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## **The possibilities of using a mixture of PDMS and phosphor in a wide range of industry applications**

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A mixture of polydimethylsiloxane (PDMS) doped with phosphor particles can be found across diverse industries having different applications. This mixture plays a particularly important role in the field of lighting, white light-emitting diodes (LED's), flexible display devices, anti-counterfeiting (AC) solutions, luminescence thermometers and many types of sensors. The field of mechanoluminescence and biomedical are booming and there is also potential for visible light communication (VLC). In this comprehensive review, the basic characteristics of PDMS and a list of selected phosphors suitable for creating a mixture of PDMS and phosphor are presented. The summary and a detailed overview of the implemented applications of this perspective mixture over the last decade is presented as well.

**Keywords:** PDMS; phosphor; white LED's; display; flexible light devices; anti-counterfeiting; luminescence thermometry; visible light communication; mechanoluminescence

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## Introduction

The combination of phosphor and polydimethylsiloxane (PDMS) is constantly finding new possibilities for its application in many areas of human activity and industrial production. This is due to the fact that the mixture of phosphorus and PDMS has many advantageous properties. PDMS is an elastomeric polymer first synthesised in the 1950s by Wacker Chemies and the first main use was the encapsulation of electronic components, acting as a dielectric insulator, protecting the components from mechanical shocks and other environmental factors within a large temperature range. Due to its high elasticity and stability of its properties, PDMS is widelly used as me-chanical sensor<sup>[1](#page-16-0),[2](#page-16-1)</sup>.

In 1998, a patent describing the use of PDMS mixed with phosphorus materials for the optical encapsulation of LED's was filled. In this patent, PDMS is referred as the best alternative to the conventional alternative at the time, polymethylmethacrylate (PMMA), polycarbonate, optical nylon, transfer molded and/or cast epoxy have been used for encapsulation. However, these materials suffered degradation of the optical characteristics over

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time, contrary to PDMS<sup>[3](#page-16-2)</sup>. Epoxy-based resins tend to yellow over time due to high temperature exposure and ab-sorption of ultraviolet (UV)/blue light<sup>[4](#page-16-3)</sup>. Depending on the choice of phosphor, it is possible to obtain the desired range of excitation and converted wavelengths. A downconversion or upconversion phosphor can be more suitable for a particular application. In the case of upconversion, the excitation wavelength is longer than the converted one, and the opposite is true for downconversion. For combination with phosphor, it is preferable to use clear polydimethylsiloxane, e.g. Sylgard 184 type. Thanks to the clear color, light losses of both excitation and converted light are minimized. PDMS is a chemically stable substance with remarkable resistance to thermal and oxidative degradation and is also resistant to UV and radiation. PDMS has the ability to withstand long-term temperatures in the range of approximately −50 °C to 200 °C and can withstand even higher temperatures for a short time. Due to its bonds, it has high adhesion and is strongly hydrophobic. PDMS elastic modulus can be between 1.32 and 2.97 MPa and tensile strength from 3.51 to 5.13 MPa, depending on the manufacturing method. It also has excellent electrical insulation properties, is resistant to various chemicals, does not cause corrosion of other materials and has gas permeability. PDMS devices usually have repeatability while being easy to manufacture and cost-effective, as with replica moulding. PDMS is biocompatible, with reported uses in microfluids, biomodels, organ-on-a-chip platforms, blood analogues and membranes for filtering<sup>[2](#page-16-1),[5,](#page-16-4)[6](#page-16-5)</sup>. The connection of phosphor and PDMS therefore leads to considerable resistance of this mixture to the chemical and physical effects of the external environment. At the same time, it maintains flexibility in setting the wavelength ranges of excitation and converted light. In addition, it is a flexible and self-supporting mixture that exhibits sufficient mechanical resistance in many areas of use<sup> $7−14$  $7−14$  $7−14$ </sup>. Among the most important are the area of white-LED's and solid-state lighting, for instance[15](#page-16-8)−[18](#page-17-0). Other important areas include anti-counterfeiting (AC) solutions<sup>[19](#page-17-1)−[22](#page-17-2)</sup>, temperature sen-sors or thermoluminescence<sup>[23−](#page-17-3)[26](#page-17-4)</sup> and mechanolumines-cence including also biomedical applications<sup>27–[30](#page-17-6)</sup>. In particular, mechanoluminescence has recently experienced a great boom. The mixture of PDMS and phosphor finds application in many sensor and detection applications<sup>[31](#page-17-7)–[34](#page-17-8)</sup>, [Fig. 1](#page-2-0) summarizes some of the different applications, discussed in more detail in the next sections, where the mixture of PDMS and phosphor materials can be found.

The areas where the blend is most matured are solidstate lighting, specially white-LED's and AC solutions. In contrast, the application in sensing, where the use of the blend has a strong perspective in different areas, has yet to be totally explored. Some of the explored areas in sensing include: pH sensor, bio-sensor (mechanical, using the mechanoluminescence capabilities and thermal, using the thermoluminescence capabilities), metal and/or gaseous particles detector, X-ray and UV detection, etc. And there are certainly opportunities and challenges in the field of visible light communications (VLC)[35−](#page-17-9)[37](#page-17-10). However, many phosphors are used in several areas, and therefore in some cases it is not easy to determine the most important area of application of a giv-en phosphor, for example<sup>[38−](#page-17-11)[40](#page-17-12)</sup>, which highlights the versatility of PDMS and phosphor blends.

It is known that a phosphor consists of a host crystal (host lattice) and a luminescence center (activator), so these two components influence the resulting properties of the phosphor. In [Table 1](#page-3-0) some of the typical host matrix, typical activator ions and respective emissions are showcased.

