

**MINI-REVIEW: FLUORESCENT ORGANIC NANOPARTICLES****D. K. Dalavi**Associate Professor, Department of Chemistry,  
Bhogawati Mahavidyalaya, Kurukali, Affiliated to Shivaji University, Kolhapur.**ABSTRACT:**

Fluorescent Organic Nanoparticles have increasing attention in research due to the wide variety of available organic molecules, the flexibility in material synthesis, their excellent water solubility, and favorable optical properties. FONs are emerging as a new class of fluorescent probe for sensing and estimation of carcinogens from the environment, bio-molecules in pharmaceutical samples and DNA due to their peculiar size dependent optical and electronic properties and superior photostability and excellent resistance to chemical degradation or photodegradation. The different kinds of methods used for the preparation of FONs are the supercritical fluid crystallization, microwave-irradiation reprecipitation, emulsion, laser ablation and reprecipitation method. The methods are employed under different conditions of temperature, precipitation time, template and stabilizer to acquire organic nanoparticles with different structures, various shapes, and of diverse sizes. The unique and more applicable properties of the prepared nanoparticles have as compared with the property of the monomer and bulk materials.

**INTRODUCTION**

Fluorescent Organic Nanoparticles (FONs) have created considerable interest since last decade owing to selective binding ability towards analyte molecules. FONs have inspired growing research efforts due to great diversity of organic molecules available, the flexibility in material synthesis, good water solubility and optical properties.<sup>1-7</sup> Inorganic materials and metal nanoparticles are reported successively in recent years but least attempts have been made to design and characterize the FONs. FONs have recently become a focus of considerable interest for sensor applications. Nonetheless, significant obstacles persist in both their development and practical use. Organic nanoparticles can be commonly described as solid particles composed of organic compounds (mainly lipids or polymeric or aromatic or polyaromatic compound) ranging in diameter from 10 nm to 1  $\mu\text{m}$ . For examples- Perylene nanoparticles, Tetracene nanoparticles, Anthracene nanoparticles, Pyrene nanoparticles, Micro/nanostructured conductive polymers, J-aggregated merocyanine microcrystals etc.

**1. Interest in FONs:**

Numerous attempts have been made to generate metal nanoparticles and inorganic semiconductor, however, there are only few and latter-day approaches to organic nanostructures. The emission color of semiconductor nanocrystal QDs is tunable by the size due to the quantum confinement effect.<sup>8</sup> Harnessing such emission for practical applications in optoelectronic devices and biological fluorescence marking is a vital challenge, which imposes stringent requirements of high stability and high fluorescence quantum yield against photodegradation.<sup>9</sup> These features are difficult to attain in semiconductor nanocrystals modified by organic ligands due to imperfect surface passivation. However, the organic ligands are labile for exchange reactions because of their weak bonding to the nanocrystal surface atoms. A verified strategy for enhancing the stability and fluorescence quantum yield is to raise a shell of a higher band gap of semiconductor on the core nanocrystals.<sup>9</sup> In such materials, the type and thickness of shell, provide further control for designing the electrical, optical, chemical and electronic properties of semiconductor nanocrystals. Additionally, the optoelectronic materials and devices based on nanowires have been extensively investigated. Most of the reported nanowires were prepared from inorganic substances. However, small organic molecules were used in the construction of 1-Dimensional nanomaterials with well-defined structures. Compared to inorganic

nanoparticles, organic nanoparticles have their own advantages and applicable potentials because they allow much more variety and flexibility in materials synthesis and nanoparticle preparation.<sup>1,4</sup>

