

Design and Optimization of a Sustainable Water-Based Ink for Inkjet Printing of Fluorescent Functional Materials

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Short abstract

Research in luminescent materials has received significant interest due to the numerous applications that have emerged, including anti-counterfeiting, display technologies and photovoltaics. A challenge in these applications lies in the deposition of luminescent films. There has been an interest in developing luminescent inks via printing processes, particularly inkjet. The growing emphasis on environmental sustainability is leading research to favour water-based inks. The objective of this work is to develop fluorescent functional materials for water-based inkjet inks.

The luminescent dyes and particles are carefully selected for stability and compatibility with water-based systems. In addition to the water, co-solvents and additives are incorporated to fine-tune surface tension and viscosity, optimizing inkjet ejectability and print quality. The inkjet inks are deposited onto various substrates to evaluate how printing parameters influence fluorescence performance.

Work results demonstrate promising fluorescence uniformity and intensity on different substrates with water-based inks balancing sustainability and functionality. The inks exhibited physicochemical properties compatible with inkjet printing and produced fluorescent thin films with significant emission intensity after printing. Optimizing concentration, thickness and surface interactions show potential for scalable, eco-friendly luminescent coatings with tailored optical properties for applications in displays, security, and sensing.

Keywords: fluorescent ink, water-based ink, inkjet

1. Introduction and background

Photoluminescence refers to the emission of light arising from electronically excited states following the absorption of photons (Valeur and Berberan-Santos 2011). This phenomenon has driven the development of a wide range of luminescent materials, ranging from organic fluorophores to quantum dots exhibiting size-dependent emission properties. Such materials have attracted considerable research interest thanks to their potential applications in numerous fields, including anti-counterfeiting technologies, display devices, photovoltaics and medical diagnostics. However, the environmental and health impacts of these materials and their processing routes have often been neglected in favour of maximizing efficiency. As an example, the toxicity of cadmium (Cd)-based quantum dots (Hu et al. 2021) widely integrated in display technologies pushed to the development of alternative materials such as indium phosphide (InP) quantum dots. Nevertheless, many luminescent inks reported in the literature still rely on organic solvent systems. Fu and al. 2025, for instance, formulated an inkjet ink using a binary solvent system, cyclohexylbenzene and decane. Although this approach avoids the dangerousness of cadmium, the use of organic solvents still raises environmental and health concerns.

Consequently, many reported formulations either rely on organic solvent or struggle to achieve competitive performance when transitioning to more sustainable systems. This highlights the need for luminescent eco-friendly materials.

For many of these applications, the controlled deposition of luminescent films is a critical step. Over the past decade, increasing attention has been devoted to the development of luminescent inks compatible with printing technologies (Xuan et al. 2020). Among the different printing techniques, drop-on-demand piezoelectric inkjet has emerged as a particularly attractive approach due to its flexibility, cost-effectiveness, and high resolution, making it suitable for both research and industrial applications. This non-impact printing technology operates by ejecting droplets of controlled volume onto a substrate. The quality and precision of the printed patterns depend not only on the electrical driving signal (waveform and voltage) but also on the physicochemical properties of the ink (viscosity, surface tension, particles size and colloidal stability). As a result, ink formulation appears as a complex and crucial step of the process, particularly when luminescent particles or dyes are incorporated. These functional materials must remain stable and uniformly distributed within the liquid phase to ensure reliable jetting and homogeneous and efficient luminescent coatings after deposition and drying.

Beyond the technical challenges associated with ink formulation and printing, environmental and health considerations must also be addressed. Water-based inks represent a promising and more sustainable alternative to solvent-based formulations. However, water-based formulations introduce additional challenges. On the one hand, the higher surface tension and slower evaporation rate of water can negatively affect print quality, film formation and adhesion. On the other hand, the fluorescence properties of luminescent compounds are sensitive to their surrounding media [Aquirresacona and al. 1989, Ibnaouf and al. 2014]. Consequently, the interactions between fluorescent compounds and the ink formulation must be carefully considered, with the assessment of the impact of each ink component on the fluorescence. Addressing these challenges requires optimization of both ink composition and printing parameters, as well as the possible incorporation of suitable additives. For instance, to prevent quenching due to aggregation of particles, which is frequently observed with organic fluorophores, Zerbib and al. [2023] introduced phenylenedipropionate 10 anions as spacer within a layered double hydroxide matrix to enhance the efficiency of sulforhodamine B fluorophores.

