Synthesis Techniques for rare Earth doped up-conversion Nano-materials for Solar cells – A brief Review

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Abstract : Extended efficiency of solar cells to ensemble more solar energy as well as its optimum conversion and utilization is believed to be a major challenge in current times. The spectral mismatch between the distribution of energy in the solar spectrum incidence and the semiconducting material band gap is a major restriction in the performance of solar cells. The conversion of wavelength of the sun is a necessary requisite to reduce spectral disruption. Of late, the solar cell converters are presumed as up-converted components and products derived from down conversion. Materials like NaCsWO₃, NaYF₄, and NaYF₄: Yb, Er are synthesized and used to overcome the problem like deficiency of up-conversion luminescence (UCL) materials and device structures. The intensity of UCL can be enhanced by a significant time when the amount of NaCsWO₃ is 2.8 m mol per cent. UCL material is considered as one of the best approaches to obtain high-efficiency perovskite solar cells (PSCs). In order to overcome these difficulties, not only were these effective up-conversion nano-particles (UCNPs) doped into the hole layer but the perovskite foil was also modified in PSCs. The highest power conversion (PCE) performance reached 18.89%. Enhanced UCLs allow for UCNPs to extend the recognition spectrum of near PSCs. The objective of this comprehensive and focused review is to highlight the different synthesis techniques used in up-conversion nano-materials, for solar cell applications along with a theoretical perspective in this regard.

1. Introduction

The global energy sector in general and solar powered energy utilization in particular is expected to rise manifold by 2050. Fossil fuels are the most exploited means of energy today. They are becoming more costly and dangerous for our ecology with each passing day. The quest for another global energy source has been initiated by finite generation and the surrounding issues linked with fossil fuels. Solar energy, biomass, hydropower, wind energy, hydrogen fuel cells are considered much more reliable and cleaner renewable sources of energy [1–3].

Sunlight is proven to be a best and widely available source of energy as it radiates almost 1002 X 10⁴ of photovoltaic electricity to the surface of planet earth. Recent studies have indicated that the use of PV technology in solar power generation can meet our energy requirements pertaining to commercial, industrial as well domestic purposes. In simple words, Solar cell is a photovoltaic instrument which inhibits a property of directly transforming the light photons into power (electricity). The advancement of solar energy technologies is expected to provide a prosperous future for solar cells with the largest energy economy. At very initial stages, a very small fraction of [4–7]light was
converted to PV (approximately one percent) by semiconductors (preferably silicon). The process of doping i.e. adding impurities (charge carriers) along with the advanced synthesis techniques, have successfully enhanced the efficiency to a large extent. Environmentally, solar cells would power the challenging energy areas from moving-to-moving stationary electronics. Photovoltaic cells can be categorized into a class of 1st, 2nd, and 3rd generation (both single-crystalline and multi-crystalline) [8–11], see figure 1.

**Figure 1.** First, second and third generation Photovoltaic Technologies

Increased visible light absorption of PSCs, leads to spread-off smooth-out effects which enables PSCs to generate more photocurrent. UCNPs could change the film perovskite by compensating the holes and gaps on the border [12–14]. Removal of perovskite surface defects mark lower carrier recombination and improves the PSC efficiency. When it comes to manufacturing first-generation solar cells, a thin slice of semiconductor called silicon wafers are used widely. The silicon solar cell technology is the most popular and widely accepted technology due to brilliant results.

Unfortunately, they are costly to frame as the cost for consumer purification of silicon substrates is high. The 2nd generation photovoltaic cells are made to eliminate the practice of cost-effective materials. The cost per watt in the 2nd generation is very less as compared to that of the 1st stage because of low-cost thin-film photovoltaic cells. Thin-film solar cells like Copper Indium Gallium Diselenide Solar Cells (CIGS) and cadmium telluride (CdTe) have the topmost cell efficiency of 20.4 and 13.4% in the second generation family. Thin-film solar cells remain less effective when compared to silicon cells of first stage. Most of the 2nd generation cells depend on substances that are rare and toxic in nature. As per the Shockley-Queisser limit, if a single-joint photovoltaic cell has a band-gap of 1.1 eV, the maximum efficiency it can achieve only 31% [15–17].

