Rev. Adv. Mater. Sci. 42 (2015) 92-101

A REVIEW ON IMPROVEMENT OF LED LIGHT EXTRACTION EFFICIENCY THROUGH A MICRO REPEATING STRUCTURE

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Received: December 24, 2014

Abstract. This paper explains the method used by researchers to improve Light Emitting Diode (LED) extraction efficiency. It also summarizes the works of different researchers on the improvement in light extraction efficiency (LEE) of an LED. In current practice, a commonality was observed where many researchers tried to improve LEE through the use of micro repeating structures (MRS). MRS comes in the shape of pyramid, lens, cylinder, perforated holes or a combination of a few random shapes. The LEE of an LED could be improved by placing a uniformly aligned or abruptly patterned MRS onto the LED surface in the desired area of projection. This review looks at the possibility of using mesoporous thin film (MTF) with ordered pore to improve LEE due to the presence of uniform MRS (a structure which consists of silica and air array).

1. INTRODUCTION

The objective of this paper is to demonstrate that LED light extraction could be improved by using thin film mesopores. Thin film mesopores have a structure which is MRS in nature. At present many researchers have reported in different methods of surface modification showing MRS structure tends to improve LEE. Amongst the various MRS reported are embossed repeating microlens [1], 2D photonic crystal as seen in Fig. 1 [2,3], Bragg's grating [4], surface-roughening and many more. The surface modifications to improve LEE of an LED are mostly repeating structures in the nano or micro scale. As an alternative, depositing MTF on LED chip is possible to improve the final luminous flux of the LED. This is because mesoporous materials could be fabricated to have repeating structures. It is possible for MRS to improve LEE because of the presence of an uneven surface for light wave to hit at random angles [5], act as a waveguide similar to a microlens [2] or that is similar to a photonic crystal for improved outcoupling effect [3].

The reason to improve LEE of LED technology is expected to have positive future prospects in the market. Features such as that it uses less energy [6], with efficiency far better than of incandescent lamps and is almost on par with fluorescent lamps [7,8] and in future is expected to exceed all other technologies ensure that this technology gets much attention. It can also show improved Color Rendering Index(CRI) [9]and has a long lifetime that goes up to 100,000 hours [10]. By referring to the statistical analysis done by the Department of Electronics of Aalto University, about one fifth of the global electricity generated is used for lighting purposes. It is estimated that 90% of electrical energy could be saved by using smart lighting technologies combined with LEDs [11]. A good example of energy

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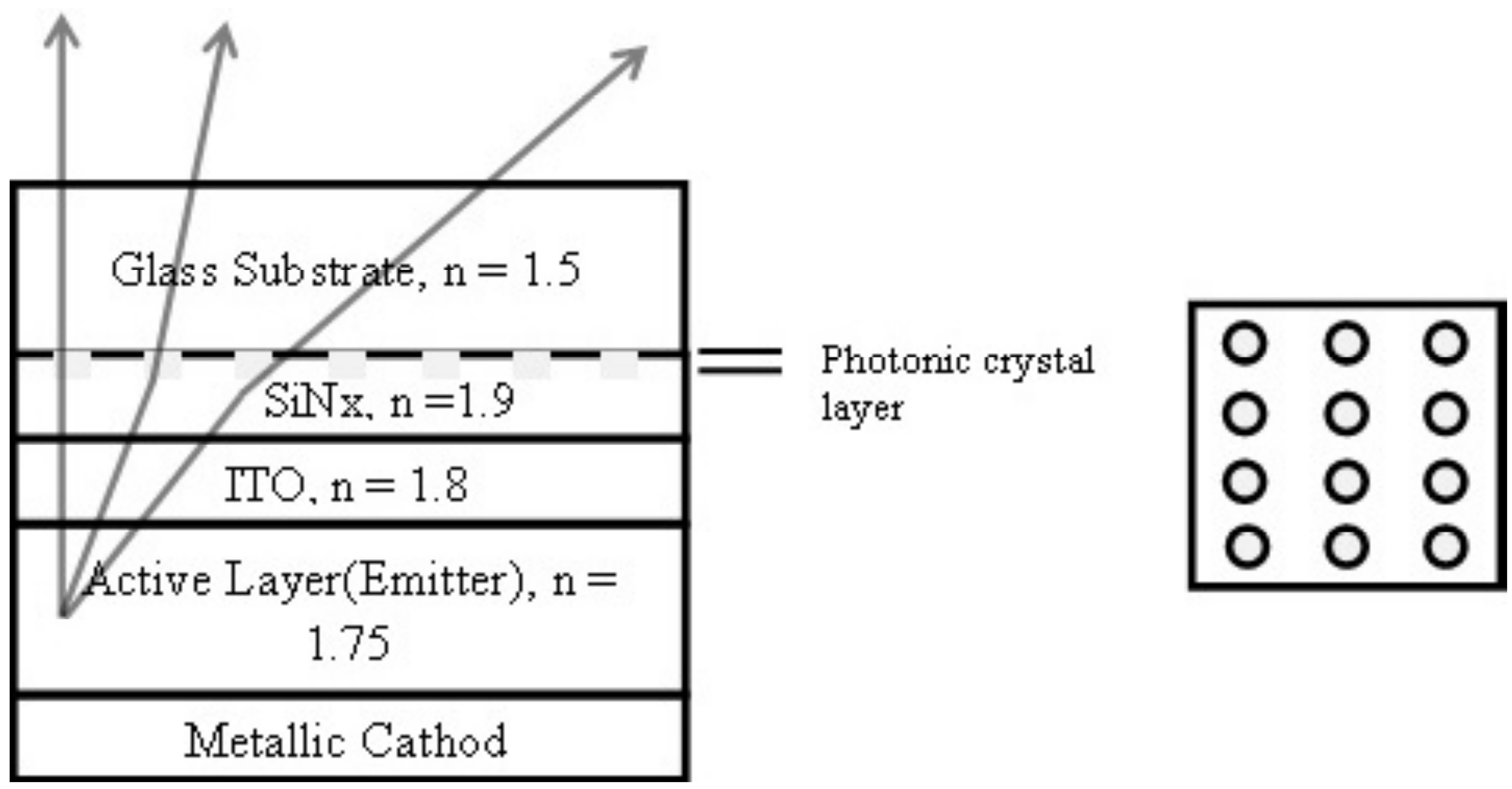

