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Fabrication and Mechanical, Dielectric and Optical Properties of Cellulose Paper Embedded with SrAl₂O₄:Eu,Dy Phosphor

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ABSTRACT: The work deals with cellulose paper filled with nanocellulose and $SrAl_2O_4$:Eu,Dy oxide phosphor. It was found that both nanocellulose and oxide improve the tensile strength of the composites obtained. The samples with the oxide demonstrate a long-lasting photoluminescence (PL) under sunlight and ultra-violet (UV) illumination. Room-temperature the PL spectra reveal a wide multicomponent band spreading over all the visible spectral regions. The short-wavelength part of the band is ascribed to the cellulose-related luminescence, while the long-wavelength PL component with maxima near 540 nm corresponds to the luminescence of the $SrAl_2O_4$:Eu,Dy phosphor. The dependency of the PL intensity on oxide concentration suggests the reabsorption of cellulose emission by the oxide and *vice versa*. The study of the dielectric properties of composite papers shows the presence of dielectric relaxations at low temperatures (T ~-50°C). Similar cellulose materials to those studied can be considered as alternatives for artificial petroleum-based polymers. Low cost, eco-friendliness, biocompatibility, and the simplicity of recycling are among the main advantages of these materials. They are produced from the cellulose which is one of the most abundant renewable materials in nature. The data on the mechanical, dielectric, and optical properties indicate that the papers studied can be used in flexible lighting devices, WLEDs, coating, markers, labels, etc.

KEYWORDS: Cellulose; long lasting phosphor; paper; photoluminescence

1 Introduction

The wide application of artificial petroleum-based polymers (plastics) in modern technology results in substantial waste generation. According to the recent data of UNEP-UN Environment Programme [1], the current rate of plastic waste production in the world is nearly 400 million tons per year with a total amount of nearly 7 billion tons accumulated so far. This waste poses significant threats to both the environment and living beings. Research shows that microplastic particles are increasingly found in the human body (e.g., lungs, liver, brain) while the effect of this pollutant is not yet fully understood [2]. Unfortunately, most artificial polymers are discarded and only 10% of total plastic waste is recycled or reused nowadays. Thus, substitution of plastics with suitable eco-friendly materials is an urgent problem for modern science and technology. The cellulose and its derivatives can be used for the abovementioned purpose. Cellulose-based



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materials are biodegradable, so they can easily be composted [3]. Cellulose sources are abundant; numerous well-developed techniques of its production in various forms (global production of $\approx 1.5 \times 10^{12}$ tons per year [4]) make this material cheap and competitive in comparison to artificial polymers.

"Paper electronics" is a technology in which cellulose derivatives are used as an alternative to plastics in electronics [5–7]. Cellulose and cellulose-based composites are used as substrates when cellulose hydrogels and nanocellulose with various additives serve as inks for printing of electrical schemes by conventional and 3D printing technologies [7–9]. The current and suggested applications of paper electronics devices include sensors (pressure, temperature, humidity, biosensors) [10–13], light emitting devices [14,15], solar cells [16], batteries [17], nanogenerators [18], etc. Although the chemical and physical properties of cellulose materials satisfy some basic requirements, the improvement of flexibility/stiffness, dielectric/conductivity, capacitance, light absorbance/emission is of high importance for enhancing the performance of the above-mentioned devices. Such improvements can be done by incorporation of various additives (fillers) into the cellulose matrix.

Nanocellulose (NC) occupies a special place among the fillers, as it both improves the mechanical properties of cellulose materials and is eco-friendly. In particular, adding 2% of nanofibrillated cellulose to wood fibers improves tensile strength of the latter from 97.5 to 141 MPa [19]. This effect was ascribed to an increase in inter-fiber stress transfer when cellulose nanofibers either cover cellulose fiber or form additional porous membranes and foams in the pores of the larger scale cellulose networks. Remarkably, only a few wt. % of NC (nanofibers or nanocrystals) significantly improve mechanical and dielectric properties of cellulose paper [19,20].