Third generation photovoltaic cells excel the Shockley-Queisser limit and are considered as an essential solution to the expensive solar cells of the First and second generation. They are flexible straightforward frames and can be printed on a wide range of adjustable substrates. This includes solar cell technology such as colloid solar cells, DSSCs, organic photovoltaics, and solution-processed bulk inorganic photo-voltaic. The third and current generation technology has brilliant capacity to produce rational high-performance solar cells [18–22].
Upconversion (UC) includes two or three photons that are absorbed to achieve light emission of less wavelength than the excited wavelength. Transformation can take place through various pathways in both organic and inorganic materials. Polycyclic aromatic hydrocarbons are organic particles which can obtain conversion by the threefold destruction, absorption enhancement. Inorganic elements also carry f and d ions. In case of photon upconversion, there are usually three mechanisms in inorganic substances, while two are in organic substances. Photon upconversion in inorganic substances occurs with the help of energy transfer upconversion (ETU), excited-state absorption (ESA), and photon avalanche (PA). These two physical processes have a common effect with upconverting emission of photons of lesser wavelength than excitation. In 1959, Nicolaas Bloembergen invented this in an early project, a solid-state IR quantum board that François Auzel first observed in 1966.

2. Upconverting Nano-particles

In bulk crystals and optical fibers, photon conversion was analyzed primarily leading to nanomaterials growth. In 1990s, the work on nanotechnology was widespread and lanthanide-doped nanostructure marked a crossroad of modern science on lanthanides. Lanthanide-doped nanoparticles are in actual translucent nanocrystals doped with certain quantities of lanthanide ions more often NaYF₄, NaGdF₄, LiYF₄, YF₃, CaF₂ or oxides such as Gd₂O₃ pairs of thulium-ytterbium (Tm³⁺, Yb³⁺) or erbium-ytterbium (Er³⁺, Yb³⁺). The more common lanthanide ions for photons shift Yb³⁺ ions which capture light at approximately 980 nm and pass it to the upconverting ion. If the ion is erbium, an irregular red and green emission is seen while near the ultraviolet region, blue and red light are used in the emission of the upconverting ion; thulium. Crystalline enrich substance and a dopant (emitter) in a conventional inorganic UC phosphorus is applied at a low concentration. The host has a crystalline matrix, while the doping agrees to create the optimum state for these centers. Some metastatic concentrations present in lanthanide ions give UC inorganic phosphors as dopants. The lanthanides contain a group of fifteen chemical components, produced from La and terminated in Lu.

Lanthanide ions have a structure of 4fⁿ 5s² 5p⁶ electrons (0<n>14) as trivalent ions (Ln³⁺) in their most firm oxidation state. Due to the presence of the partially filled 4f inner cell, their significant magnetic and optical properties arise. For lanthanide ions with n electrons, there are 14 desirable configurations possible. Due to large number of energy levels, the sharp f-f transition bands are mostly Ln³⁺ ions because the filled 5s² or 5p⁶ subshells are protected by four electrons with Ln³⁺ ions. The transformation between different 4fⁿ states is limited to Laporte, resulting in significantly long-term (up to 0.1s) and poor transition. The lanthanide ion has an exciting life and absorbs a sequence of ideal second photon energy even at a low excitation level and has the tendency to reach a higher excited state. If the band distance between energy levels in the lanthanide ion is similar, they become excited by monochromatic light to a high degree. But the energy demanded is the same per stage of absorption. This makes upconverted green and red pollution visible. UC pollution is constrained by two factors in single-doped UC phosphors. At high eminent dopant densities, cross-relaxation can be a problem which could depress excitation energy by enhancing the emitter's concentration in phosphors. The Er³⁺ and Tm³⁺ concentration in most UC products is not between 3% and 0.5%. The excitation light cannot be efficiently absorbed by the Er³⁺ and Tm³⁺ at such low concentrations, which lead to low efficiency in UC. Whereas, upconverting green and red emissions, two factors in single-doped UC phosphors show limiting UC emissions.

3. Synthesis Techniques of UCNPs

The various Techniques of UNCPs are discussed below. We have provided a focused approach based on the various techniques along with their related crucial aspects.
3.1. **Decomposition by using Thermal Energy**

The available investigations signify as per the studies, it is observed that the organometallic precursors will decompose if we use a high boiling point of 300 to 350°C along with the long alkyl chain surfactants. Among all the available organo-metallic precursors today, metallic trifluoroacetate salts and rare-earth oleates are used most frequently. This is because the surfactants employed could be oleic acid (OA), oleyl amine (OM) or trioctylphosphine oxide (TOPO) and the solvent used is 1-octadecene (ODE). This method produces UC nanocrystals that are rich in quality, uniform in size, distribution and have exceptional optical properties. These nano-crystals have shown the property of high crystallinity.