Fig. 1. Pillar structure of the 2D photonic crystal. The photonic crystal is acting as a waveguide, Reprinted with permission from Y.-J. Lee, S.-H. Kim, J. Huh, G.-H. Kim, Y.-H. Lee, S.-H. Cho, Y.-C. Kim and Y.R. Do // *Applied Physics Letters* **82** (2003) 3779. Copyright 2003, AIP Publishing LLC and from "Review on the light extraction techniques in organic electroluminescentdevices", Kanchan Saxena, V.K. Jain, Dalip Singh Mehta, Optical Materials 32 (2009) 221–233, © 2009 Elsevier.

savings can be seen in New York City where 70% of its traffic lights have been converted to LED technology, and the estimated annual savings for maintenance and electricity goes up to $6 million [12]. The total estimated electricity saving in the US is estimated to be around $125 billion from 2005 to 2025 [10]. Cumulative savings for the total amount of electricity and maintenance in the US is estimated to reach as high as $300 billion from 2009 to 2030 [12].

2. MESOPOROUS THIN FILM MATERIAL AND ITS POTENTIAL APPLICATION ON IMPROVING LIGHT EXTRACTION EFFICIENCY

Mesoporous materials which are applicable for film, bulk or powder forms are defined by IUPAC as materials with pore sizes between 2 and 50 nm respectively [13]. The presence of pores in the material makes it low density, having a large surface area which is suitable for catalysis, and in certain cases, the ability to retain matter. In other words, it is a material which has different properties from a bulk material [14] and reported as transparent [15-17]. There are many types of porous material films made out of metal oxide such as Cobalt(II,III) Oxide [18], Titanium Dioxide(Titania) [19], Zirconia [15], nickel oxide [20], zeolite materials such as mordenite [21], carbon [22] and silica.

Many applications are possible for mesoporous materials such as:

i) Enhancing ionic conductivity by caging the ions in the inert mesoporous material [23],
ii) Acting as a molecular sieve to control diffusion [24-27], or acting as adsorbent for material separation and purification [25,28,29] or, in certain cases, for material storage and transportation purposes [28,30],
iii) For catalysis purposes [26-28,31,32],
iv) Used as waveguide [32,33],
v) A template to fabricate nanomaterials [34] and
vi) Used in fuel cell application such as in direct methanol fuel cell as an electrocatalyst [22] or in solid oxide fuel cell [15].

Mesoporous material is an MRS due to the presence of repeating silica and air array. A new application besides the ones mentioned above is possible, which is to improve LEE on LEDs. Some of the examples of the structures reported to improve LEE as seen in Fig. 2: a) [35], b) [36], c) [37], and d) [38]. Fig. 2e [39] show mesoporous structures fabricated through the use of a surfactant template. It shows that it is thus possible for MRS pattern to improve LEE.