Different properties of phosphors with  $Ce^{3+}$  activator

<span id="page-2-0"></span>

**Fig. 1 | Some applications of PDMS:phosphor, highlighting the heterogeneity of applications where the blend has potential uses.**

<span id="page-3-0"></span>

**Table 1 | Phosphor Blends of PDMS:host matrix:activator ion and respective wavelengths of emission.**

Introducing  $Gd^{3+}$  ions, which are larger and thus have a increased crystal field than  $Y^{3+}$  causes a red shift of the  $Ce^{3+}$  emission in YAG :  $Ce^{3+}$ . A substitution of  $Al^{3+}$ ions for the larger  $Gd^{3+}$  shifts the spectrum in the other direction, due to the weaker crystal field of the Ce<sup>3+ [51](#page-18-1)[,52](#page-18-2)</sup>. located in different types of host crystals (Fluorides, Oxy-halides, Aluminates, Silicates and others) are known<sup>[50](#page-18-0)</sup>.

## Typical manufacturing process of PDMS and phosphor mixture

The process of preparing the PDMS phosphor mixture has several stages. Due to its suitable physical and chemical properties, PDMS type Sylgard 184 (Dow company) is very often used. There are many types of PDMS, but Sylgard 184 type PDMS is used very often in practice. It will therefore be presented a possible recommended procedure manufacturing process of PDMS and phosphor mixture for Sylgard 184. For other types of PDMS, the procedure can be similar, however, there can be some specific differences. Sylgard 184 is a two-component silicone elastomer that consists of a "base" and a "curing agent" with a recommended mixing ratio of 10∶1 (base∶curing agent) by weight. After mixing these two components, the mixture is usually stirred briefly and intensively.

As a result of mixing, a large number of air bubbles form in the mixture, which must be removed. To remove the bubbles, either a vacuum chamber is used, or they are allowed to leave on their own. To improve and speed up the process of spontaneous bubble removal, this mixture can be temporarily placed in a refrigerator (e.g. at a temperature of around 5 degrees Celsius). Then comes the phase of mixing PDMS and phosphor in a precisely selected mass ratio, when the required amount of PDMS and powdered phosphor is weighed on precise scales and mixed together, while this mixture is placed in a suitable container, e.g. into a test tube.

Next, the process of homogenization of the PDMS phosphor mixture occurs. This process can be implemented with the help of an ultrasonic bath, or a mechanical shaker, or a combination of both of these options. However, longer-term use of the ultrasonic bath can lead to more intense heating of the PDMS phosphor mixture, which then leads to faster unwanted thermal curing of the mixture. To delay this thermal curing, it is advisable to place the test tube with the PDMS phosphor mixture and the shaker in a cooling box (e.g. with a temperature of around 5 degrees Celsius).

It is advisable that this process of mechanical shaking of the PDMS phosphor mixture takes place for at least 2–3 hours. However, with the use of a cooling box, it is then possible to extend the shaking time of the PDMS phosphor mixture to 10 hours. It seems appropriate to place the test tube with the weighed PDMS phosphor mixture in a shaker that combines the possibility of rotational and vibrational movement, which can be, for example, a rotary shaker of the Multi Bio RS-24 type (Biosan company).

After the end of the phase of homogenization of the PDMS phosphor mixture, it is necessary to place the PDMS phosphor mixture in the desired place. Thin layers of PDMS phosphor mixtures are often created on suitable surfaces (e.g. microscope slides, wafers, etc.) using spin coating or dip coating techniques. Sometimes the possibility of pouring PDMS phosphor mixtures into a suitably shaped container is also used.

Then comes the final phase, which is the thermal curing of the PDMS phosphor mixture. Surfaces with an applied layer of PDMS phosphor or containers into which this mixture was poured are placed in an oven, where they are thermally cured. The specified thermal curing time is dependent on temperature and may also depend

on the volume of the PDMS phosphor mixture, on the mass ratio of PDMS : phosphor, on the type of phosphor used, etc. The following values are given in [Table 2](#page-4-0) for pure PDMS type Sylgard 184 (without phosphor). In addition to the above, each researcher working with a mixture of PDMS phosphors can have their own specific production processes of combining PDMS with phosphors, as well as customized equipment such as their own home-made shaker etc.

<span id="page-4-0"></span>

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phor YAG:Ce<sup>3+</sup> for white-LED and solid-state lighting YAG:Ce<sup>3+</sup>. Using the spin coating technique, it is possi-184: YAG: $Ce^{3+}$ ) applied to the central part of the micromixture PDMS and YAG: $Ce^{3+}$ . The price of the PDMS phosphor mixture depends on the price of its components and on the weight ratio of PDMS and phosphor. In the case of the most used PDMS type Sylgard 184, the price for 1 kg of Sylgard 184 can be around 160 euros. The price of the frequently used phoscan be, for example, around 9 euros per gram of ble to apply a sufficient layer on the central part of the standard microscope slide with an amount of 0.2 g of the PDMS phosphor mixture. In the case of a considerably large weight ratio of *PDMS*∶*phosphor* 2∶1, the total price for 0.2 g of the *PDMS*∶*phosphor* mixture (Sylgard scope slide can be approx. 0.62 euros (a[pprox. 0.](#page-4-1)02 euros for PDMS and 0.6 euros for phosphor). [Figure 2](#page-4-1) displays a photograph from a microscope slide containing the

<span id="page-4-1"></span>

**Fig. 2 | Photograph of a microscope slide containing emitting samples with PDMS and YAG:Ce3+ (main emission peak at 555 nm) on the left side and the samples on the right side contain PDMS, YAG:Ce3+ and CaS:Eu2+ (main emission peak at 650 nm), using as excitation source blue light**. Figure reproduced from ref[.](#page-17-10)[37](#page-17-10), under a Creative Commons Attribution International License.

However, in the case of using another application technique on a microscope slide, e.g. the dip coating technique, the total price of the mixture PDMS phosphor applied to a microscope slide will probably be much higher. This will be caused by the need to mix a large volume of the PDMS phosphor mixture into the container into which the microscope slide will be immersed, and logically most of the PDMS phosphor mixture will remain unused. So the use of the Dip coating technique for applying the PDMS phosphor layer can be quite cost-inefficient.

On the other hand, the application of the PDMS phosphor layers using the spin coating technique also produces waste. This is created by the excess of the PDMS phosphor mixture being transposed onto the inner walls of the spin coater thanks to the forces acting during the rotation of the layer. Even so, the application of layers of the PDMS phosphor mixture using the spin coating technique is much more cost-effective in contrast to the dip coating technique.