The details regarding different hosts, as well as their shape and size distribution is mentioned in Table 1. These researches performed in the past are associated to thermal energy. One could observe that different types of host’s correspond to different shape and size distribution.

| Host    | Shape                        | Size distribution (nm) |
|---------|------------------------------|------------------------|
| NaYF₄   | Sphere, nanorod, nanocube,   | 15-60 narrow           |
|         | Nanoplate, nanoprism         |                        |
| NaGdF₄  | Sphere                       | 2-30 narrow            |
| NaYbF₄  | Sphere, polyhedron           | 15-60 narrow           |
| NaScF₄  | Sphere, hexagonal            | 20-40 narrow           |
| LiREF₄  | Polyhedron, rhombic,         | 20-90 narrow           |
|         | Nanoplate                    |                        |
| KREF₄   | Nanowire, nanocube and       | 7-20 narrow            |
|         | nanopolyhedra                |                        |
| MF₂     | Sphere, nanocube,            | 3-100 narrow           |
|         | Nanowire                     |                        |
| MFCl    | Nanocube                     | 12-20 narrow           |
| KMnF₃   | Nanocube                     | 10-40 narrow           |
| NaMnF₄  | Sphere                       | 8-10 narrow            |
| RE₂O₃   | Nanoplate, nanodisk          | 5-10 narrow            |
| YOF     | Nanosphere                   | 14-16 narrow           |

3.2. **Hydro Solvothermal method**

This method involves using solvent under a given pressure and at a temperature that is above its critical value. This adds to the improvement of reactivity and solubility of antecedents that as a result promote fine crystallization of the finished products. Specialized reaction vessels called autoclaves are used to carry out these reactions. These vessels are of 20 inner Teflon liners, an outer stainless-steel
shell with a cap of stainless steel. This method is used to obtain products that usually have few defects and high crystallinity [16], see table 2.

| Host        | Surfactant           | Shape                  |
|-------------|----------------------|------------------------|
| NaYF$_4$    | OA(oleic acid)       | Sphere, Nanoplate,     |
| NaYF$_4$    | OA(oleic acid)       | Nanorod, nanodisc     |
| NaYF$_4$    | Cit$^3$-             | Microplate             |
| NaYF$_4$    | EDTA(Ethylenediaminetetraacetic acid) | Sphere |
| NaYF$_4$    | PVP(Polyvinylpyrrolidone) | Sphere |
| NaYF$_4$    | PEI(Polyethylenimine) | Sphere                 |
| NaYF$_4$    | PAA(Polyacrylic acid) | Nanorod                |
| NaGdF$_4$   | OA(oleic acid)       | Sphere, polyhedron     |
| NaREF$_4$   | OA(oleic acid)       | Polyhedron, Nanorod,  |
|             |                      | nanodisc               |
| GdF$_4$     | EDTA(Ethylenediaminetetraacetic acid) | Sphere |
|             | PEI(Polyethylenimine) | Sphere                 |
| BaY$_2$F$_8$| OA(oleic acid)       | Nanobelt               |
| Ba$_2$GdF$_7$| EG                   | Pseudo octahedron      |
| Ca$_x$YF$_{3+2x}$ | CTAB(Cetyltrimethylammonium bromide) | Micro-cubic, rhombic  |
| CaF$_2$     | OA(oleic acid)       | Sphere                 |
| SrF$_2$     | OA(oleic acid)       | Sphere                 |

3.3. **Co-Precipitation Method**

Co-precipitation is one of the early and most practical UCNP synthesizing methods. The surfactants have a significant role in regulating the microstructure's scale, morphology, transformation and magnetic properties. Although the co-precipitation process is straightforward, it also requires post-heat (annealing) therapy to promote UC fluorescent intensity. It allows the NPs to accumulate and grow.
The hydrophilicity of products reduces after certain processes like organic capping reagents and heat treatments to increase ductility, see table 3

Table 3. Details of Host, Surfactants in CO-precipitation method

| Host       | Surfactant                          | Shape                  |
|------------|-------------------------------------|------------------------|
| NaYF₄      | EDTA(Ethylenediamine tetraacetic acid) | Sphere                |
| REF₃       | Ammonium di-n-octadecyldithiophosphate | Elongated particle   |
| NaGdF₄     | Na₂EDTA,PVP,SDS                      | Rod shape aggregate   |
| LaF₃       | Ammonium di-n-octadecyldithiophosphate | Sphere               |
| La₂O₃      | Urea                                | Nano-aggregate,nanosphere |
| Y₂O₃       | CTAB(Cetyltrimethylammonium bromide) | Sphere                |

3.4. Microwave-Assisted Synthesis
In recent years, the UCNPs synthesis has attracted a great deal of interest through the microwave-assisted approach. Microwave heating is better than traditional heating as it provides benefits such as eminent heating rates and less reaction time. This type of heating also poses an unwanted side reaction [13].