3. REPEATING MICRO STRUCTURE ON LED TO IMPROVE LIGHT EXTRACTION EFFICIENCY

Jeong [40] reported that the internal quantum efficiency of an OLED is nearing 100%, whereas the light extracted is only 20%.The rest are lost within the OLED unit structure. Below are the reviews of experiments done by researchers to improve LEE.

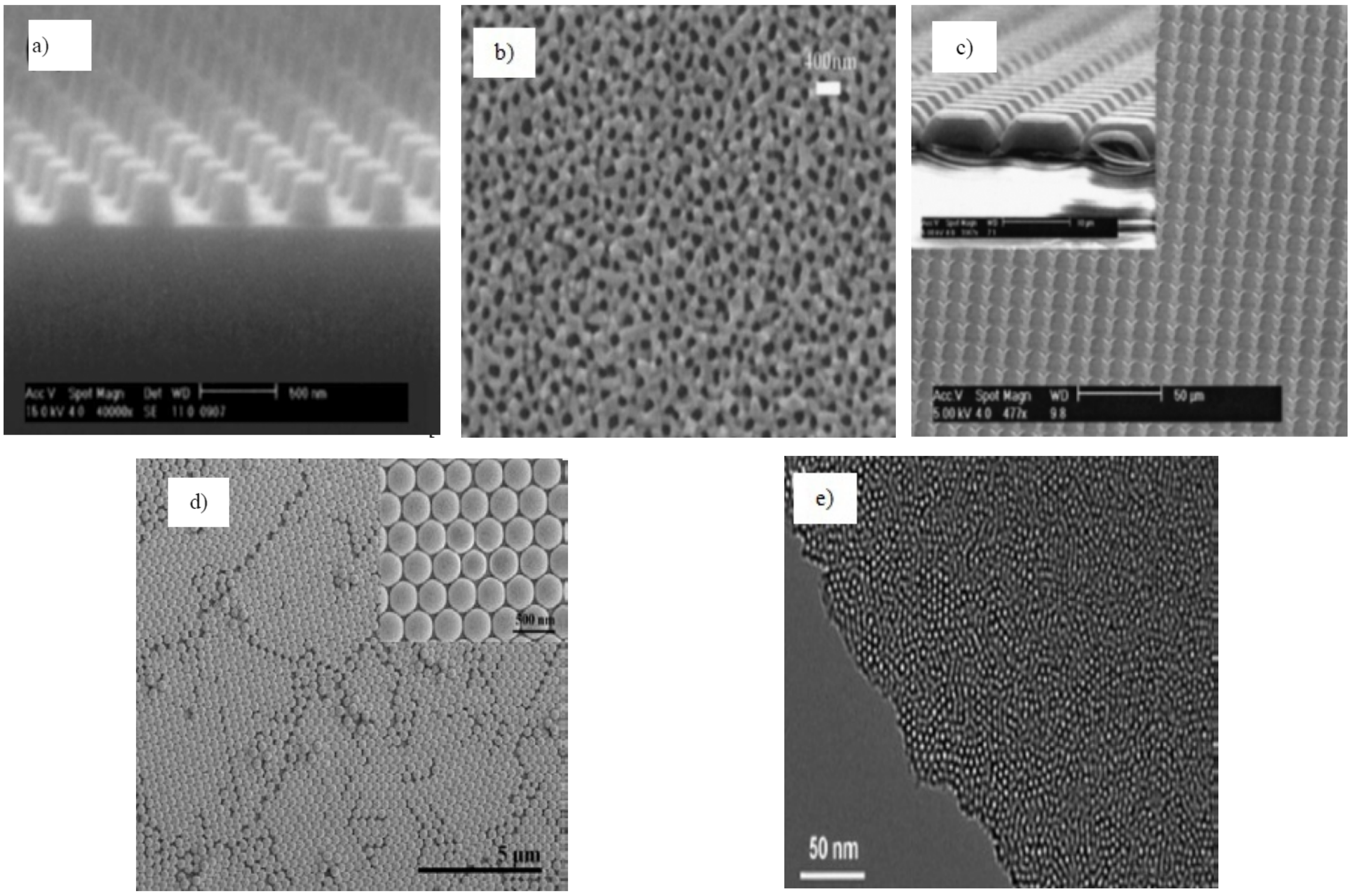

Fig. 2. Shows some of the MRS structures to improve LEE of an LED. Picture a) to d) are the examples of MRS used on LED. Picture e) shows mesopores fabricated through surfactant template. (a) Reprinted with permission from Y.R. Do, Y.-C. Kim, Y.-W. Song and Y.-H. Lee // *Journal of Applied Physics* **96** (2004) 7629. Copyright 2004, AIP Publishing LLC; (b) Reprinted with permission from H.J. Peng, Y.L. Ho, X.J. Yu and H.S. Kwok // *Journal of Applied Physics* **96** (2004) 1649. Copyright 2004, AIP Publishing LLC; (c) Reprinted with permission from S. Möller and S.R. Forrest // *Journal of Applied Physics* **91** (2002) 3324.. Copyright 2002, AIP Publishing LLC; (d) Reprinted with permission from C.S. Choi, S.-M. Lee, M.S. Lim, K.C. Choi, D. Kim, D.Y. Jeon, Y. Yang and O.O. Park // *OPTICS EXPRESS* **20** (2012) 309, Copyright 2012 The Optical Society; (e) reprinted with permission from Z. Teng, G. Zheng, Y. Dou, W. Li, C.-Y. Mou, Xuehua Zhang, A.M. Asiri and D. Zhao // *Angewandte Chemie International Edition* **51** (2012) 2173, (c) 2012 John Wiley & Sons Inc.