Producing very small volumes of PDMS phosphor mixture samples can also be generally cost-inefficient, as it is practically difficult to homogeneously mix a very small volume of PDMS phosphor mixture and at the same time transfer it to the requested location. E.g. larger volumes, on the order of milliliters, are better mixed in the test tube, however, part of this volume will remain inefficiently used due to adhesion to the walls of the test tube.

In section *Applications - PDMS and phosphor* some of the most reported applications such as, white-LED's and solid-state lighting, anti-counterfeiting solutions, temperature sensors, thermoluminescence, mechanoluminiscense and bio-medical are reviewed, with some of its mechanisms and manufacturing methods being explored. In section *Discussion and future perspectives* some of the advantages and disadvantages of using these mixtures are reviewed, as well as future developments.

## Applications - PDMS and phosphor

four main areas of application of the PDMS : phosphor *PDMS* : *phosphor* mixture, which will be mentioned in a The combination of PDMS and phosphorus creates a mixture that finds application in many industries. The blend were mentioned in the introduction and will be detailed in the following subsections. In addition, there are many other possibilities of using the separate subsection.

justable *PDMS* : *phosphor* weight ratio. PDMS serves as a Individual applications are determined depending on the specific type of phosphorus. When using powdered phosphorus, it is common practice to create a homogeneous mixture of PDMS and phosphorus with an adbinder, mechanical protection and is important from the point of view of guiding the excitation radiation to the luminescence centers. For flexible bio-sensors its biocompatibility and hyperelasticity are some highlighted features. In the case of specific applications, such as gas detection, its permeability allows to better sense these environments.

#### White-LED's and display technology

*PDMS* : *phosphor* mixture is the area of white-LED's and The most important area for the use of the solid-state lighting. These are often phosphors excited by narrowband UV radiation or blue light present in the optical packaging of the chip. In the case of phosphorescent white-LED's, the weight ratio of the mixture is carefully engineered in a way that part of the excitation energy (UV or blue) is converted into a long-wave broadband component and part maintains the short-wave component. The combination of these components causes the emission of white light. Usually, this coating can be either applied mixed in the optical packaging silicone (as in typical commercial white-LED's) (see Fig.  $3(a)$ ), on the surface of the LED chip (see Fig.  $3(b)$ ) or on the surface of the optical packaging (see Fig.  $3(c)$ ). Using the second method reduces backscatering of blue light from the phosphor to the chip, causing reabsorption and reducing the overall luminescence efficiency. However, in commercial applications, the costs due to extra processing make the first a more viable commercial

application<sup>[54](#page-18-5)</sup>.

phor LEDs are YAG:Ce<sup>3+</sup> (yellow emitter activated  $BaMgAl_{10}O_{17}: Eu^{2+}/Ba_2SiO_4: Eu^{2+}/CaAlSiN_3: Eu^{2+56}.$ YAG:Ce<sup>3+</sup> is a commercial solution widely used for (YAG : Ce;  $Y_3Al_5O_{12}$ : Ce<sup>3+</sup>; YAG phosphor). The com-The most common phosphor blends for white phosmainly by 440–480 nm light, with an absorption peak at 460 nm) or a combination of blue/green/red phosphors (activated with 360–410 nm light), such as white-LED's and solid-state lighting. Some authors use different variants of the designation of this phosphor bination of blue/green/red phosphors can offer superior color quality in exchange for a more complex design of the coating and less efficient power source.

Many issues of phosphors in these applications are temperature quenching and degradation. Several protective measures can be taken to reduce these issues, such as, forming protective surface layers against moisture and high temperature in fluoride and sulphide based phosphors; surface coating in silicate and oxide based phosphors increases its Photoluminescence (PL) efficiencies; surface passivation treatment in nitrite and oxynitride based phosphors, reducing thermal degradation of PL intensity<sup>[57](#page-18-7)</sup>.

ble light. The combination of *PDMS* : *phosphor* allows to To note that this technique was already employed in fluorescent lamps, with the latter using as excitement source the UV produced by the eletric discharge in gas enclosed into vacuum tubes. The main criterion to apply a phosphor into an LED for white light is that they must show strong absorption in UV/near-ultraviolet (NUV) while having an efficient emission in the region of visieasily achieve a white tone using a single LED, contrasting with the red-green-blue (RGB) solution, which

<span id="page-5-0"></span>

**Fig. 3 | Schematic diagram of LED packaging.** (**a**) Proximate phosphor configuration, where the phosphor is homogeneously distributed across the optical packaging host. (**b**) Proximate conformal phosphor configuration, with the phosphor deposited on the LED surface. (**c**) Remote phosphor configuration, with the layer of the phosphor on the surface of the host matrix. Figure reproduced with permission from ref.[55](#page-18-8), Sage Publications.

requires 3 LEDs (working with different voltages) in the same optical encapsulation. The fore mentioned further reduces packaging costs, size and consumption.

*PDMS* : *phosphor* encapsulation unlocked illumination In ref.[58](#page-18-9) a comparison study using both technologies concluded the luminous efficacy of a phosphor-white device typically equals more than twice the efficacy of the corresponding RGB combination. The blue LED with worldwide with a reduced energy consumption, luminous efficiency, durability and more eco-friendly com-pared with previous illuminating technologies<sup>[59](#page-18-10)</sup>.

vices. The main importance of the PDMS : phosphor lay-The solid-state lighting area also includes display deer in the case of display devices lies in flexible luminescent films and backlight function. The first exploits the flexibility and stretchability of PDMS. The latter exploits the variation of the light emitted by the phosphor, controlling the intensity of the illuminating LED. Despite being a matured technology with good quantum yield while maintaining high thermal and chemical stability, the size of the phosphor particles is in the micron level, increase difficulties in integrating these coatings with micro-LED pixels from displays, affecting color conversion efficiency and uniformity of the devices<sup>60-[66](#page-18-12)</sup>.