Cellulose-based films offer quite high transparency in the visible spectral range (transmittance up to 90.7% was reported for cellulose nanopaper [21] and nearly 90% for regenerated cellulose film [22]). The cellulose and its derivatives luminesce under ultraviolet (UV) and visible light illumination [23–25]. However, the intensity of the luminescence of pure microcrystalline cellulose and nanocellulose is insufficient for practical application in lighting devices. In order to improve the luminescence properties a cellulose matrix can be incorporated with luminescent particles, such as carbon or semiconductor quantum dots [22,26], coordination complexes [27], or inorganic phosphors [28–30]. The rare-earth ions doped oxide phosphors attract the most attention as they offer a high luminescence intensity, long operational stability as well as low toxicity in comparison with semiconductor quantum dots or coordination complexes.

In this work, we report the effect of NC and SrAl₂O₄:Eu,Dy oxide with long-lasting luminescence on the mechanical, dielectric and optical properties of a cellulose paper. To the best of our knowledge a number of studies have explored on cellulose-based materials with SrAl₂O₄:Eu,Dy phosphor—cotton fibers [30], cellulose acetate nanofibrous films [31], regenerated cellulose fibers [32], and nanofibrillated cellulose films [33,34]. These materials were proposed for use in anti-counterfeiting purposes, luminous flexible equipment, interior decoration, night lighting, and portable logos or labels. In contrast to the abovementioned studies, in the present paper we report the simultaneous effect of cellulose and the oxide phosphor on mechanical and luminescence properties of the composite paper has been reported in this work. Additionally, this work serves as the first detailed study of the dielectric properties of the cellulose/nanocellulose/SrAl₂O₄:Eu,Dy composite paper. In our opinion, the data discussed in this work are necessary when using these composites in flexible paper electronics as substrate or active light emitting elements.

2 Materials and Methods

2.1 Sample Preparation

A bleached sulfate pulp from coniferous wood has been used as a fibrillar matrix for preparation of paper samples embedded with SrAl₂O₄:Eu,Dy luminescent oxide. This type of cellulose was chosen as one that provides the desirable parameters of tensile strength of composite paper and also allows studying their mechanical, dielectric and optical properties.

The process of sample preparation consists of the next steps. In the first stage, industrially produced sheets of wood pulp (as a source for the fibrillar matrix) were cut into pieces measuring ~50 mm × 50 mm. Then, these pieces were soaked in distilled water until they swelled with further grinding in a laboratory grinding mill (Fig. 1). The resulting suspension was characterized by the Schopper-Riegler test resulting in 55 ± 3 °SR degrees. To improve the mechanical properties of the composites, a suspension of NC from non-wood plant material was added to the cellulose mass at the consumption of 0.5 wt. % of the cellulose pulp. Nanocellulose acts as a strengthening component that binds the the fibers of the cellulose matrix. This NC was extracted from cellulose by ecologically friendly organosolv method with the application of techniques described elsewhere [35]. The paper gluing process was increased by adding a solution of alkyl ketene dimer with a consumption of 0.3 wt. % to the mass of the original wood pulp. The rationale behind the oxide filler concentration range accounted for the results of the work [33] where similar materials were studied. It was shown in [33] that 40 wt. % and more of the SrAl₂O₄:Eu,Dy filler decrease the mechanical properties of the paper. Consequently, concentration range 0–23 wt. % of the oxide powder has been taken for the research being described. The required mass of the oxide filler was weighted using an analytical balance (precision \pm 0.001 g) and added to the suspensions noted above to prepare the luminescent composite paper.



Figure 1: Scheme of preparation of cellulose-based paper samples embedded with SrAl₂O₄:Eu,Dy luminescent oxide

The obtained suspensions have been thoroughly stirred using magnetic stirring. On the next stage, the paper samples have been obtained by casting the suspension prepared using Büchner funnel and Bunsen flask in the form of disks 120 mm in diameter. The drying process was performed in a drying chamber under $T = 97^{\circ}C$ and pressure p = 93.1 kPa. The measured density of fibrillar suspension was 0.5 g/l and final density of dried paper disks was 100 ± 5 g/m². The compositions and abbreviation of all samples produced are noted in Table 1 below.