One of the MRS is 2D photonic crystal which looks somewhat like a waveguide in the shape of fiber optics. The top view of a 2D structure is illustrated on the right corner and the side view as seen in the larger picture in Fig. 1 respectively. Through understanding the workings of a fiber optics, light wave is guided within the fibers through total internal reflection (TIR). TIR occurs if light wave is incident above the critical angle and the refractive index of cladding is lower than the core [41]. It is possible to amplify the guided mode and the narrowing of emission respectively, as shown by Marlow [42], with a mesoporous silica fiber. As an alternative, Yi [43] uses a different approach of an omnidirectional reflecting waveguide by having a lower refractive index core covered by layers of alternating Si and Si_3N_4 with higher refractive index.

An illustration is shown in Fig. 3 to simplify the theory behind light being guided out through perpendicular pores from an emitter by Lee [2] and Mehta [3]. This is somewhat analogous to a 2D photonic crystal, as seen in Fig. 1, due to different refraction index. Tsai [44] states that by using a two-dimensional photonic crystal, a 3.5 times in output power was observed within the angle of 30°. In another paper, Do [35] reported an increase of 50% of LEE.

A different type of MRS structure was reported by Tuohioja [1] which is through embossing repeated microlens structure onto the encapsulant material of an LED package. Tuohioja collimated light output by narrowing the spread from a 108° to 45° angle and increased the luminous intensity by 70%. The narrowing of spread was described by Schreiber [5], which is a result of the reduction of stray light and