A green (545 nm) phosphor, BaLa $_2$ ZnO $_5$  : Tb $^{3+}$ , acti-*PDMS* : *phosphor* combination for flexible displays use, vated with 255 nm light, color purity of 91.03%, quantum efficiency of 49.72% and decay lifetime of 0.520 ms is reported in ref.<sup>[47](#page-17-19)</sup>. The author suggests a with chemical stability in environments such as water, but also alkali and acid solutions. [Figure 4\(a\)](#page-7-0) is a series of photographs of the manufactured films under daylight and UV light, with the latter having a strong PL effect. The author explores the behavior of the films under me-chanical stress [\(Fig. 4\(b\)](#page-7-0) and  $4(c)$ ) as well as its flexibility (Fig.  $4(d)$ ), also suggesting applications in anti-counterfeiting applications and dermatoglyphics, further explained in the next subsections.

Some reported phosphors used for the purpose, as well as metrics related to this subset are listed in [Table 3.](#page-8-0) Correlated color temperature (CCT) is a widely used index to correlate the temperature of an ideal black-body radiator to that of the light source. The color rendering index (CRI) compares the colors in artificial light to the sun-light or standartized light source<sup>[67](#page-18-13)</sup>.

Changing the host matrix has influence on the behavior of the phosphor material. For the same powder of phosphor, the reported CCT was 4433 K, 6044 K and

*PDMS* : *phosphor* is in head-up displays (HUD), where 6413 K, for epoxy resin, PDMS and PMMA<sup>[43](#page-17-15)</sup>. A low CRI is related to a deficiency in red light, which usually tends to increase the CCT (higher CCT represents colder light, more close to blue)<sup>[78](#page-18-14)</sup>. Applying the PDMS coating allowed for it to maintain its characteristics over 300 h exposure, contrary to the reference without the encapsulation, which after 120 h had lost nearly 50 percent luminescence[68](#page-18-15). Another common application for the transparency of PDMS allows for a transparent display, not interfering with the field of view of the driver, while the phosphor projection displays information for the driver.

#### Anti-counterfeiting solutions

In this area, the *PDMS* : *phosphor* mixture is mainly used in the form of AC markers, which are invisible in visible light, but are clearly visible when excited by UV radiation. The production of these security tags is low-cost, and using standard screen printing techniques, it is possible to apply them to surfaces such as metal, fabric, pa-per, and others<sup>[38](#page-17-11)</sup>. These techniques generally imply laying the ink into a hollow patterned screen and subsequent curing of the ink in the material to be marked.

These mixtures are usually prepared with a powdered PDMS mixed with deionized water, where the phosphor is dissolved using ultrasonic baths to ensure uniform dispersion. The solutions are then degassed using vacuum to remove bubbles and cast-dried to form the transparent AC marker, as can be seen in [Fig. 5\(a\)](#page-9-0). In [Fig. 5\(b\)](#page-9-0) several printed markers are tested under different environmental conditions, such as different surfaces, long term storage, photo-stability and thermal treatment. [Ta](#page-9-1)[ble 4](#page-9-1) contains some of the most reported mixtures from the last decades with applications in anti-counterfeiting solutions, using phosphor materials emitting at different wavelengths, combined with PDMS.

A mixture of PDMS and  $La_2MoO_6: Sm^{3+}$  is reported in ref.[38](#page-17-11) emitting at 601 nm with excitation light of 365 nm and a quantum yield of 66.2%. In this application characteristics such as chemical and environmental stability are required. These materials must withstand chemical exposures and still ensure durability and relia-bility. In ref.<sup>[57](#page-18-7)</sup> the stability of the suggested AC marker is demonstrated by evaluating the degradation in different solvents and bleaches such as ethanol, acetone, detergent and hydrogen peroxide The printed security pattern must maintain its phosphorescence during the life cycle

<span id="page-7-0"></span>

**Fig. 4** | (a) Photographic images of the BaLa<sub>2</sub>ZnO<sub>5</sub>:Tb<sup>3+</sup>/PDMS composite folding films under daylight and UV light. (b) The  $BaLazZnO<sub>5</sub>$ :Tb<sup>3+</sup>/PDMS composite film was under applied mechanical stress and photographed before and after applying the mechanical stress under UV 270 nm light. (**c**) PL spectra of films after applying mechanical stress. (**d**) The flexibility of films under different weights (250, 500, and 750 gm). Figure reproduced with permission from ref.<sup>[47](#page-17-19)</sup>, Elsevier.

of the object to be protected by the AC marker. The photo-stability must endure prolonged UV exposure and thermal stress. Ensuring all these parameters enables to use these materials as consistent, hard to replicate authentication methods.

#### Temperature sensors and thermoluminescence

There are many phosphors that exhibit a significant dependence of luminescent behavior on temperature.

Y2O2S : Er*,* Yb mixed with PDMS to form a light, flexible There are several ways to perform temperature measurements using thermoluminescence, including fluorescence intensity ratio (FIR), luminescence lifetime and emission peak/band width (ref.<sup>[23](#page-17-3)</sup>). Reference<sup>[98](#page-19-0)</sup> reports a green emitting phosphor (550 nm) composed of thin film thermometer. This very cost-effective device was tested in electronic circuit boards and achieved decimal accuracy for specifically engineered temperature



<span id="page-8-0"></span>

ranges. The device uses a upconversion mechanism, emitting high-energy photons by fusing low-energy pho-tons<sup>[99](#page-19-1)</sup>, as visible in [Fig. 6.](#page-10-0)

Typically in cells or biological tissues, UV excitation may cause interfering luminescence from the surrounding medium, affecting measurements, in contrast to IR excitation, which stimulates visible fluorescence only on the target phosphor<sup>[98](#page-19-0)</sup>. In the case of upconversion phosphors, temperature can be measured using FIR relation with temperature, as in ref. $101$  where its use is suggested in silicon chips, where the difference in value for the calibration measurements can be related with the low thermal conductivity of PDMS (0.23 W/mK), especially compared with the silicon chip (167.00 W/mK).

of the emissions as reported in ref.<sup>[98](#page-19-0)</sup>. A PDMS : phosphor tions and previous reported  $\pm$  0.05 °C resolution and target of  $\pm$  0.03 °C, using a single layer deposited using spin Temperature can also be measured by analysing the decay times of the phosphorescent state. The temperature increase decreases the decay times and the rise time mixture is reported in ref.[26](#page-17-4) with thermometry applicacoating, which can further reduce manufacturing costs

<span id="page-9-0"></span>

**Fig. 5 |** (**a**) Diagram of *PDMS*:*phosphor* AC inks manufacturing process. (**b**): (α) AC labels created using screen-printing mode on different surfaces and visualized under day light and UV 365 nm light. (β) Long term storage of AC labels up to 270 days. (γ) Photo-stability test examined up to 120 min using UV source. (δ) Developed AC image on marble and thermally treated up to 250 °C for 20 min (β1 & δ1) Pixel profiles of experi-ments on developed AC labels (γ<sup>1</sup>) PL measurement on UV irradiated AC label. Figure reproduced with permission from ref.<sup>[38](#page-17-11)</sup>, Elsevier.