In this work, we aim to develop functional luminescent inks suitable for inkjet printing. The main challenge lies in incorporating luminescent particles into a water-based fluid while maintaining the physicochemical properties for stable inkjet printing. Particular attention is given to the development of non-toxic and sustainable formulations favouring water-based systems, while optimizing optical performance and durability. The strategy for selecting luminescent materials relies on a close collaboration with the ICCF – Luminescent Materials group, whose expertise in the synthesis of luminescent compounds enables the development of tailored particles specifically designed for integration into water-based inkjet-printable formulations. The inks are deposited onto a range of substrates (polymer film, papers) using inkjet printing to evaluate their ability to produce stable luminescent films and reproduce a broad spectrum of colors obtained by fluorescence. This study focuses on gaining a deeper understanding of the system key parameters, including ink properties and printing conditions, that influence the optical performance of the resulting luminescent film.

2. Materials and Methods

2.1 Materials

Different fluorescent organic dyes were purchased from Sigma-Aldrich: Sulforhodamine B (SRB) and 7-amino-4-methylcoumarin (C120). An aqueous suspension of fluorescent nanoparticles $\text{YVO}_4:\text{Eu}^{3+}$ was provided by the University of Clermont-Ferrand (ICCF, Luminescent materials Group). Glycerol and ethanol were purchased from ROTH and the surfactant TWEEN® 20 was obtained from Sigma-Aldrich. A polyurethane binder (U475) was provided by Alberdingk-Boley. For all experiments, distilled water was used.

Three different substrates were used: a polyethylene terephthalate (PET) film, (MELINEX® ST504, from DuPont Teijin Film), an optical brightener agent (OBA)-free paper (Dune 80 g.m^{-2} from Clairefontaine) and tracing paper (Canson Satin 90 g.m^{-2}). The PET and paper were selected to observe the influence of the infiltration in porous and non-porous substrate. Tracing paper was selected in order to minimize interference during the measurement of the emission spectra of the printed luminescent films.

2.2 Ink formulation

Fluorescent organic dyes were first dissolved in the appropriate solvent: SRB was dissolved in distilled water, while C120 was dissolved in water/ethanol mix. The dyes were weighed and introduced into the solvent at the chosen concentration.

$\text{YVO}_4:\text{Eu}^{3+}$ suspension was diluted at the chosen concentration.

Then, all the solutions and suspensions were mixed at room temperature using a magnetic stirrer (10min). An ultrasonic probe (Branson Sonifier 250) was subsequently used to homogenize the mixtures of C120 and $\text{YVO}_4:\text{Eu}^{3+}$, 5min in total is applied at a cycle output between 20 and 30%.

For ink preparation, the compounds were weighed and placed in a beaker in the following order: fluorescent solution or suspension, ethanol, glycerol and Tween 20. The blend was mixed with the magnetic stirrer (10 min) and stored at room temperature for 24h to 48h prior to printing experiments.

The polyurethane binder was added to the mix at the final stage of the formulation and mixed using a magnetic stirrer (10 min). Different binder contents were investigated (5 wt%, 8 wt%, 10 wt%, 12 wt% and 15 wt%) in order to study their influence on ink adhesion on PET substrates and their impact on fluorescence properties.

This first step allows us to determine the ink's composition and verify the compatibility of the fluorescent particles with each component. Subsequently, the ink's composition will remain fixed, only the concentration of fluorescent particles will change.

2.3 Substrates characterization

The surface roughness of the classical paper and tracing paper substrates was measured using an InfiniteFocus optical profilometer (Bruker Alicona, Graz, Austria). The average roughness (R_a) value of the PET substrate was taken from the manufacturer's datasheet.

The surface free energy of the substrates was determined from contact angle measurements (Optical contact angle goniometers, OCA50, DataPhysics Instruments, Filderstadt, Germany) using the Owens-Wendt model with the following probe liquids: water, glycerol and bromonaphthalene. The excitation and emission spectra of the different substrates were measured using fluorescence and absorption spectrometer (DUETTA, HORIBA Scientific, Kyoto, Japan).

2.4 Ink characterization

Fluorescence of liquid formulations

The absorption spectra of the luminescent fluids were measured using a UV spectrophotometer (Shimadzu UV-1800, Japan) in a 1 cm quartz tank with water as the baseline reference. Emission spectra were recorded under excitation optimized for the fluorescent particles using a UV-Vis spectrophotometer (Ocean Optics HR-6XR500-5, OceanView software, US), with fluorescence intensity corrected for the instrument response.