#### OVERVIEW OF FONs:

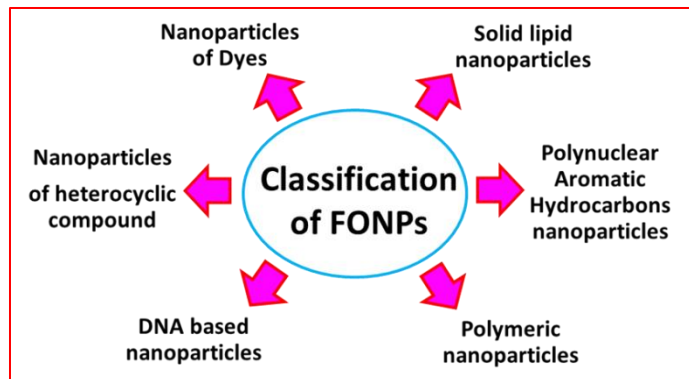
FONs are emerging as a new class of fluorescent probe for sensing and estimation of carcinogens from the environment, bio-molecules in pharmaceutical samples and DNA due to their peculiar size dependent optical and electronic properties and superior photostability and excellent resistance to chemical degradation or photodegradation. Methods used for the preparation of organic nanoparticles are mainly based on either reprecipitation, milling or through chemical reaction techniques.<sup>9,10</sup> As far as the application is concerned, FONs are expected to hold the higher potentials because they allow much more variability and flexibility in materials synthesis and nanoparticle preparation.<sup>11,12</sup> Functional ONPs have been attracted in a scientific and technological area of materials. Several kinds of functional organic nanocrystals relating to opto-electrical materials, nano-medicine and bio-photonics are mentioned. ONPs occupy the intermediate state between isolated molecules and the bulk crystal. It is known that the nanoparticles have unique physicochemical properties that are not observed in their bulk materials due to the changes in lattice state caused by an increase in the surface area of nanoparticles and therefore, to lattice softening. It is believed that, lattice softening results in weaker interaction between adjacent molecules, finally resulting in wider band gaps. Thus, it is confirmed that not only can the wide panel of physical properties provided by functional compounds be fully exploited, but also these properties modulated by the particle size and shape.

Unfortunately, the investigation of organic nanoparticles has been paid little attention, as compared to the inorganic, metallic and semiconductor nanoparticle until several articles are reported on FONs. However, organic nanoparticles are of attention not only because they represent an intermediate occurrence between single molecules and bulk materials but also they offer potential applications in science and technology. In 1992, Kasai *et al.* firstly reported a novel reprecipitation method for organic nanoparticles.<sup>13</sup> It has demonstrated that the size of organic microcrystals found to be easily controlled by the preparation conditions such as choice of good solvents and poor solvents, dropping conditions into water, temperature, surfactant and treatment of UV-irradiation. Despite of the range of 100 nm, size dependent optical properties were perceived for these organic nanocrystals, an order of magnitude larger than the scale where the inorganic nanocrystals generally exhibit size dependent optical properties.<sup>11</sup> These aqueous suspension of organic nanocrystals possess great potential for use in fundamental and applied optics.<sup>13</sup> Systematic research work on organic microcrystals have been initiated by Nakanishi and co-workers who have revealed that the optical Kerr shutter responses of polydiacetylene and J-aggregated merocyanine suspension of microcrystals in aqueous medium.<sup>14</sup> It has been shown that the phthalocyanine and PNPs exhibit very diverse and size dependent fluorescent properties from those of bulk samples. This was a significant breakthrough and had a great impact on the future development of "Nanoscience". In aqueous medium, the size-dependent absorption (color) and fluorescence were found in a much larger size range than for metals and semiconductor particles. Detail understanding of these phenomena would contribute to a variety of applications of organics such as color of dyes and pigments, luminescence, photography, photoconductivity, electroluminescence and nonlinear optical effects; by using agricultural chemicals, medicines and so on. Then in upcoming years, a method based on reprecipitation, many kinds of organic nanocrystals successfully prepared from low molecular weight PAHs. The researchers, Yao *et al.* reported similar results with pyrazoline nanoparticles and S. Y. Park and coworkers prepared a new class of organic nanoparticles (CN-MBE nanoparticles) with a mean diameter of 30-40 nm, which have exhibited a strongly enhanced fluorescence emission.<sup>15</sup> H. Y. Kim and coworkers developed preparation method for class of PAHs like nanocrystals of tetracene in aqueous solution.<sup>1</sup> In 2004, Kang *et al.* reported a novel sonication method for preparation of anthracene nanoparticles.<sup>16</sup> In same year, Wang L. *et al.* developed sensor based on ONPs for direct fluorescence quantification of Cr(VI).<sup>16</sup> M. EI-Kemary and H. Yao