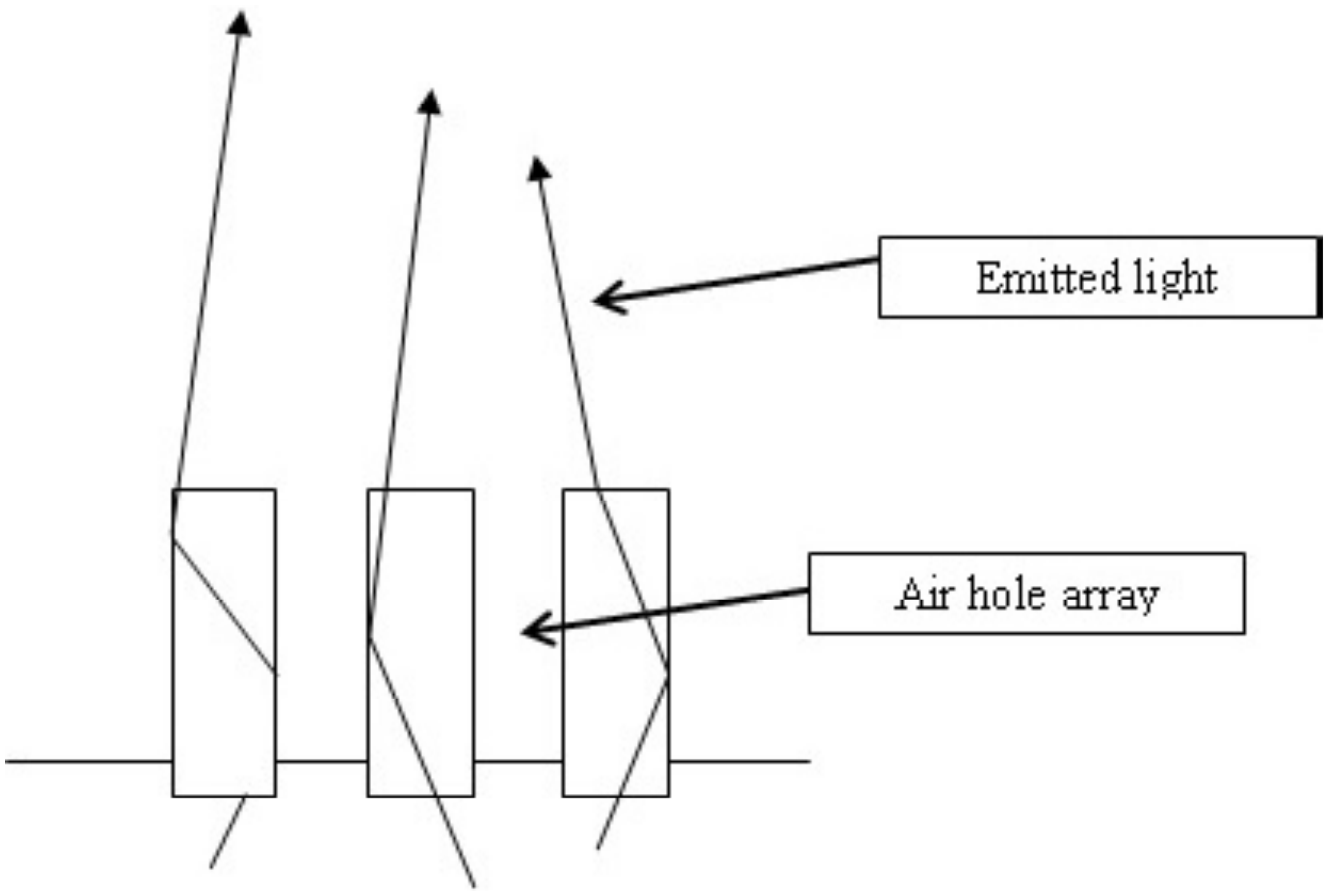

Fig. 3. A simple illustration of a repeating pillar structure followed by air hole array. The arrow shows illustration of light being guided out of the repeating pillar structure.

increase in homogeneity of light. However the percentage of increase in intensity was not mentioned by Schreiber.

Aside from the methods mentioned above, Ziebarth [4] used Bragg gratings fabricated through soft lithography. The forward emission intensity was increased by 49% and 70% through the use of one-dimensional and two-dimensional gratings respectively. The quantum efficiency was increased by 15% and 25% respectively. The difference is in the direction of light incident upon these structures.

On the contrary, a naturally occurring MRS is reported by Bay [45], whereby a rough misfitting surface such as the abdominal structure of a firefly is much more efficient at extracting light compared to a smooth surface. This abrupt pattern of repeating scales joined together produces diffuse transmission and it could improve LEE up to 55% [46]. Bay's [46] evaluation showed that the period and height of the MRS should be 5 μm and 6 μm respectively.

Since abrupt patterns could improve LEE, surface-roughening which consists of abrupt patterns in different shapes is proven to improve LEE. For example a well or inversed pyramid structure [6] or a truncated micro-pyramid [47] formed on the LED chip top surface is reported to increase the LEE at 73% and 60% respectively. An example of surface roughening could be seen in an SEM scan in Fig. 4b. Fig. 4a which is a mirror polished wafer was taken as a comparison. The surface-texturing was able to guide light out of the LED chip by allowing light to hit the interface at arbitrary incident angles [35]. This could be termed as a waveguide which is similar to the 2D photonic crystal and also microlens but reported in different terminologies.

3.1. Repeating micropores and the difference of structural dimensions effect on brightness in LED technology

The main interest is to understand the optical properties of mesoporous structure which is an MRS and its effect on luminous flux when deposited on LED chip. The effect of MTF deposition on LED light output had been proven by Peng [36] by growing OLED structures on the nanoporous anodic aluminium oxide. It was reported that the coupling efficiency was improved by around 50% with this method. Coupling efficiency is inhibited by TIR and basically the modification method needs to promote forward scattering. Choi [38] used perforated WO_3 and was able to increase the OLED external quantum efficiency by 39.3% and observed a decrease in operating voltage. It is possible that these modification on LED will improve the LEE as compared to OLED as stated by Jeong [40].