Ink properties:

The density of the ink was determined by collecting 10 mL of the ink and weighing it. Three measurements were performed for each formulation.

Rheological measurements were carried out using Rheometer MCR 302 (Anton Paar, Ostfildern, Germany), software Rheocompass, equipped with a cone-plate geometry (CP-50, 1° cone angle, 50 mm diameter). The temperature was controlled at 30 °C. The viscosity was measured at a shear rate of 1000 s⁻¹.

Surface tension was measured using a Sigma700 tensiometer (software OneAttention, Biolin Scientific, Sweden) with a platinum De Noüy ring (diameter 1 cm).

Based on the measured ink properties, the jetting behaviour and stability of the ink can be predicted from the dimensionless numbers Reynolds, Weber and **Z**, defined as Derby (2010):

$$\begin{aligned} We &= \frac{v^2 \rho d_n}{\gamma}, \\ Re &= \frac{v \rho d_n}{\eta}, \\ Z &= \frac{\sqrt{\gamma \rho d_n}}{\eta} = \frac{Re}{\sqrt{We}}, \end{aligned}$$

Where γ , ρ and η are the surface tension (N/m), the density (kg/m³) and the dynamic viscosity (Pa.s) of the formulated ink. d_n is the characteristic length corresponding to the nozzle diameter of the Dimatix SAMBA printhead. v is the velocity of the ejection (m/s). The conditions of ejectability predicted by the dimensionless Z number has been defined differently over the years. Jang et al. 2009 found that the processability of an inkjet ink is ejectable for a Z number between 4 and 14. In Derby (2010), the window is limited to $1 < Z < 10$.

2.5 Printing

The formulated inks (2 mL) were loaded into a SAMBA cartridge (Fujifilm, Dimatix) and printed using a Dimatix 2800 (Fujifilm Dimatix, Santa Clara, CA, USA). The SAMBA printhead is equipped with piezoelectric actuators enabling the ejection of droplets through micron-scale nozzles. Prior to printing, the inks were filtered using a syringe filter (pore size 1.2 µm) to prevent nozzle clogging. The driving voltage was set at 25 V and the printing temperature was maintained at 30 °C in order to ensure stable jetting conditions. The waveform parameters were adjusted to obtain stable droplet formation without satellite droplets. Droplet formation was monitored using the stroboscopic camera system of the printer. The volume of the drops is measured using the Dimatix's drop volume measurement function. Square patterns of 1 cm per 1 cm were printed with a drop spacing of 20 µm, allowing the formation of continuous thin film after droplet coalescence on the substrate. For each ink, 3 patterns are printed.

2.6 Fluorescent and print characterization

The morphology and quality of the printed deposit were observed using optical microscopy (ZEISS Axio Imager M1, Carl Zeiss AG, Oberkochen, Germany).

Fluorescence spectrum measurements of the printed layers were performed using a spectrometer fluorescence and absorption spectrometer (DUETTA, HORIBA Scientific, Kyoto, Japan) equipped with a reflection probe. A fixed angle of 45° between the probe and the sample surface was kept using a dedicated probe holder. The color coordinates from CIE L*a*b color space are deduced from the emission spectra.

3. Results and discussion

3.1 Ink formulation: physicochemical properties for inkjet ejectability of fluorescent fluid.



Figure 1: Example of SRB ink formulation, observed on the left under daylight and on the right under UV light (365nm).

Table 1 summarizes the viscosity, surface tension, density, and corresponding Z values of the different ink formulations.

Table 1: Fluid parameters and Z values of the inks with different contents of solvents.

H ₂ O:EtOH:Gly (wt%)	Dynamic viscosity (1000s ⁻¹ , 30°C) (mPa.s)	Surface Tension (mN/m)	Density (kg/m ³)	Z
70:20:10	2,09	33,0	975,7	11,79
70:25:5	2,01	32,4	958,9	11,87
60:30:10	2,39	30,1	965,4	9,63
60:35:5	2,20	30,2	950,6	10,15

The calculated Z values range between 9.6 and 11.9, which falls within the printable window reported in the literature. The obtained values indicate that the formulated inks possess physicochemical properties compatible with inkjet printing and should allow stable droplet ejection without excessive satellite formation.