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Sample abbreviations	Cellulose pulp, g	NC, 10 ⁻³ g	Oxide , 10 ⁻² g	NC, %	Oxide, wt. %
PO	1	0	0	0	0
PN	1	5	0	0.50	0
PN1	1	5	1	0.49	0.99
PN2	1	5	2	0.49	1.95
PN3	1	5	3	0.48	2.90
PN5	1	5	5	0.47	4.74
PN10	1	5	10	0.45	9.05
PN30	1	5	30	0.38	22.99

Table 1: The content of the samples in grams (g) and in %, and their abbreviations

2.2 Characterization Techniques

Optical images of the samples have been obtained using XS-3330 MICROmed microscope equipped with a digital camera.

The Tescan Vega 3 electron microscope was used for study of the samples morphology. Accelerating voltage was from 1 to 25 kV and maximal resolution riches 3 nm.

The physico-mechanical parameters, namely tensile strength of the composite papers, have been studied for the case of 15 mm wide strips that were cut from the abovementioned paper discs. These studies have been performed on RBM-30-2M vertical bursting machine. The values of the tensile strength (*TS* in N/m²) were calculated by the formula $TS = P/(t \cdot a)$, where *t*—sample thickness, in m; *a*—sample width, in m.

The dielectric properties of the composite paper, namely real and imaginary parts of complex dielectric permittivity, have been studied in -190° C -90° C temperature range and in the 5 to 50 kHz frequency range. An automated installation based on a P5083 AC bridge and a thermostated four-electrode cell has been used for the measurements of capacity and dielectric loss tangent [36,37].

The photoluminescence properties have been studied using a modernized DFS-12 spectrometer (equipped with FEU-100 photomultiplier tube) and a Horiba/Jobin-Yvon Fluorolog-3 spectrofluorometer. The ultraviolet (UV) diodes ($\lambda_{rad/ex} = 265$, 365 and 395 nm) as well as powerful xenon arc lamps were used as sources of the PL excitation.

3 Results and Discussion

A general view of the composite paper strips under daylight illumination can be found in Fig. 2a. The surface of the samples appears in a yellowish hue that can be ascribed to absorption of the violet/blue light by cellulose and the oxide accompanied by some luminescence of these components. After illumination by daylight or UV light, composite paper samples exhibit an intensive, long-lasting luminescence) (Fig. 2b).

3.1 Optical and Scanning Electron Microscopy

Fig. 3 shows that the samples studied consist of intertwined cellulose fibrils (thickness ~10 μ) that form a porous network. The structure of the samples suggests that incorporated SrAl₂O₄:Eu,Dy oxide nano/microparticles are located both on the surfaces of fibrils and between them, in pores. The samples without oxide, PN0 and PN1, are characterized by weak blue luminescence after illumination with UV light at $\lambda_{ex} = 265$ nm (Fig. 3b,c) due to the own luminescence of the cellulose host. The larger oxide particles and their agglomerates can be easily seen in the optical microscope images of the samples, PN1-PN30, owing to the bright green luminescence of these particles (Fig. 3c). It should be noted that the visible sizes of particles

and agglomerates in Fig. 3c exceeds their real sizes due to the luminescent radiation scattering on the samples' inhomogeneities. The particles of the oxide filler are observed in SEM images as bright light dots with a size of 0.1 to 10 μ including their clusters (Fig. 3d,e).



Figure 2: Photos of the strip samples taken under daylight illumination (a) and after 10 min of daylight exposure taken in a dark room (b)



Figure 3: Optical, under UV light illumination (left; the scale bar is 100 μ), and SEM images (right) of P0 (a), PN3 (b), PN30 (c), and PN10 (d, e) samples

3.2 Tensile Strength

A dependence of tensile strength on sample compositions is shown in Fig. 4. It was found that 0.5 wt. % nanocellulose improves the tensile strength of cellulose paper on about 2%. The improvement of mechanical properties (namely tensile strength and index) of paper by nanocellulose fibers or nanocrystals has been widely described in the literature [38–40].