Therefore, to prove that MRS is able to improve LEE, structure and dimensions of the MRS from different researchers are summarized into Table 1. The data in Table 1 is analyzed through Minitab software by using a general regression method. Data with ranged values are averaged such as the one extracted from Do [35] and Ziebarth [4]. A general regression plot was performed as observed in Fig. 5. The regression shows an 80.07% R-squared value which is a good linear fit. The regression is then

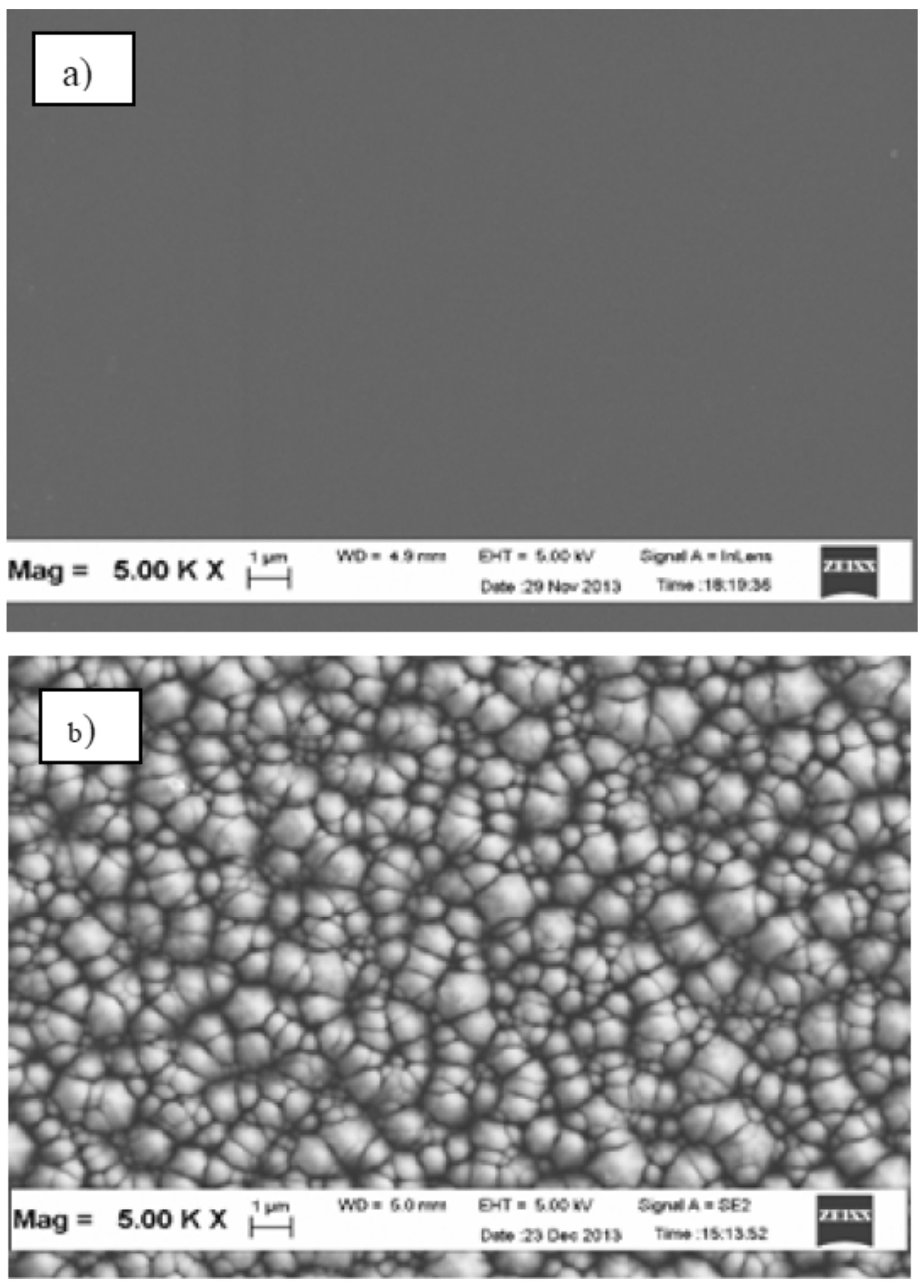

Fig. 4. SEM images of LED chips. Mirror polished wafer is shown in Fig. 4a and a roughened wafer in Fig. 4b.

simplified into an equation which is shown in Eq.(1). With the regression equation, an estimation of the dimensions of the MRS's structure height and diameter could be calculated for optimum light extraction at 80% *R*-squared. It is stated in the equation that the height of the structure plays a significant role in improving brightness with a factor of 19.0362. The second contributing factor at 2.7217 for brightness improvement is the diameter of the structure. Finally, it could be observed that there is an interaction between the height and diameter in the equation at a factor of -2.5707. Since the regression is only at 80% R-squared, Eq.(1) will only be true at 80% confidence level. In order to improve the regression, probably more data points are required. The regression in Eq.(1) could be used as a quick prediction of the structure's dimension on brightness improvement using the height and diameter as the parameters.

$$\begin{aligned}&\text{Brightness Improvement, \%} = -54.1901 + \\ &19.0362\ \text{Height} + 2.72174\ \text{Diameter} + \\ &0.184699\ \text{Wavelength} - \\ &2.5705\ \text{Height*Diameter}.\end{aligned} \qquad (1)$$

The experiments in Table 1 show a model where diffraction through MRS to improve LEE of an LED is true. It may be concluded that any micro-structure deposited, fabricated or etched on an LED chip either in repeating semi-sphere (lens shape), rod, pyramid or random structures (Surface-roughening) may improve the overall LEE.