In addition to jetting stability, the inks must exhibit good adhesion to the substrates, especially on the non-porous PET films. This constraint is less critical for porous substrates such as paper, where drying occurs partly through infiltration in the porous network. In contrast, for PET substrates, the solvent evaporates mainly at the surface, which may result in poor fixation of the dye molecules and in a non-homogeneous distribution of active molecules. To address this issue, a binder was incorporated into the ink formulation to improve adhesion of the fluorescent materials to the substrate. A comparative study was therefore conducted using commercial

polyurethane binder at different concentrations to determine the optimal formulation and ensuring good adhesion while preserving fluorescence properties.

Finally, the compounds of the ink are required to not hinder the fluorescence of the materials. To verify the fluorescence properties, the emission spectra of the formulated inks were measured at an excitation of 365 nm. The spectra, showed in Figure 2, confirm the fluorescent properties of the dyes are preserved after formulation.

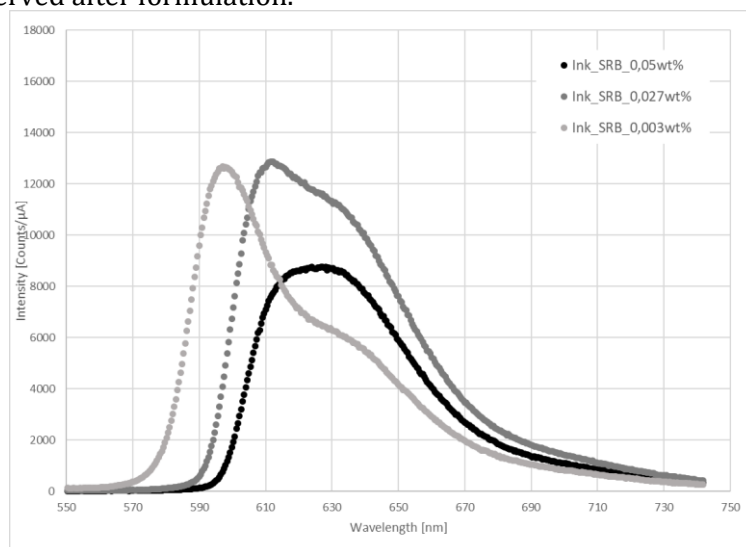


Figure 2: Emission spectra of the formulated inks.

3.2 Characterizations of substrates

The physicochemical properties of the different substrates are summarized in Figure 3 and Table 2.

Table 2: Physicochemical properties of the substrates

	PET	Paper	Tracing paper
Surface free energy	42.5 ± 5.6 mN/m	40.6 ± 3.6 mN/m	29.8 ± 4.8 mN/m
Average Roughness	1.5 nm	1.67 ± 0.06μm	2.53 ± 0.90μm

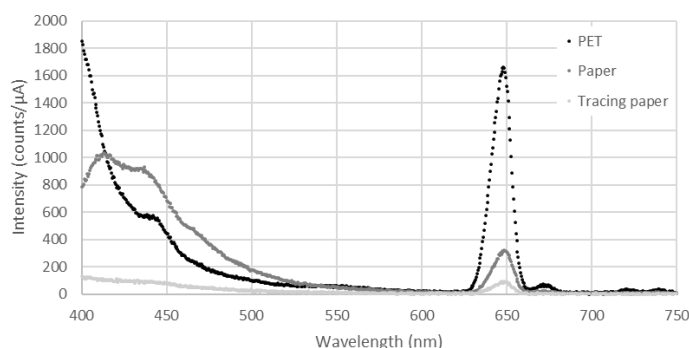


Figure 3: Emission spectra of the substrates for an excitation at 365nm.

The substrates partially absorb UV light radiation, which may influence the fluorescence measurements of the printed films.

3.3 Printing of fluorescent thin film.

The purpose of the printing process is to produce homogeneous fluorescent films with uniform thickness and consistent optical properties. Several inkjet printing parameters influence the final print quality, including the printing temperature, the piezoelectric waveform, the driving voltage and the drop spacing. Proper tuning of these variables is necessary to ensure stable droplet formation and correct printing quality while minimizing defects like non-homogeneous deposition or satellite droplets which can compromise film uniformity and fluorescence performance. Figure 4 illustrates the ejectability window and the droplet formation behaviour of the SRB ink under a driving voltage of 25 V at 30°C.