reported the synthesis, characterization and optical properties of ONPs of piroxicam anti-inflammatory drug.<sup>17</sup> Wang J. *et al.* in 2007 reported fascinating assembly of one-dimensional organic luminescent nanowires based on quinacridone derivatives.<sup>18</sup> Olive *et al.* published an article on fluorescence amplification in self-assembled ONPs by excitation energy migration and transfer.<sup>19</sup> The self-assembly of a low molecular weight organic chromophore occurs upon reprecipitation in water and yields wide disk-like nanoparticles of 120 nm range as shown by FCS and AFM. Fluorescence polarization spectroscopy also reveals that the efficiency of the donor-to-acceptor energy transfer process is amplified by a donor to donor excitation energy migration.<sup>20</sup> Yilong Lei *et al.* reported phase- and shape-controlled synthesis of single crystalline perylene nanosheets and its optical properties.<sup>21</sup> H. B. Fu and J. N. Yao prepared the nanoparticles of pyrazoline derivative of varying size and their optical properties were studied as a function of size.<sup>3</sup> T. Enseki and H. Yao have developed the new route for the synthesis of FONs of carbocyanine dyes by using an ion association technique.<sup>22</sup> Recently our research group contributed significantly by reporting FONs as a probe for sensing applications.<sup>23</sup> The reprecipitation method have been modified in the laboratory and applied successfully to obtain the surface modified nanoparticles of fluorescent PAHs and their derivatives. The photostability, particle size distribution, surface morphology and sensing ability for fluorescence quenching of the nanoparticles were studied by fluorescence and UV-visible spectroscopy, DLS, Zeta sizer, TEM, and SEM techniques. Aqueous suspension of surface modified nanoparticles exhibited AIEE. This fluorescence enhancement provides an optimal 'on-state' for sensitive 'turn off' quantitative analysis. The analytical fluorimetric method developed in the present work based on quenching and enhancement of fluorescence of nanoparticles satisfy the current demand of novel, selective method of quantitative investigation of specific analyte. In the category of chemical approaches, some of the synthetic methods are based on considerable exhaustive, and therefore time consuming synthetic routes (Sugimoto *et al.* 2002; Spatz *et al.* 1999). As a result, methods which rely on solubility changes to induce nanoparticle formation are recently becoming attractive. Among these methods, a simple less expensive and suitable technique proved to be particularly effective for the preparation of ONPs dispersion is the so-called reprecipitation method.

#### CLASSIFY FONs:

Organic nanoparticles can be designed for wide applications. It can be distinguished on the basis of type of simple molecules. The first one is polymeric nanoparticles constitute by far the most studied organic particles in the literature, elaborated for drug delivery and for biomedicine purposes. Solid lipid nanoparticles (SLNs) are composed of lipid matrices derived generally from glycerol esters of fatty acids. SLNs are highly stable (for several years), good biocompatible and low toxic, hence their nanoparticles are considered as promising drug delivery systems and a good alternative to polymeric nanoparticles especially for parenteral method. The versatility of functionalization, the design and synthesis of heterocyclic compound analogues suited for molecular recognition are feasible. One of the goals in the field of structural DNA nanotechnology is the use of DNA to build up 2- and 3-D nanostructures. The research in this field is encouraged by the significant structural features of DNA as well as by its unique and reversible recognition properties. DNA can serve as a linker or template to form DNA-hybrid structures with other materials.<sup>24</sup> This approach can be used for the improvement of new detection strategies as well as nano-electronic structures and devices using DNA nanoparticles. Many of the FONs belonging to the class of PAHs and dyes are used as fluorescent probe for sensing and estimation of carcinogens from environment, bio-molecules in pharmaceutical samples, nucleic acids and drugs from the metabolic product samples.<sup>25</sup> Exclusively, PAHs are very fascinating class of materials. They exhibit very wide range of fascinating properties such as great color, emission, semiconducting properties, photoconductivity and very distinct photochemical and chemical performance.<sup>26</sup>



### METHODS FOR PREPARATION OF FONPS:

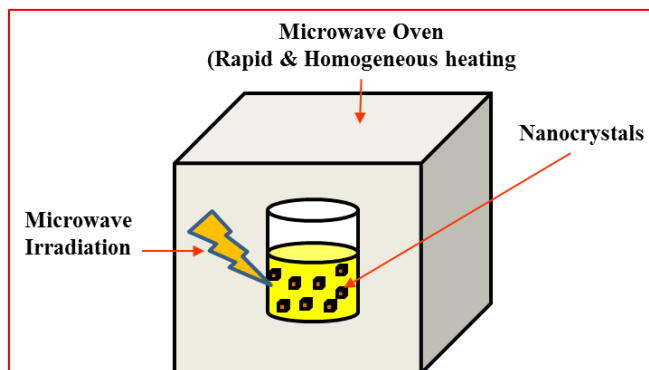
The methods of preparation of inorganic and metal nanoparticles are lengthy, complicated and expensive. In contrast preparation of fluorescent organic nanoparticles is simple, quick and cost effective method. Organic nanoparticles have attracted much attention as advanced materials for applications, such as display elements, inks, toners, drugs, and so on.<sup>27</sup> The different kinds of methods used for the preparation of FONs are the supercritical fluid crystallization, microwave-irradiation reprecipitation, emulsion, laser ablation and reprecipitation method. These methods are employed under different conditions of temperature, precipitation time, template and stabilizer to acquire organic nanoparticles with different structures, various shapes, and of diverse sizes. The unique and more applicable properties of the prepared nanoparticles have as compared with the property of the monomer and bulk materials. It is well-known that properties of many nano-scaled aggregates (QDs, rods, tubes and wires) have unique size dependent properties which are substantially different from that of bulk materials even composed of the same atoms or molecules. The method used for preparation of organic nanoparticles are discussed below.