<span id="page-9-1"></span>

when optimized. A device achieving repeatability and cyclic temperature measurement with the worst resolution recorded being 0.0011%.K and the best 0.00436%.K (423 K) in the temperature range of 353–523 K is reported in ref.[24](#page-17-23), so for short-term it can withstand higher temperatures than the specified by the manufac-

turer, however is not recommended since it can alter the intrinsic characteristics of PDMS. [Table 5](#page-10-1) contains some reported mixtures with applications in temperature sensors and thermoluminescence. PDMS Sylgard 184 has a temperature range specified by the manufacturer as −50 °C to 200 °C, limiting its use to this operational range.

<span id="page-10-0"></span>

**Fig. 6 | Diagram of the upconversion mechanism where the energy absorbed by ion 1 and 3 (E1) is transferred to ion 2 which then emits a photon with higher energy than the ones absorbed when returning to the fundamental state (G1).** Figure reproduced with permission from ref.<sup>[100](#page-19-18)</sup>, Elsevier.

#### Bio-medical applications

The mixture of PMDS and phosphor can be a hot topic for biosensors due to several key reasons like enhanced sensitivity (where Phosphor materials can exhibit strong and stable phosphorescence, which can be highly sensitive to changes in the environment. This makes it possible to detect low concentrations of biological targets with high sensitivity; and the combination of PDMS with phosphor can amplify the detection signal, making it easier to detect small changes in the biosensor's environment). Secondly, both PDMS and phosphors are chemically stable materials, which helps in maintaining the integrity and functionality of the biosensor over time as well as these materials can also provide thermal stability, ensuring that the biosensor performs reliably across a range of temperatures. Third point, PDMS is a flexible and stretchable polymer, which can be useful for developing wearable biosensor[s](#page-16-4) or sensors that need to con-form to irregular surfaces<sup>[5](#page-16-4)</sup>. The PDMS matrix enables a

strain sensing in a wide range of strain, spanning up to several hundred percent in comparison to the conventional rigid matrix composites and ceramic-based mechanoluminescence (ML) sensors<sup>[106](#page-19-19)</sup>. It is reported that the mixture of PDMS with ML phosphors is suit-able for monitoring of human motions<sup>[107](#page-19-20)</sup>. Mechanoluminescence usually implies applying pressure to a previously excited ion releases a trapped hole to the valence band of the material. This trapped hole can recombine with the ion leading to a unstable state. When returning to a stable state the ion will emit the extra energy in the form of phosphorescent light, which can be correlated with the applied pressure<sup>[27](#page-17-5),[28](#page-17-27)</sup>. [Figure 7\(a\)](#page-11-0) illustrates a ML process example.

In this way, such mixture can be used in various types of biosensors, including optical, electrochemical, and mechanical sensors, providing a broad range of applica-tions<sup>[108,](#page-19-21)[109](#page-19-22)</sup>. Not less important is biocompatibility feature: both PDMS and certain phosphor materials are biocompatible, making them safe for use in direct contact with biological tissues and fluids. This biocompatibility ensures that the sensors do not induce adverse biological responses, which is critical for *in vivo* applications. Due to the high elasticity of PDMS (and therefore the PDMS phosphor mixture), at relatively small compressive and tensile forces acting on the PDMS (or the PDMS phosphor mixture), there are negligible changes in the compression or stretching of the PDMS or the PDMS phosphor mixture. Hence, this high elasticity is the main factor for the easy and highly efficient use of mechanoluminescence in the mixture of PDMS and ML phosphor. And this key feature is then easily used in many pressure and tensile biosensors. Customizable properties (by adjusting the ratio of PDMS to phosphor, the mechanical properties of the sensor can be tuned to meet specific requirements, such as elasticity and hardness) and optical tuning, such as emission wavelength and intensity, can be adjusted by choosing different types of phosphors, allowing for the customization of the sensor for specific applications.

**Table 5 | Phosphors combined with PDMS for temperature sensors or thermoluminescence.**

<span id="page-10-1"></span>

Phosphor	Ref.	Phosphor	Ref.
LuNbO <sub>4</sub> : $Eu^{3+}$ /Sm <sup>3+</sup>	ref $^{24}$	La <sub>2</sub> O <sub>2</sub> S:Eu	ref $26$
$Ca_2Sb_2O_7 : Eu^{3+}$	ref $^{25}$	La <sub>3</sub> Ta <sub>0.8</sub> Sb <sub>0</sub> .2O <sub>7</sub> : 0.04Bi <sup>3+</sup> , zSm <sup>3+</sup> (0 $\le$ z $\le$ 0.03)	ref $^{23}$
$SrWO4: [Er3+]/[Yb3+]$	ref <sup>99</sup>	$NaNbo3: Pr3+, hexagonal boron nitride nanosheets$	ref $102$
$Y_2O_2S : Er, Yb, La_2O_2S : Yb, Er$	ref <sup>98</sup>	La <sub>2</sub> O <sub>2</sub> S:Eu	ref $103$
La <sub>2</sub> O <sub>2</sub> S:Eu	ref $104$	NaGdTi <sub>2</sub> O <sub>6</sub> : $Pr^{3+}$ , $Er^{3+}$	ref <sub>.105</sub>

<span id="page-11-0"></span>Rendeiro R et al. Opto-Electron Adv 7, 240133 (2024) <https://doi.org/10.29026/oea.2024.240133>



