bogdan, N.; Rodriguez, E.M.; Sanz-Rodriguez, F.; De La Cruzm, C.I.; Juarranz, A.; Jaque, D.; Sole, J.G.; Capobianco, J.A. Bio-functionalization of ligand-free upconverting lanthanide doped nanoparticles for bio-imaging and cell targeting. *Nanoscale* **2012**, *4*, 3647–3650. [[CrossRef](#)]
126. Xie, X.H.; Song, J.L.; Hu, L.Y.; Zhang, S.Y.; Wang, Y.T.; Zhao, Y.L.; Lu, Q. Tailor-made PL-UC-C3 nanoparticles for fluorescence/computed tomography imaging-guided cascade amplified photothermal therapy. *Int. J. Nanomed.* **2018**, *13*, 7633–7646. [[CrossRef](#)]
127. Tian, L.X.; Guo, H.Q.; Li, J.; Yan, L.S.; Zhu, E.Z.; Liu, X.M.; Li, K.X. Fabrication of a Near-infrared Excitation Surface Molecular Imprinting Ratiometric Fluorescent Probe for Sensitive and Rapid detecting Perfluorooctane Sulfonate in Complex Matrix. *J. Hazard. Mater.* **2021**, *413*, 125353. [[CrossRef](#)]
128. Su, H.; Luo, J.X.; Zhao, J.B. A Commercial Enzyme Inhibition-Colorimetric Assay Kit for Rapid Detection of Pesticide Residues was Developed for the Determination of Sensitivity to 10 Organophosphorus Pesticides. *Food Saf. Food Qual.* **2019**, *10*, 5512–5516.
129. Zhang, B.H.; Wei, W. Study on the Effect of Enzyme Inhibition Method in the Detection of Pesticide Residues in Vegetables. *South China Agric.* **2018**, *12*, 105–106.
130. Li, Z.M.; Wang, Y.; Leng, H.X.; Li, X.A.; Kang, X. Simultaneous Determination of Clenbuterol and Ractopamine Residues in Food by Protein chip. *Instrum. Anal.* **2019**, *38*, 913–919.
131. Wang, P.Y.; Li, H.H.; Hassan, M.M.; Guo, Z.; Zhang, Z.Z.; Chen, Q. Fabricating an Acetylcholinesterase Modulated UCNPs-Cu<sup>2+</sup> Fluorescence Biosensor for Ultrasensitive Detection of Organophosphorus Pesticides-Diazinon in Food. *J. Agric. Food Chem.* **2019**, *67*, 4071–4079. [[CrossRef](#)] [[PubMed](#)]
132. Liu, M.L.; Zhang, L.X.; Jiang, S.; Fu, Z.F. A Facile Luminescence Resonance Energy Transfer Method for Detecting Cyano-Containing Pesticides in Herbal Medicines. *Microchem. J.* **2020**, *152*, 104451. [[CrossRef](#)]
133. Chen, H.; Ding, Y.; Yang, Q.; Barnych, B.; González-Sapienza, G.; Hammock, B.D.; Wang, M.H.; Hua, X.D. Fluorescent “Turn off-on” Small-Molecule-Monitoring Nanoplatform Based on Dendrimer-like Peptides as Competitors. *ACS Appl. Mater. Interfaces* **2019**, *11*, 33380–33389. [[CrossRef](#)] [[PubMed](#)]
134. Xie, G.L. Detection Technology of Veterinary Drug Residues in Food of Animal Origin. *Sci. Technol. Inf. Anim. Husb. Vet.* **2019**, *11*, 63.
135. Hlavacek, A.; Farka, Z.; Hubner, M.; Hornakova, V.; Nemecek, D.; Niessner, R.; Skladal, P.; Knopp, D.; Gorris, H.H. Competitive Up-conversion-Linked Immunosorbent Assay for the Sensitive Detection of Diclofenac. *Anal. Chem.* **2016**, *88*, 6011–6017. [[CrossRef](#)]

136. Xu, S.; Xu, S.H.; Zhu, Y.S.; Xu, W.; Zhou, P.W.; Zhou, C.Y.; Dong, B.; Song, H.W. A Novel up-conversion, Fluorescence resonance Energy Transfer Biosensor (FRET) for Sensitive Detection of Lead Atoms in Human Serum. *Nanoscale* **2014**, *6*, 12573–12579. [[CrossRef](#)]
137. Yuan, L.; Lin, W.Y.; Xie, Y.N.; Chen, B.; Zhu, S.S. Single Fluorescent Probe Responds to H<sub>2</sub>O<sub>2</sub>, NO, and H<sub>2</sub>O<sub>2</sub> with Three Different Sets of Fluorescence Signals. *J. Am. Chem. Soc.* **2012**, *134*, 1305–1315. [[CrossRef](#)] [[PubMed](#)]
138. Kaur, K.; Bhardwaj, V.; Kaur, N.; Singh, N. Fluorescent Primary Sensor for Zinc and Resultant Complex as Secondary Sensor towards Phosphorylated Biomolecules: INHIBIT Logic Gate. *Inorg. Chim. Acta* **2013**, *399*, 1–5. [[CrossRef](#)]
139. Tang, L.J.; Cai, M.J.; Zhao, J.; Zhong, K.L.; Hou, S.H.; Bian, Y.J. A Highly Selective and Ratiometric Fluorescent Sensor for Relay Recognition of Zinc(ii) and Sulfide Ions Based on Modulation of Excited-state Intramolecular Proton transfer. *RSC Adv.* **2013**, *3*, 16802–16809. [[CrossRef](#)]

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