#### 1 Supercritical Fluid Crystallization Method (SCFC):

Phthalocyanine and few other organic compounds are hardly soluble in organic solvent due to  $\pi$ - $\pi$  stack rigid structure.<sup>28</sup> Komai *et al.* in 1998 overcome the solubility problem by the preparing phthalocyanine nanocrystals using SCFC method.<sup>29</sup> Supercritical fluid (SCF) has been expected to have a high solubility against insoluble organic compounds, since it shows high solubility under high temperatures and high pressure. Thus, the SCF approach proved to be an alternative method for making phthalocyanine nanocrystals, instead of the acid pasting method. The properties of phthalocyanine are very interesting in terms of both optics and electronics in the fields of nonlinear optics, organic photoconductors and pigments.<sup>9</sup>

#### 2 Microwave Irradiation Reprecipitation Method:

The vital role of microwave radiation (2.45 GHz) is that the microwave is well absorbed by water molecule, resulting in effective consistent



**Fig.1:** Experimental setup of microwave-irradiation reprecipitation method.

and rapid heating of water. The uniform microwave heating gives well defined and size controlled organic nanocrystals without aggregation of particles. Baba *et al.* reported microwave irradiation method for a novel fabrication of microcrystals of organic molecules in 2000 and nanowires of diacetylene in 2007 using this method.<sup>9,30</sup>

### 3 Emulsion Method:

The monodispersed PAH nanocrystals fabricated in a stabilizer-free oil-in-water emulsion.<sup>31,32</sup> The organic nanocrystals fabricated by using low-molecular-weight hydrocarbons like benzene, xylene, toluene, tetrachloromethane *etc.* as solvents and emulsifiers. Micro-emulsions are homogeneous in macroscale and micro-heterogeneous in nanoscale dispersion of two immiscible liquids consisting of nano-sized domains of one or both liquids in the other, stabilized by an interfacial film of surface active molecules. Depending on the proportion of various components and hydrophilic– lipophilic balance (HLB) value of the used surfactant micro-emulsions can be classified as water-in-oil (W/O), oil-in-water (O/W) and intermediate bi-continuous structural types that can turn reversibly from one type to the other. The dispersed phase consists of monodispersed droplets in the size range of 5 – 100 nm. The stability of the micro-emulsion can be influenced by salt, concentration of reagents, temperature and pressure.<sup>33</sup>

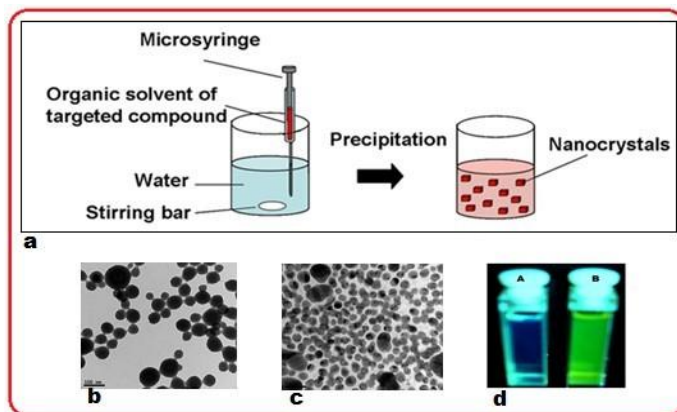
### 4 Laser Ablation Method:

The material used in this method is hydrophobic fluorescent organic compound dissolved in volatile organic solvent as analytical reagent followed by sonication process for particular time in minutes.<sup>34</sup> The suspending/mixture was put into a glass cuvette. The crystals were suspended in organic solvents by stirring vigorously and irradiated by a regenerated amplified femtosecond Ti:sapphire laser pulse (800 nm, 120 fs, 1 kHz). The laser beam was focused to the behind of the cuvette with a 400 mm focal length lens. Regrettably, intense beam of laser light used in laser ablation technique which cause photochemical damage. FONs have recently attracted a significant amount of research interest because of their peculiar size-dependent optical and electronic properties.<sup>20,21</sup> FONs have great potential applications because they allow much more variability and flexibility in material synthesis and nanoparticle preparation. Reprecipitation is a popular method for the preparation of FONs because of its easy and versatile operation.<sup>35</sup>