**Fig. 7 | The ML properties of the sensor for visual sensing.** (**a**) ML mechanism of SrAl2O4:Eu2+, Dy3+. (**b**) The dependence of the ML spectra at varying pressure in the range from 1 MPa to 30 MPa. Figure reproduced with permission from ref.[27](#page-17-5), IOP Publishing.

ties. New bio-medical applications of *PDMS* : *phosphor* to 30 MPa using PDMS:SrAl<sub>2</sub>O<sub>4</sub> :  $Eu^{2+}$ , Dy<sup>3+</sup> was re- $ZnGa_2O_4: Cr^{3+}$  and are activated using mechanoluminescent  $SrAl<sub>2</sub>O<sub>4</sub>$ : Eu<sup>2+</sup>, remotely actived using ultrason-Both PDMS and phosphors are relatively inexpensive materials, which can help in reducing the overall cost of biosensor production. For scalable manufacturing, the fabrication process for PDMS-based biosensors is scalable, allowing for mass production and wider adoption in various fields. Phosphorescent signals typically have longer lifetimes compared to fluorescent signals, which can help in reducing background noise and improving the signal-to-noise ratio in biosensor readings. As last point, PDMS is commonly used in microfluidic devices due to its ease of molding and bonding. Integrating phosphor materials into PDMS can create advanced microfluidic biosensors with enhanced detection capabiliblends are being published every year, showcasing that it is a high interest topic both for industrial and academical communities. According to ref.[110](#page-19-27) ML also occurs if the device is stretched, allowing its application into tension sensors. A pressure sensor measuring from 1 MPa ported by ref.[27](#page-17-5) with ML intensity linear relation with the pressure applied, visible in [Fig. 7\(b\).](#page-11-0) These devices are bio-compatible and flexible, allowing its application into the human tissues or flexible fabrics<sup>[110](#page-19-27)</sup>. Mechanoluminescence has also been used in Photothermal Therapy, where tumor cells are selectively destroyed without affecting normal tissues, using photothermal agents. These agents usually have persistent luminescent, such as, ic waves. A device capable of multicolor fluorescence imaging is reported in ref.[111](#page-19-28). This device uses PDMS light-guide plates to guide the transmitted light from the light source only to the fluorescent samples. PDMS host

*PDMS* : *phosphor* based devices can be used to detect tuworks in ML using different PDMS : phosphor mixtures. matrix can be doped with phosphors to enhance the PL intensity of the signal, obtaining higher contrast imaging of the samples. This technology can also be used to mon-itor in real-time drug releases in specific targets<sup>[112](#page-19-29)</sup>. mor biomarkers, metabolites, biomolecules, and other signal parameters from living cells. Luminescent properties of nanoparticles gained notoriety as potential in cell stimulation and tissue growth, providing more efficient tissue engineering strategies<sup>[112](#page-19-29)</sup>. [Table 6](#page-12-0) comprises several

#### Other selected applications

One of the possibilities of using the *PDMS* : *phosphor* mixture is in the area of reducing biological pollution in marine infrastructure, heavily affected by bio-foulings. Diatoms, unicellular algae, are known to be one of the major bio-fouling agent. A mixture of PDMS and Waterproof long afterglow phosphors (WLAP) is used to influence the physiological activity of diatoms. WLAP absorbs and stores energy from daylight, which then emits weak fluorescence at night, which limits the physiological activities of diatoms. According to ref.[126](#page-20-0), with the use of PDMS and WLAP, attachment reduction rate was 34.5% compared with the blank control sample and a diatom removal rate after wash of 42.3%.

growing. Therefore, the mixture of PDMS : phosphor can As referred in subsection *White-LED's and display technology*, white emitting LEDs using phosphor coatings is widely used as illumination, however, for certain applications other spectrums are required, such as plants also be engineered to emit in specific wavelengths preferred by the plants. With this approach the efficiency of indoor growing can be further optimized, because all the

#### <span id="page-12-0"></span>Rendeiro R et al. Opto-Electron Adv 7, 240133 (2024) <https://doi.org/10.29026/oea.2024.240133>



**Table 6 | Phosphors combined with PDMS for mechanoluminiscence.**

the plant. A mixture using  $Sr_2ScSbO_6$ :  $Mn^{4+}$  is reported  $Rb_xK_{2-x}CaPO_4F:Eu^{2+}(0 \le x \le 2)$ , which emitted in 495 light emitted can be in fact the required and absorbed by in ref.[127](#page-20-1) with an emitting peak at 697 nm if under 365 nm UV light excitation, matching with the far-red light photosensitive pigment of the plant. According to ref.<sup>[128](#page-20-2)</sup>, using 313 nm and 380 nm exciting light in nm and 665 nm, increased the growth rate of Chlorella 12.2% when compared with the control group. Plants require UV and blue wavelengths which can degrade the host material after long exposure, requiring the host ma-trix to be resistant to these radiation, such as PDMS<sup>[129](#page-20-3)</sup>.

current frequency applied to the PDMS : phosphor de-The phosphor capabilities can also be harnessed to modulate the emitted light by varying the alternating vices and PDMS allows it to have stretchability. According to ref.[130](#page-20-4) a device built with PDMS:ZnS:Cu emitted green (0.165, 0.320) with 1 kHz to blue (0.121, 0.131) with 100 kHz. A device where varying the current frequency from 100 Hz to 500 Hz changes the color coordinates from (0.24, 0.42) to (0.23, 0.32) is reported in ref.[131](#page-20-5), with ZnS:(Cu, Cl) as phosphor material. In [Fig.](#page-12-1)  $8(a)$  is possible to observe the schematic diagram of the device, AC source and emission.