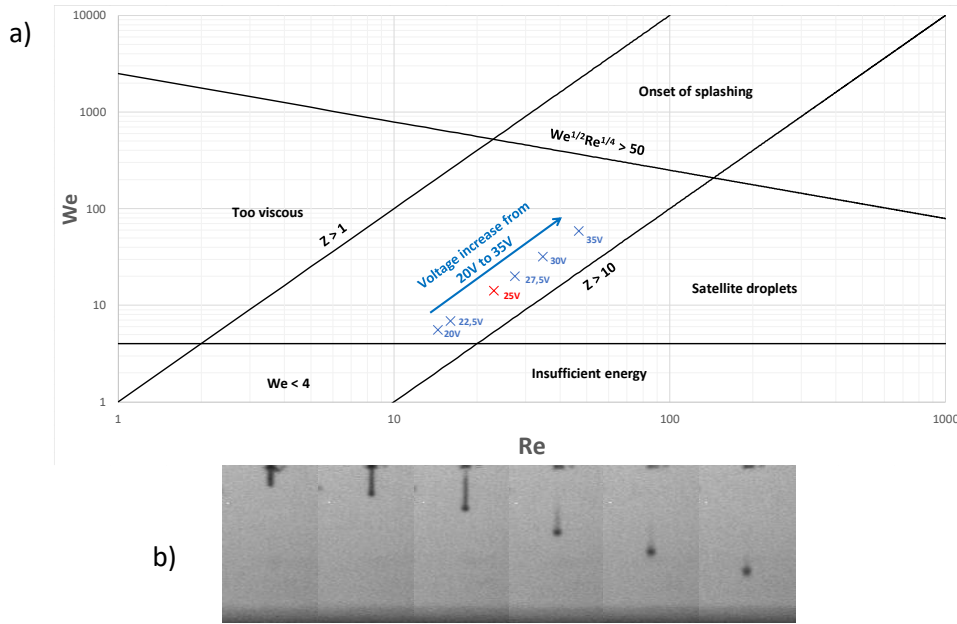


Figure 4: a) Ejectability window for SRB ink defined by Derby (2010) depending on the signal voltage; b) Droplet formation for SRB ink with pulse voltage of 25V at 30°C.

A critical aspect of the process is the analysis of single droplet deposition on the substrate. By analysing droplet size, spacing, and spreading behaviour on the substrate, the printing conditions can be adjusted to achieve continuous film formation. Figure 5 shows optical microscopy observations of single deposited SRB droplets on PET.

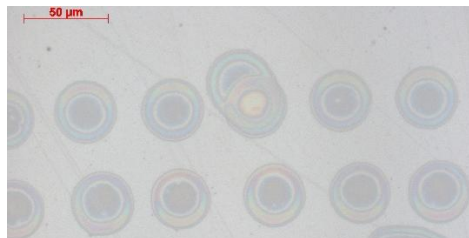


Figure 5: Optical microscope of dry single drop SRB ink on PET

During the printing process, the coffee ring effect was observed. This phenomenon occurs due to solvent evaporation, which induces a capillary flow that transports solutes toward the border of the droplet. As a result, material accumulation occurs at the droplet perimeter, leading to non-uniform fluorescence intensity across the film. For organic fluorophores such as SRB, this defect may influence fluorescence performance. The organic fluorophores like SRB are known to be

quenched when they are aggregated (aggregation-caused quenching). The accumulation of fluorophores at the perimeter of the drop could therefore locally increase aggregation and potentially reduce fluorescence intensity.

3.4 Optimization of fluorescent film performance.

For optimizing the fluorescence, two parameters are important: the concentration of fluorescent materials and the thickness of the printed film. The ink's composition selected is the one determine thanks to the experiments of 3.1. Only the concentration of fluorophores is changed.

The concentration of fluorescent materials is particularly crucial for organic fluorophores. At high concentrations, fluorophore molecules may aggregate, leading to aggregation-caused quenching (ACQ). To investigate this effect, a series of experiments were conducted using varying concentrations (as shown in Figure 6). The purpose was to identify the concentration range providing the highest fluorescence intensity while avoiding aggregation-induced quenching.

Similarly, the influence of the printed film thickness was investigated by printing multiple layers. Increasing the film thickness may enhance emission by increasing the quantity of fluorescent material present in the film. However, excessive thickness may lead to self-absorption or light scattering, which can reduce the overall fluorescence intensity. Inversely, a too thin layer may not contain sufficient fluorescent material to generate strong emission.