The effect of several parameters for the pure phase forming of NaYF₄ was investigated by Yang and co-workers: Yb, Er. They have displayed that the state of developed nanoparticles will eventually be changed by enhancing the NH₄F quantity of EG solvent, see table 4.

Table 4. Details of Host, Surfactants, shape in microwave synthesis

| Host      | Surfactant          | Shape                      |
|-----------|---------------------|----------------------------|
| NaYF₄     | OA(oleic acid)      | Hexagonal and cubic        |
| NaYF₄     | OA(oleic acid)      | Sphere, bipyramidal microdisk |
| NaGdF₄    | PEI(Polyethylenimine) | Quadral cubic             |
| BaYF₃     | PEI (Polyethylenimine) | Sphere                    |

3.5. Ionic Liquids Assisted Synthesis
Liquids are said to be one of the best mediums for the synthesis of nanomaterials owing to their low viscosity and high resilience qualities. They also have minimum vapor pressure and better ionic conductivity that helps in the synthesis process. Furthermore, ionic liquids possess the highest dissolution and stabilization capability for metallization which enables many of them to function in inorganic synthesis of solvents, caps or surfactants. Ionic fluids are considered to be as excellent microwave absorbers and are also used for microwave heating as solvents and F- sources. This process decelerates the reaction rate considerably (5–20 min) which leads to the production of strong crystallized products.

3.6. Gas-Phase Synthesis Methods
This approach incorporates the pulsed laser vaporization characteristics of the target material with a regulated vapor condensing technique. This process creates very crystalline upconverting nanocrystals and removes the post-reinforcement process that is generally essential for many other processes. Another probable technique is a gas-phases flame montage, which is a single-step process that can yield highly pure nanocrystal quality of low primary size.

4. Solar Cell Applications
In 1996, Gibert first proposed an optimum utilization of the use of upconverting phosphors to enhance the solar cell capacity. The gas solar cell was consolidated by a material that is doped by Yb\(^{3+}\) and Er\(^{3+}\) with an efficiency of 2.5% at elevated excitation levels. Many research groups subsequently participated in the production of upconversion solar cells. The components for upconverting doped individually by Er\(^{3+}\) or Ho\(^{3+}\) are ideal for fine c-Si solar cells of 1100 nm as light can be used in long wavelengths 1500 nm with short-well-length 5 and visible emissions 980, 800, 660, 550 nm.

4.1. First-Generation Photovoltaic Cell
A first-generation solar cell is also termed as a 1st generation photovoltaic cell. It consists of a single layer p-n junction diode, single-crystal with a comparatively larger area. These cells are used to generate a notable amount of electrical energy from light sources with the wavelengths of sunlight. To make these cells, silicon wafers are employed in the diffusion process.

4.2. Second Generation Solar Cells
There are mainly three types of solar cells that fall in the category of second-generation solar cells, one is silicon while the other two are made from non-silicon materials called CIGS and CdTe.

4.3. Third-Generation Photovoltaic Cells
The class of solar cells which have the ability to overcome the Shockley-Queisser limit of 31-41% power efficiency for a single band-gap solar cells are commonly called third-generation solar cells. It consists of a range of alternatives to the cells of the 1st and 2nd generation respectively. The most common third-generation systems comprise of multi-layer cells from amorphous silicon or gallium arsenide.

4.4. Fourth-Generation Photovoltaic Cells
These cells have emerged into a whole new level of application that is referred to as the latest solar cell technology. In this technology, the organic and inorganic materials are combined in such a way that it increases efficiency and reduces the overall cost of solar cells, see table 5.