Another potential application is in Visible Light Com-munication (VLC), which according to ref.<sup>[86](#page-19-6)</sup> can replace Wi-Fi in the future, being able to combine illumination and communication. A device capable of optical communications is reported in ref. $35$ , using as information units a multi-color display, activated by pressure on the copper outer layer. The author suggests to use the different emitted colors to code data to be transmitted, as visible

<span id="page-12-1"></span>

**Fig. 8 |** (**a**) Schematic of light emission from the mixed ZnS composite and PDMS with applied AC bias. (**b**) Schematic diagram of self-powered optical communication system consisting of information inputs (instantly dynamic self-powered multi-color display), information acquisitions (cameras), information processing (MCU), and information display (display screens). Four information units (00, 01, 10, and 11) and corresponding states of multi-color self-powered ACEL system. Figure reproduced with permission from: (a) ref.<sup>[131](#page-20-5)</sup>, under a Creative Commons Attribution Non-Commercial License; (b) ref.[35](#page-17-9), Elsevier.

#### in [Fig. 8\(b\)](#page-12-1).

However, one issue related with using this technology for communication is the decay time of the phosphor. In optical communication the higher the bit rate and transferred data the higher the frequency at which the light must be modulated. The high decay times of the phos-phors limit its available bit rate<sup>[132](#page-20-17)</sup>.

Carbon dots have been reported to react with metal ions, altering its luminiscence when excited under UV light and thus with potential use in metal sensing<sup>[31](#page-17-7)</sup>. According to ref.[133](#page-20-18) several carbon dots obtained from different biowaste sources are evidenced to have phosphorecent properties, with each interacting in its own manner with the metal ions. The author also suggests this relation to develop metal ion detectors.

PDMS:Ba<sub>2</sub>LuNbO<sub>6</sub>: Tb<sup>3+</sup> is reported in ref.<sup>[134](#page-20-19)</sup>, with a X-ray photons are highly energetic and typically are sensed using scintillators and charge-couple device (CCD) sensors. There is a need to use scintillators, which convert the X-ray energy into visible photons, that can be detected by a photodetector, as a CCD sensor or Positive-Intrinsic-Negative (PIN) diode. In this realm, PDSM:phosphor mixtures are of high interest, due to the cost-effective X-ray sensing solution, especially when compared with typicall scintillators made of crystals, such as Cesium Iodide (CsI). Photodetectors for visible light are the most documented and matured, allowing for a high resolution very cost-effective sensing. In this application the higher the resolution possible the better. Resolution is thus a function of the scintillator and the photodetector. A flexible scintillator film made of spatial resolution of 12.5 lp/mm, compared with a commercial scintillator in [Fig. 9\(a\).](#page-13-0)

the mixture PDMS: NaBaScSi<sub>2</sub>O<sub>7</sub>: Eu<sup>2+</sup> and, using ampli-Fiber optical pH sensors typically use as detection techniques wavelength or amplitude modulation. The first is based on refractive index changes in the sensing layers of the optical fiber, usually having high requirements for the coating and material properties. The latter involves measuring the variation in the output intensity of fluorescent indicators. A sensor manufactured with tude modulation, displayed a linear response, visible in [Fig. 9\(b\)](#page-13-0) and a sensitivity of 0.05/pH in a range of 6.86–9.18 pH $^{135}$  $^{135}$  $^{135}$ .

ing a PDMS: phosphor junction converting the peak Tryptophan, which is an amino acid, plays an important role in the production of serotonin, melatonin, niacin, and nicotinamide<sup>[136](#page-20-21)</sup>. Tryptophan fluorescence can be used to track cellular proliferation in events such as wounds closure, neoplasm and others chronic conditions with the emission peak located around  $345 \text{ nm}^{137}$  $345 \text{ nm}^{137}$  $345 \text{ nm}^{137}$ . However, in general, the transmission of optical fibers in this wavelength range is lower. The author suggests usemission to a 450–650 nm range, improving UV emit-ters monitoring<sup>[137](#page-20-22)</sup>.

Metasurfaces are a new brand of optical engineered materials which use sub-wavelength structures to control the light, as if classical refractive optics, but with the advantage of being flat and thin. To manufacture metasurfaces is thus required to produce structures such as grooves in the nanometer scale. A combination of PDMS:phosphor is suggested in ref.<sup>[33](#page-17-30)</sup> for a metasurface engineered flexible sticker with luminescent characteristics. These films can found applications in photovoltaic panels. The upconversion mechanism overviewed in section *Temperature sensors and thermoluminescence* as po-

<span id="page-13-0"></span>

**Fig. 9** | (a) X-ray energy absorption spectra of Ba<sub>2</sub>LuNbO<sub>6</sub>:Tb<sup>3+</sup> and the commercial scintillator Bi<sub>4</sub>Ge<sub>3</sub>O<sub>12</sub>. (b) Normalized optical loss relation with pH. Figure reproduced with permission from: (a) ref.<sup>[134](#page-20-19)</sup> and (b) ref.<sup>[135](#page-20-20)</sup>, respectively.

PDMS:  $β$ -NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup>, a phosphor with peak tential uses in photovoltaic applications. Commercial photovoltaic panels are manufactured using silicon, which has lower absorption in the low-energy end of the visible spectrum, with trace amounts of IR photons being absorbed. In a way to increase efficiency of photovoltaic panels, ref.[138](#page-20-23) suggests coating the panels with emission at 543.5 nm.

fective sensor for analytes, such as dissolved  $CO_2$ , with ing (DLR) using a film of PDMS: $Ru(dpp)_{3}^{2+}$  used as refachieving a  $R^2 = 0.993$ , as visible in [Fig. 10\(b\)](#page-14-0). Combin-Another suggested application is a robust and cost-efits scheme visible in Fig.  $10(a)^{34}$  $10(a)^{34}$  $10(a)^{34}$ . This device uses a phase-based sensing technique, luminophore referencerence luminophore and other fluorescent indicator, ing bio-compatibility and dissolved gas detection allows for real time monitoring in tissues, cell cultures or water as reported in ref.[139](#page-20-24), achieving a low limit of detection of 0.01 mg/L, a high sensitivity of 16.9 and a short response time (22 s).