Figure 4: Dependence of tensile strength on composition of the samples

The simultaneous effect of nanocellulose and oxide fillers provides increase of the tensile strength by up to 16%. The improvement of tensile strength by oxide addition has been previously reported in [41,42]. In instance, the cellulose nanocrystals and ZnO significantly improved the mechanical properties of poly(vinyl alcohol)/chitosan blend films [41]. Mechanical properties of the modified cellulose acetate membrane had been successfully improved by ZrO₂ layers [42]. At the same time particles SiO₂@SrAl₂O₄:Eu,Dy degraded mechanical properties of polymer films based on TEMPO-oxidized nanofibrillated cellulose [33]. In case of regenerated cellulose, incorporation up to 8 wt. % of SrAl₂O₄:Eu,Dy particles into fibers has almost no effect on the rigidity of the material, while further increasing of the oxide content to 12 wt. % leads to significant decrease (almost twice) of tensile strength [32]. It was supposed that inorganic particles disturb oriented fiber structure, although they increase the number of entanglement points between cellulose fibers [32]. These processes have an opposite effect on the mechanical properties of polymer composites.

A tensile strength dependence on the amount of the filler (Fig. 4) is nonlinear and reveals a tendency to saturation despite the increasing amount of solid oxide particles. Consequently, the results confirmed an assumption that incorporated particles "reduced the number and strength of the hydrogen bonds" which provides the binding cellulose fibrils [33]. At the same time, it should be pointed out that in our research tensile strength is increased when the content of the oxide reaches 23 wt. %, while as it is reported in the work [32] an incorporation of more than 8 mas. % of SrAl₂O₄:Eu,Dy is accompanied by the tensile strength decrease. Thus, our results can be explained by an impact of the nanocellulose introduced to the composition of the studied papers.

3.3 Dielectric Properties

The temperature dependences of the real $\varepsilon'(T)$ and imaginary $\varepsilon''(T)$ parts of complex dielectric permittivity at four frequencies for PN and PN30 samples are shown in Fig. 5. The curves of $\varepsilon'(T)$ for the samples contain band at low temperature (maxima near -20° C). This band moves toward higher temperatures when frequency increases. This shift indicates the relaxation nature of this band.

Curves of imaginary part $\varepsilon''(T)$ of complex dielectric permittivity for the samples also reveal a band in $-190^{\circ}\text{C}-0^{\circ}\text{C}$ temperature range. As in case of $\varepsilon'(T)$ dependence, the shift of band maxima toward higher temperature with increasing of frequency additionally indicates the relaxation nature of this band.

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Figure 5: Temperature dependencies of real (a, c) and imaginary (b, d) parts of complex dielectric permittivity for PN (a, b) and PN30 (c, d) samples

In order to analyze the effect of oxide content on the abovementioned relaxation process the dependencies of $\varepsilon''(T)$ for the studied samples are shown at the same frequency (Fig. 6). It worth noting, that SrAl₂O₄:Eu,Dy content increase leads to the relaxation region shifting to higher temperatures and the intensity of relaxations increasing.



Figure 6: Temperature dependencies of imaginary part of complex dielectric permittivity obtained for frequency f = 10 kHz

The parameters of the abovementioned relaxation processes can be estimated from the model system of the identical relaxers each of those have two energetically non-equivalent positions separated by potential barrier [43–45]. Within a framework of this model the dependencies of real and imaginary parts of complex dielectric permittivity can be described by Eqs. (1) and (2), as it was proposed earlier [43]:

$$\varepsilon'(\omega, T) = \varepsilon_{\infty} + \frac{\Delta \varepsilon'(T)}{1 + \omega^2 \tau^2(T)}$$
(1)

$$\varepsilon''(\omega,T) = \frac{\Delta\varepsilon'(T)\omega\tau(T)}{1+\omega^2\tau^2(T)},\tag{2}$$

where ε_{∞} —dielectric permittivity at high frequencies (related with elastic polarization mechanisms), $\Delta \varepsilon'(T)$ —dependence of dielectric permittivity increment on temperature, ω —angular frequency, τ —relaxation time.