| Dopant ion | Host lattice | Excitation | Emission (nm) | Solar cell |
|------------|--------------|------------|---------------|------------|
|            |              |            |               |            |
| Er<sup>3+</sup> | NaYF<sub>4</sub> | 1523 | 550,660,800,980 | c-Si |
| Er<sup>3+</sup> | CaF<sub>2</sub> | 1550 | 660,980 | c-Si |
| Er<sup>3+</sup> | Gd<sub>2</sub>(MoO<sub>4</sub>)<sub>3</sub> | 1530 | 545,665,800,980 | c-Si |
| Er<sup>3+</sup> | NaGdF<sub>4</sub> | 1530 | 527,540,653 | c-Si |
| Er<sup>3+</sup> | Y<sub>2</sub>O<sub>3</sub> | 1538 | 562,659,801,987 | c-Si |
| Er<sup>3+</sup> | Gd<sub>2</sub>O<sub>3</sub>S | 1510 | 540,660,820,990 | c-Si |
| Er<sup>3+</sup> | Fluoride glass | 1532 | 550,660,820,980 | c-Si |
| Er<sup>3+</sup> | BaY<sub>2</sub>F<sub>8</sub> | 1557 | 540,670,800,970 | c-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup>, In<sup>3+</sup> | LiNbO<sub>3</sub> | 1550/980 | 530,558,672 | c-Si/a-Si |
| Ho<sup>3+</sup> | Glass ceramics pbF<sub>2</sub> nanocrystal | 1170 | 650,910 | c-Si |
| Yb<sup>3+</sup>, Ho<sup>3+</sup> | Fluorindate glass | 1155 | 550,650,750,905,980 | c-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | NaYF<sub>4</sub> | 980 | 525,540,640-660 | a-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup>, Gd<sup>3+</sup> | NaYF<sub>4</sub> | 980 | 540,660 | a-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | NaYF<sub>4</sub> | 980 | 520,538,656 | a-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | NaYF<sub>4</sub> | 1560,980 | 540,650,803,980 | a-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | Gd<sub>2</sub>O<sub>3</sub>S | 980 | 510-560,650-680 | a-Si |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | LaF<sub>3</sub> | 980 | 543,655 | DSSC |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | YF<sub>3</sub> | 980 | 525,545,656 | DSSC |
| Yb<sup>3+</sup>, Er<sup>3+</sup> | NaYF<sub>4</sub> | 980 | 520-570,650-700 | DSSC |
| Yb<sup>3+</sup>, Er<sup>3+</sup>, Fe<sup>3+</sup> | NaGdF<sub>4</sub> | 980 | 525,540,653 | DSSC |

5. **Recent updates and Theoretical Perspective**
A fundamental and extensive approach to enhance the solar cell efficiency is available. A brilliant review of the theoretical and experimental research performed in this domain is mentioned in. Various types of rare-earth elements up-conversion nano-particles with respect to perovskite cell has been discussed. A more specific review on the recent advances in using rare-earth doped nano-materials in mesoporous electrodes, perovskite cells is available. Loss of energy in perovskite solar cells and better
procedures, methods employed to manage the photons in PSCs to make them cost-competitive is described. The best utilization of widely available solar energy through extensively efficient solar cells to address the ever-increasing demand of energy in present and future is an extremely hot topic of research. Renewable sources will soon deplete and humans cannot rely on them in the longer run. Moreover, they cause huge environmental degradation and long lasting impact on ecology and environment. The best doping methods with effective host material to amass the solar energy is a challenging task. The theoretical investigations and predictions through simulations by optimizing the characteristics, physical properties and interplay of host compound, could provide useful advancements for more effective conversion of sunlight to electricity. The use of up-conversion nanoparticles in solar technology is expected to revolutionize the domain of optimum utilization of solar energy. The doping could enhance the performance of cells by tapping more radiation and then covert that light to PV energy.

● How much concentration is required to achieve the best conversion rate in order to garner the solar energy could be estimated through simulation models.
● The theoreticians can develop models and compare the results with available experimental data at occasions to find out the type and quality of different type of doping materials which can yield best results.

A theoretical effort pertaining to high photoluminescence quantum efficiency with respect to Red light emitting inorganic La$_2$CaZnO$_5$ is also available in literature.

6. **Summary and Outlook**

We presented various methods (with different dopants, host materials) to synthesize solar cells with enhanced efficiency and discussed the related fundamental aspects. It is evident that there is a spectral mismatch between the solar radiations falling on planet with reference to many types of semiconductors. The major problem that affects the effective solar cell execution is the non-absorption of sub band photons by semiconductors. This obstructs the performance of solar cell to a large extent. The available literature indicate that a wide range of spectral converters such as up-conversion nanomaterials have the capability to convert large fraction of light into visible band. The information provided in the review will provide impetus to advanced research to mark progress in the application of nano-materials for up-conversion in the synthesis techniques of solar cells. Moreover, the methods reviewed and discussed in article may provide specific inputs to theorists. Assembling the various properties of different host materials theoretical Physicists could suggest best configuration and composition to manufacture solar cells with high conversion rate from solar energy to electricity. More experimental research is necessary to raise the conversion rate of solar cells with best host materials which could provide economical and clean source of energy. In a fair assumption, we believe that even if 0.1 % of solar radiation energy falling on earth could be trapped with about more than 20 % of conversion rate, the most of our energy requirements (commercial, domestic, industrial, or research etc) may be fulfilled.

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