### 5 Reprecipitation Method:

The reprecipitation method is a bottom-up type approach with a solvent displacement way to prepare organic nanocrystals as their water dispersion. Number of PAHs are allowed to be an applicant for targeting of FONs by using this method. Nakanishi and coworkers proposed a very simple reprecipitation method for the fabrication of organic nanocrystals.<sup>36</sup>

The method involves a rapid mixing of small amount of concentrated stock solution of the target compound dissolved in a good solvent with excess of a poor solvent. The great disparity between the solubilities of the target compound in the good and poor solvents, and the good compatibility of the two solvents are essential for this method. The rapid mixing of the stock solution and the poor solvent changes the micro-environment of the molecules of target compound. The molecules are exposed to the poor solvent surroundings in a very short time, inducing the nucleation and growth of the molecules to nanostructures. The size and morphology of nanostructures can be controlled by changing concentration, temperature, stirring speed, and adding surfactants. Then the precipitation creates nanocrystals dispersed in water. The surface of nanocrystals is modified by cationic, anionic and nonionic surfactants and also using polymers according to the need of sensing analyte molecules. The role of surfactant and polymer is to stabilize nanoparticles also to control the nucleation growth of nanoparticles and prevents further aggregation of nanoparticles into bigger molecules. Different organic compounds were reported to form nanostructures namely  $\pi$ -conjugate compounds, nonlinear optical dyes, perylene dyes, fullerene, 9-anthraldehyde and polymers.<sup>37,38</sup>



**Fig. 2:** (a) Scheme of the reprecipitation method, (b, c) TEM images of FONs, (d) Visible light exposure of aqueous suspension of FONs.

### PHOTOPHYSICAL PROPERTIES OF FONs:

Optical properties are the fundamental of nanoparticles. Thus, the measurement of absorption and emission spectrum is essential to understanding the optical properties and applications of nanoparticles.<sup>39,40</sup> Commonly used techniques include electronic absorption (UV-visible), photoluminescence (PL), infrared red (IR) absorption, dynamic light scattering, Raman scattering, as well as time-resolved which gives information about properties of the materials. Some of these properties are elaborated in this section.

#### 1 UV-Visible Absorption Spectroscopy:

The wavelength of light required for electronic transitions is typically in the UV and visible region of the electromagnetic spectrum, electronic absorption spectroscopy is often called UV-vis spectroscopy. Nanoparticles have optical properties that are sensitive to size, shape, concentration, agglomeration state and refractive index around the nanoparticle surface, which makes UV-vis spectroscopy a valuable tool for identifying, characterizing and studying nanomaterials. It is recently used to obtain qualitative and/or quantitative information regarding the aggregation of molecules into nano/ micro-aggregates from that of the isolated fluorophore which is discussed in H- and J-type aggregation.<sup>35,41,42</sup>

#### 2 H-type and J-type Aggregation:

This phenomenon discussed in terms of the intramolecular and intermolecular effects exerted due to fluorophore aggregation.<sup>1</sup> Intramolecular effects usually owing to the twisted conformations and the rotation of chromophores have a tendency to suppress the radiative process in the dilute solution of isolated fluorophore, whereas planar conformations of chromophores induced in the aggregate state activate the radiation process. Intermolecular effects by aggregation in  $\pi$ -conjugated organic chromophores are correlated with the aggregation geometry such as H or J-type aggregation.<sup>1,35</sup>

#### 3 H-type Aggregation:

H-aggregation in which the face to face interaction of molecules *i.e.* molecules allied parallel to each other with strong intermolecular interaction have a tendency to induce the nonradiative deactivation process. The higher electronic state carries all oscillator strength whereas transitions from the ground state to a lower coupled excited state of molecules are forbidden. Therefore, formation of H-aggregates is characterized by hypsochromically shifted (blue-shifted) absorption peaks with respect that of the isolated fluorophore. In this case, internal conversion from a higher to a lower electronic state is much faster than emission. Hence the emission of the H-structure is effectively quenched.

#### 4 J-type Aggregation:

J-aggregation in which the head to tail (end to end) interaction of molecules leads arrangement into a slanted stack. The transition from the ground state to the lower couple excited state of molecules is allowed

and the J-aggregate state activate the radiation process. Generally, the formation of J-aggregates is characterized by bathochromically shifted (red-shifted) and intensely narrow absorption peaks with respect that of the isolated fluorophore and hence, the emission is stronger.

## CONCLUSION

FONs are emerging as a new class of fluorescent probe for sensing and estimation of carcinogens from the environment, bio-molecules in pharmaceutical samples and DNA due to their peculiar size dependent optical and electronic properties and superior photostability and excellent resistance to chemical degradation or photodegradation.

## ACKNOWLEDGEMENTS

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