Relaxation time can be described by Eq. (3):

$$\tau = \frac{2\pi}{\omega_0} \frac{\exp(\frac{U}{kT})}{1 + \exp(\frac{-V}{kT})}$$
(3)

where ω_0 —frequency of the relaxer vibrations at equilibrium position, *U*—activation energy, *V*–difference in energy for relaxer in 1 and 2 positions, *k*—Boltzmann constant.

The temperature dependence of dielectric permittivity increment can be described by Eq. (4):

$$\Delta \varepsilon'(T) = \varepsilon_0 - \varepsilon_\infty = \frac{N\mu^2}{3k\varepsilon_0 T} \cdot \frac{\exp(\frac{-V}{kT})}{\left[1 + \exp(\frac{-V}{kT})\right]^2}$$
(4)

where *N* is relaxers concentration, μ —average difference of dipole moment of relaxers in positions 1 and 2, ϵ_0 —electric constant.

Approximation of temperature dependencies of $\varepsilon'(T)$ by Eq. (4) at the high temperature part of relaxation band (-15°C-0°C region at Fig. 5) where $\omega\tau \ll 1$ allowed us to estimate the $N\mu^2$ and V parameters (obtained values are collected in Table 2). It can be seen from the Table 2, that energy difference V for relaxer in two states as well as concentration of relaxers increase with oxide content increasing. Both the $N\mu^2$ and V parameters reach their maximum values at ~5 wt. % of oxide in composites, as it follows from $N\mu^2$ and V dependencies on concentration (Fig. 7).

Table 2: Energetical characteristics of relaxation processes

Sample	$\mathbf{N}\boldsymbol{\mu}^2$, $\mathbf{C}^2\mathbf{m}^{-1}$	$V, kJ \cdot mol^{-1}$	$\Delta S/k$	U, kJ·mol ⁻¹
PN	$2.92970 \cdot 10^{-31}$	6.3	55	30
PN1	$3.08745 \cdot 10^{-31}$	6.8	57	36
PN2	$3.20223 \cdot 10^{-31}$	6.9	58	38
PN3	$4.49074 \cdot 10^{-31}$	7.5	58	39
PN5	$4.82798 \cdot 10^{-31}$	7.8	58	40
PN10	$4.72026 \cdot 10^{-31}$	8.0	61	45
PN30	$5.32423 \cdot 10^{-31}$	7.5	61	47



Figure 7: The dependencies of relaxer concentration (black squares) and energy difference V (red circles) on SrAl₂O₄:Eu,Dy concentration

In case of U >> V and when the entropy of activation is taken into account for relaxation time, the value of the last one can be calculated by Eq. (5):

$$\tau = \tau_0 \exp\left(\frac{U - T\Delta S}{kT}\right),\tag{5}$$

where $\tau_0 = 10^{-12}$ c, ΔS —entropy of activation [40]. The condition of maximum for relaxation process at $\varepsilon''(T, f)$ dependence can be written as $\omega \tau = 1$. Consequently, some relaxer parameters can be obtained by approximation of $\ln f(1/T_r)$ dependencies with $\ln f = -\ln 2\pi\tau_0 + \Delta S/k + U/kT$ formula (see. Fig. 8). The estimated entropy of activation $\Delta S/k$ and activation energy U are collected in Table 2. It is seen, that both abovementioned parameters rise with $\mathrm{SrAl_2O_4}$:Eu,Dy concentration increasing. The increase of the activation energy indicates that $\mathrm{SrAl_2O_4}$:Eu,Dy particles prevent some transformations of cellulose-building units.

It is known that cellulose consists of anhydroglucose units carrying two hydroxyl groups (-OH) and one methylol group ($-CH_2-OH$). According to Einfeldt et al. [46], there are three types of dipolar and movable sites—the pyranose ring (movable by orientational motions around the glucosidic bonds) and two side groups with rotation mobility around C-O linkage. The rotations of the methylol side-group are responsible for low-temperature relaxations [45–47]. Thus, we suppose that SrAl₂O₄:Eu,Dy particles prevent the reorientations of the ($-CH_2-OH$) methylol side-groups in the cellulose molecules.