3.2. Surfactant template mesoporous silica film properties and its potential in optoelectronic materials

Silica, alumina, and indium phosphide (InP) are materials that could produce high quality pores [48]. Mesoporous Silica is a better material due to its promising characteristics in the growth of silicon-based optoelectronic devices [49]. It was reported that since mesoporous silica thin films (MSTF) fabricated through surfactant templates have elastic modulii conforming to IC processing [50] thus opto-

Table 1. Dimensions of the MRS and its effect on brightness improvement on LED.

Height, nm	Diameter, nm	Period[a], nm	Wavelength, nm	Brightness Improvement, %	Journal
3000	10000	Na	560	55	[45,46]
50-400[b]	150-500[b]	300-700[b]	530	50	[35]
7000	8000	16000	All Visible[d]	70	[1]
40-60	≈500[c]	375	640	70	[4]
≈40[c]	196	350	530	46	[44]
5000	5000	10000	470	73	[6]
≈1000[c]	≈2000[c]	Na	465	60	[47]
50	255	330	550	39.3	[38]

[a] Period is the lattice constant of the MRS
[b] Ranged values
[c] An approximation of the dimensions
[d] Visible light spectrum

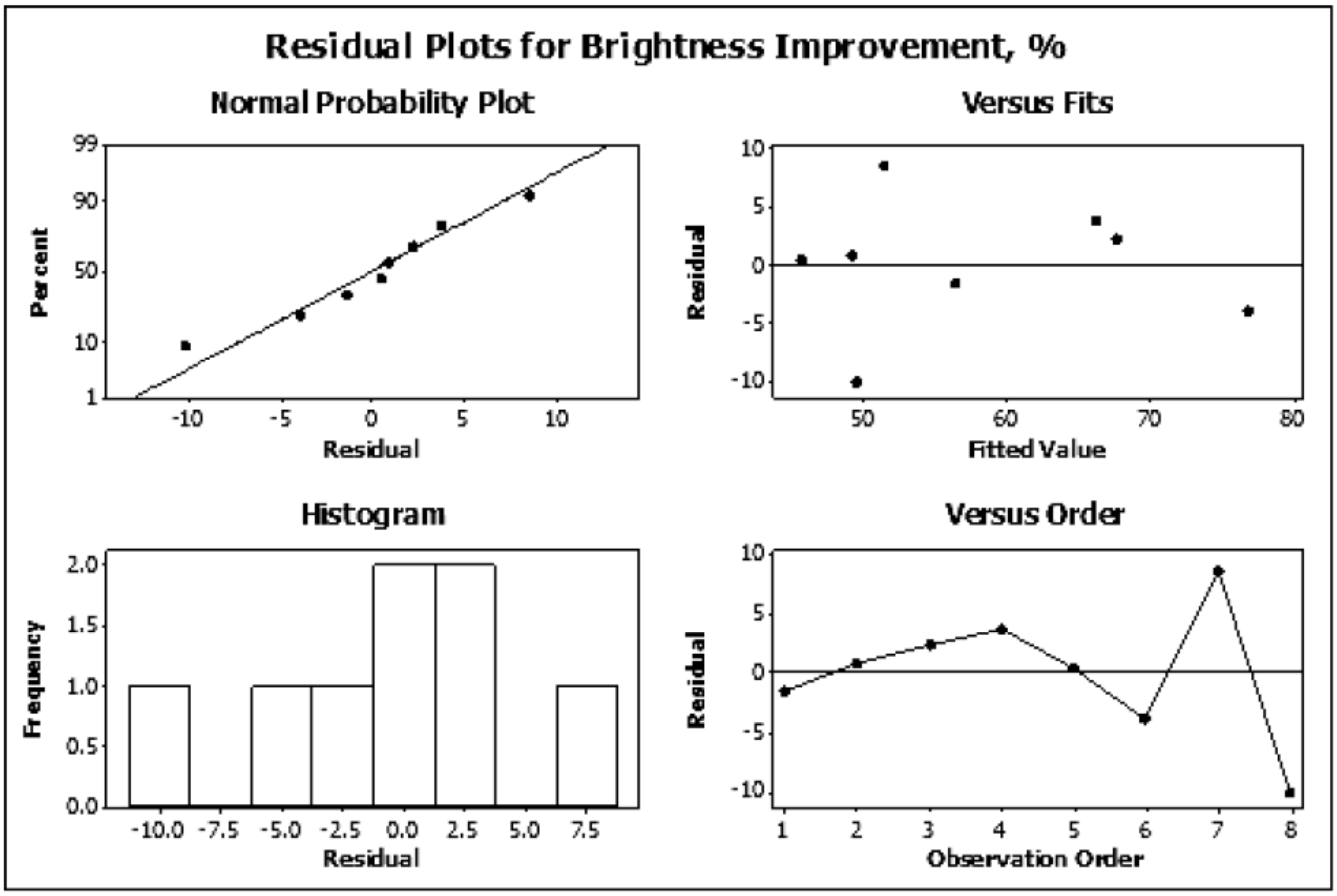

Fig. 5. Graph of residual plot for Brightness vs Height, Diameter and Wavelength. The graph shows a good fit for a linear model.

electronic applications could utilize this characteristic for further study as it will not stress the package due to the minimal difference in coefficient of thermal expansion(CTE). This is because the chip passivation layers are fabricated from silicon dioxide. The porous silica is also able to exhibit photoluminescence properties at room temperature [51] which opens up applications for optoelectronics. The MSTF is mostly reported to be transparent [16,17], uniform and the thickness of the film ranges from 300 nm to a few hundred μm [16].

The porosity is mostly attributed through surfactant template growth [52]. Many researchers uses surfactant template growth. For example Yeh [53] utilizes ternary surfactant and silica condensation, Johansson [54] produced large 18 nm mesopores through P123 surfactant, Ko [19] fabricated mesoporous titania film with vertical pores through F-127 surfactant in 1-butanol solution and Kemmochi [55] uses F127 surfactant to produce silica mesopores as a template for growth of carbon nanotubes. There are many reports on the use of surfactant template method for fabricating ordered mesoporous silica. However, there is no actual application reported yet.