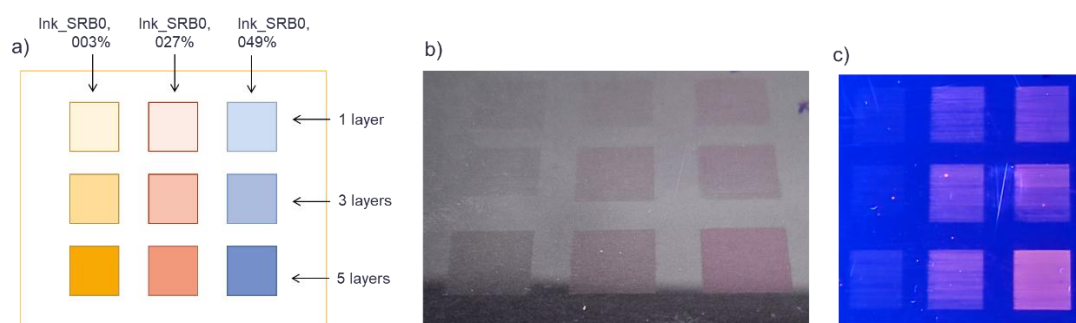


Figure 6: a) Printed patterns 1x1cm square, with increasing SRB concentration (lines) and increasing number of printed layers (columns). b) Results of the print on PET under daylight. c) Results of the print on PET under UV light (365nm).

The fluorescence properties measured in liquid formulations do not necessarily guarantee strong fluorescence after printing. The interaction between the ink and the substrate plays a crucial role in determining the final optical performance. In porous substrates such as paper, the ink penetrates into the fibre network, whereas on non-porous substrates such as PET, the fluorescent materials remain mainly at the surface. These differences strongly influence the distribution of fluorophores and consequently the emission properties of the printed films. Figure 7 shows the emission spectra of the printed films on the different substrates under excitation at 365 nm. By increasing the superposition of printed layer, the intensity of the fluorescence increases as well.

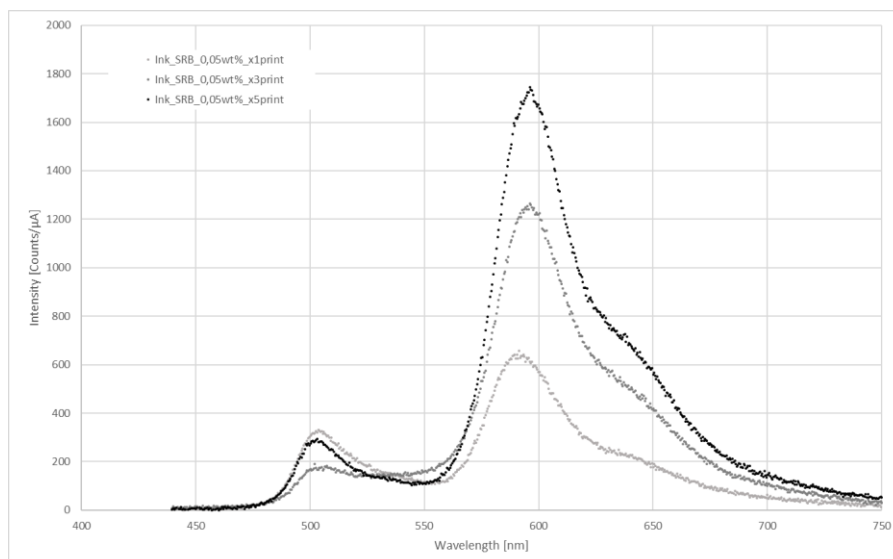


Figure 7: Emission spectra of printed SRB ink (0,05wt% of dye content) films on paper

4. Conclusions

In this work, several fluorescent inks were successfully formulated. The developed inks are water-based formulations designed to meet the physicochemical requirements for inkjet printing while maintaining strong fluorescence properties. The addition of a binder enabled good adhesion on non-porous substrates such as PET. The inks exhibited physicochemical properties compatible with inkjet printing and produced fluorescent thin films with significant emission intensity after printing. Further investigations were conducted to optimize the fluorescence performance by studying the influence of dye concentration, film thickness, and substrate properties. Current work focuses on gaining a better understanding of the interactions between ink formulation, printing process parameters and fluorescence performance. By successfully combining luminescent materials with eco-friendly ink formulations, printing techniques and fluorescence high-performance, this work paves the way for sustainable functional coatings with potential applications in security printing, sensing, and optical devices.

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