3.4 Photoluminescence Studies

As was shown earlier [33], the composite paper with SrAl₂O₄:Eu,Dy filler reveals intensive green luminescence even under daylight illumination. The photoluminescence (PL) spectra of the studied samples are shown in Fig. 9a. The PL spectrum of initial paper P0 consists of an asymmetric complex band with an overall maximum in the blue spectral region near 450 nm. This band has been numerously reported for various forms of cellulose but the nature of this luminescence is still under discussion [23–25,48,49]. It is known that nanocellulose (nanocrystal and nanofibrils) reveals blue luminescence [50], thus this emission

should be imposed with emission from cellulose fibers. In fact, a nanocellulose filler increases the intensity of short-wavelength part of emission of the composite paper but has almost no effect at the region of $\lambda > 500$ nm.



Figure 8: The dependences of logarithm of frequency on inverse temperature $(1/T_r)$ of relaxation band maxima at $\varepsilon''(T)$ curves for composite samples with different SrAl₂O₄:Eu,Dy concentration



Figure 9: (a) The PL spectra of the samples measured under excitation at $\lambda_{ex} = 365$ nm, T = 290 K; (b) the dependencies of integrated luminescence intensities on oxide concentrations

Significant changes in the luminescence of the papers studied have been observed for the samples embedded with oxide filler. The luminescence of these samples is long-lasting; thus, it can be registered for many hours after the photoexcitation has been turned off. Depending on cellulose source, the typical lifetime constants for cellulose-related luminescence range only from a few to several hundreds of milliseconds [24]. Thus, the green luminescence of the studied composite samples can be ascribed solely to SrAl₂O₄: Eu,Dy oxide emission. This PL is related to electron and hole trapping and releasing. Several possible

trapping/releasing mechanisms have been discussed so far: 1) hole released by Eu^{2+} ion to the valence band (VB) \rightarrow hole trapped by Dy^{3+} ion \rightarrow hole released by Dy^{4+} ion \rightarrow phosphorescence as a result of hole recombination with excited Eu^{1+} ion [51]; 2) electron released by Eu^{2+} ion to the conduction band (CB) \rightarrow electron trapped by Dy^{3+} ion \rightarrow thermal release of the electron from Dy^{2+} ion \rightarrow luminescence as a result of hole recombination with excited Eu^{3+} ion [52]; 3) luminescence of $SrAl_2O_4$:Eu,Dy originates from the photo-oxidation of Eu^{2+} cations under UV irradiation, and electron traps are oxygen vacancies located nearby photo-generated Eu^{3+} ions [53]; 4) this is the model that included the models #2 and #3 [54], etc. However, additional experimental data requires for deeper understanding of the energy trapping/releasing mechanisms of the $Sr(Ca)Al_2O_4$:Eu,Dy luminescence [55–58].

A wide band of the green PL with a maximum near 540 nm dominates in the PL spectra regardless of the SrAl₂O₄:Eu,Dy concentration. It is observed that the blue emission from cellulose/nanocellulose is considerably suppressed at 2 wt. % of oxide. This effect is depicted in the dependencies of the integrated PL intensities on oxide concentration (Fig. 9b). As it follows from the data, the dependency of green PL on SrAl₂O₄:Eu,Dy content has two linear regions: 1) when X < 3 wt. % (this part can be approximated by line I (X) = 6X); 2) when 3 < X < 30 wt. %. This part can be approximated by the line I (X) = 9 + 3.2X. In our opinion, the radical change in the increase index results from a sharp increase in the rate of excitation energy transfer from the matrix to the Eu²⁺ luminescence centers in the oxide filler when the distance between oxide particles is shortening.