ref.<sup>[98](#page-19-0)</sup>. The PDMS:  $Y_2O_2S$ : Er, Yb mixture can be intro-An application in heat flux sensing is suggested in duced in fluids, with the phosphor embedded into the PDMS matrix to form small sensing clusters, and using thermoluminescence, previous referred in section *Temperature sensors and thermoluminescence* is thus possible to map the heat flux along liquid solutions. Pressure-sensitive paints, used in the aerospace industry for aerodynamic studies of the aircraft are of great interest. A matrix of PDMS with Tetra (pentafluorophenyl)porphine (PtTFPP) was reported by ref.<sup>[140](#page-20-25)</sup> to have a short response time and very low photo-degradation rate. Using the re*PDMS* : *phosphor* mote optical sensing capabilities of the several uses where a blend of *PDMS* : *phosphor* was  $films<sup>33</sup> suggests using them as non-destructive, remote,$  $films<sup>33</sup> suggests using them as non-destructive, remote,$  $films<sup>33</sup> suggests using them as non-destructive, remote,$ instantaneous, and customizable sensors to identify geometrical defects in aerogels and elastomers, materials that serve as critical structural components while operating in extreme conditions, such as PDMS. Another well investigated use of these misture is in dermatoglyphics studies. Dermatoglyphics is the scientific study of indicators in the skin such as fingerprints. Using these materials can enhance the ability to distinguish patterns and to improve imaging qualities<sup>[47](#page-17-19)</sup>. [Table 7](#page-15-0) summarizes some of applied.

#### Discussion and future perspectives

The PDMS : phosphor mixture has many advantages, ing PDMS : phosphor based solutions is the easy manuwith a symbiotic relationship existing and bringing several advantages. The optical tunability in all the visible spectrum that engineered phosphorescent materials hosted in a PDMS matrix offers is of high interest in many industries. It is a core technology for cleaner energetic transition in illumination and displays solutions worldwide and will continue to further help, for instance in food production, with higher efficiency indoor growing systems. PDMS is a well known bio-compatible material, and this characteristic as well as its polymeric structure giving it a elastic and stretchable form, allows for several bio-medical applications, such as sensors or bio-imaging. Another well documented advantage of usfacturing with high repeatability and cost-effective solutions. Some of these characteristics in phosphor materi-

<span id="page-14-0"></span>

Fig. 10 | (a) Schematic of sensor probe excitation configuration and collection scheme. (**b**) Calibration curve of PDMS-based CO<sub>2</sub> sensor. Figure reproduced with permission from ref.[34](#page-17-8), Elsevier.

#### <span id="page-15-0"></span>Rendeiro R et al. Opto-Electron Adv 7, 240133 (2024) <https://doi.org/10.29026/oea.2024.240133>



**Table 7 | Phosphors combined with PDMS for uses in various applications.**

<span id="page-15-1"></span>

**Fig. 11 | Capabilities of the** *PDMS***:***phosphor* **mixture for future perspectives applications.**

als are summarized in [Fig. 11](#page-15-1).

However, despite all the advantages of these blends, there are certain issues to be solved, such as optical quenching. As referred before, PDMS has a low thermal conductivity, which can affect the measurements if used in thermometry applications but also affects its ability to disperse heat. This creates others problems also referred such as temperature quenching, that with time reduce the PL intensity. The luminance and efficiency of the phosphor over time is reduced due to poor thermal management causing crackings in the interface between the host and the phosphor<sup>[55](#page-18-8)</sup>. Its temperature range can also be a limiting factor for the mixture if the application requires temperatures lower than −50 °C and higher than 200 °C, or a use of a specifically tuned PDMS, since most commercial available PDMS only withstand the referred temperature. The mechanical properties of PDMS are changed after introducing the phosphor, which may lead to increased brittleness or reduced flexibility. Using PMDS:phosphor blends requires a trade-off between mechanical robustness, and thus durability, and optical efficiency, which needs to be engineered for the specific application. If there is a phase separation or uneven distribution between the PDMS and the phosphor the luminescence will be non-uniform and the performance will be reduced. This however, is a feature that can be exploited for new anti-counterfeiting solutions.

On the other hand, it is very likely that many new applications will be discovered in some areas, especially in the field of mechanoluminescence where new bio-medi-

areas, the PDMS : phosphor mixture has so far been little be to find a better use of the *PDMS*: *phosphor* mixture cal applications are reported frequently. Combining mechanoluminescence with piezoelectricity on a device using PDMS as host, as reported in ref.<sup>[118](#page-20-9)</sup>, provides a comprehensive understanding of mechanical events, providing qualitative and quantitative information about strain-related issues and use artificial intelligence (AI) to further optimize the device. PDMS alone has been reported to be used to manufacture bioinspired multifunctional flexible optical sensor achieving sub-millimeter spatial resolution for force location sensing, plus a  $\mu$ N resolution in force assessment<sup>[156](#page-21-7)</sup>. Electronic skin, wearable electronic sensors mimicking the functionalities of the human skin, manufactured using PDMS have been reported by ref.[157](#page-21-8). These new bioinspired sensors can leverage 3D printing techniques and AI to manufacture complex structures and mixed phosphors can help to boost the collected signal or to retrieve information about the health of the sensor. Here AI will speed the development of sensor technology due to the complex correlations and high-throughput of data in the biomedical contexts, as p.e. in ref.<sup>[2](#page-16-1)</sup>, where AI is used to distinguish between signals produced by different pressure and movements in mechanoluminescence devices. In some used, e.g. in the area of VLC. Perhaps the challenge could for this area as well, using different phosphor with different energy levels to be able to expand the available range. In photovoltaics area new explored solutions can increase the efficiency of new devices, as well as being able to increase its longevity, with easy to manufacture PDMS films.

## **Conclusion**

the PDMS : phosphor mixture over the last decade have related to the use of the PDMS : phosphor mixture. In this comprehensive review, all known applications of been presented. Overviews of phosphors used for individual applications were also presented here, as well as some advantages, disadvantages and trade-offs of using these blends. We believe that this detailed breakdown of phosphors can be very useful for further research work

To conclude, the combination of PDMS with phosphorescent materials is developing and booming due to its unique optical and mechanical properties while being easy to manufacture and cost-effective. Its versatility and ability to combine with other technologies is a core advantage. Despite having some issues to be solved it is a

standard in a wide range of industries from biomedical to illumination, with significant potential applications to be discovered using new processing techniques and innovative material design, while managing trade-offs for each particular application.

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#### Competing interests

The authors declare no competing financial interests.



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