The PL excitation spectra have been measured at emission registration in the blue and green PL bands (Fig. 10). The blue luminescence is effectively excited in a wide band with maxima near 370 nm (Fig. 10a). As for the samples with oxide, the maximum of the excitation band is shifted toward longer wavelengths due to overlapping of cellulose- and oxide-related emission (see Fig. 9a). Excitation spectra of the green emission substantially differ from those for the blue PL. In particular, there are at least three PL excitation bands with maxima near 320, 370 and 420 nm. It was reported earlier that PL excitation spectrum of SrAl₂O₄: Eu,Dy phosphor consists of a wide single band in 270–450 nm region with maximum near 350 nm [59]. This band has been ascribed to the inter-configurational transitions from the ground $4f^7$ state to the excited $4f^65d^1$ state in Eu²⁺ ions. Interestingly, the PL excitation band with maxima near 420 nm was observed earlier for TEMPO-oxidized nanofibrillated cellulose filled with SiO₂@SrAl₂O₄: Eu,Dy particles [33]. In our opinion, the bands at 320 and 420 nm are more likely to be connected with cellulose-related light absorption centers. Detailed studies of the PL characteristics, especially of the PL decay and thermo-stimulated glow curves should be performed in the future to assign specific PL excitation bands to cellulose- or oxide luminescence centers.

From the viewpoint of the light emitting devices application, it is important to know the color characteristics of the composite papers, which are chromaticity (x, y) and (u', v') coordinates according to CIE 1931 and CIE 1976 standards; correlated color temperature (CCT) and color purity (CP). The data have been estimated from the PL spectra measured at PL excitation at 365 nm (Fig. 9) and the data were collected in Table 3. The CCT values were calculated by the well-known empirical McCamy formula [60]. The *CP* values have been calculated by the method described in [61]. The obtained CCT values correspond to the "cold" radiation (T > 4500 K). Low color purity values are associated with the spread of the PL band over the entire visible light range. This result indicates that the studied materials could not be applicable where high color rendering is necessary (e.g., displays), but they can be used as components of low-power lighting devices. The observed non-monotonous behavior of CCT and CP regarding oxide content is more likely to be related with the reabsorption of cellulose emission by oxide and *vice versa*.



Figure 10: The PL excitation spectra measured at λ_{em} = 450 (a) and 550 nm (b); T = 290 K

Sample	X	у	u'	v'	ССТ, К	СР, %
PO	0.2768	0.3238	0.1748	0.4603	8842	10.2
PN	0.2559	0.2826	0.1741	0.4326	13,275	22.2
PN1	0.3037	0.3972	0.1697	0.4993	6498	17.3
PN2	0.3348	0.5048	0.1596	0.5416	5462	59.5
PN3	0.3235	0.4557	0.1654	0.5244	5739	40.8
PN5	0.3268	0.4704	0.1636	0.5298	5647	42.8
PN10	0.3344	0.5353	0.1528	0.5503	5473	73.3
PN30	0.3493	0.5477	0.1574	0.5555	5202	78.7

Table 3: Color characteristics of composite papers luminescence

4 Conclusions

The composite paper samples consisting of a cellulose paper as a matrix, nanocellulose and an oxide SrAl₂O₄:Eu,Dy long-lasting phosphor as fillers have been produced and studied. The composites incorporated with the luminescent oxide consist of intertwined cellulose fibrils with oxide particles between them.

The combined effect of nanocellulose and oxide phosphor on mechanical, luminescent, and dielectric properties of the produced papers has been studied for the first time.

Temperature dependencies of dielectric permittivity indicate the presence of dielectric relaxations. The analysis of fitting parameters and their relation with the content of oxide particles suggests the presence of interaction between cellulose matrix and filler.

The prepared composite papers showed both blue and green luminescence bands under UV and visible light exposure. The former is attributed to the cellulose fibrils, while the latter is due to the radiation transition in Eu^{2+} ions. The green luminescence intensity significantly increases, while the intensity of the blue band decreases with increasing oxide filler content.

The effect of the fillers ensured an increase in the tensile strength of the paper by up to 16% and its luminescence intensity by a factor of 8.3.

The properties of these composite make well-suited for the use as a portable logo and label substrate, or as an active light-transforming material of flexible paper electronics.

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Availability of Data and Materials: The data that support the findings of this study are available from the corresponding author upon reasonable request.

Ethics Approval: Not applicable.

Conflicts of Interest: The authors declare no conflicts of interest to report regarding the present study.

Nomenclature

- CB Conduction band
- NC Nanocellulose
- PL Photoluminescence
- UV Ultraviolet
- VB Valence band

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