Surfactant template produces mesoporous structure and in order to make it into thin film layer, the process is normally based upon a surfactant template solution which is then spin coated [19], casted [56], EISA method after being dipped or spin-coated respectively [57,58], Sol-gel method [52], multi-step oxidation-etching process [59] and sputter deposited film through RF magnetron sputtering [60,61]. The mesoporous material pore alignment could also be controlled to be vertical to the deposited plane. Richman [62] reported that a method for vertical pores is through coating the substrate surface with a neutral material which does not have any affinity towards any of the surfactant blocks. It is also important for the reaction to be ideally slow in order for the assembly to take place. Thus a lower pH is favored for slow reaction and ideally around pH 2 to 3 for regular and homogeneous films [53,62]. Yeh [53] reported that at pH 6, the mesoporous sheets become irregular and not controllable.

Besides, MSTF is a versatile material due to the capability of having its adjustable pore structure and size, controllable amount of pores, adjustable surface chemistry [14,24,50,63], excellent stability [64] and having well-organized pores [65]. It has the flexibility of having its pore structure engineered thus the dielectric constant of the material could be adjusted. This is due to the presence of air causing the material to exhibit high insulating properties [27]

Table 2. Packing factor on the change of the surfactant mesophase.

Packing factor (*g*)	Mesophase
<1/3	Cubic (Pm3n)
1/3 < g < 1/2	Hexagonal (p6m)
1/2 <g < 2/3	Cubic (Ia3d)
1	Lamellar

Note: The increase in surfactant concentration represents higher packing factor value

and having its refractive index tunable [51]. The tunable refractive index in certain cases was tuned to a lower refractive index to enable it to be used as an optical cladding for waveguides [33].

The pore characteristics, refractive index and arrangement could be engineered depending on its application. There are many ways of engineering mesopores such as controlling the surfactant concentration [13], temperature, solution pH condition [53] and composition of reactants [66]. Surfactant concentration could affect mesoporous material packing factor which could be represented by the equation below:

$$g = \frac{V}{a_0 l}, \qquad (2)$$

g is the packing factor, *V* is the overall volume of the surfactant chains with co-solvent or organic additive between the chains, a_0 is the effective head group area of the micelles surface and *l* is the kinetic surfactant tail length. The packing factor value of the surfactant is represented in Table 2 [13,67]

To elaborate more, a binary phase diagram in Fig. 6 [68] shows the effect of increasing surfactant concentration and the changes in surfactant agglomeration characteristics. From the diagram, when concentration of surfactant is low the surfactant appears as free molecule chains. At increased concentration CMC1, critical micelle concentration is reached where surfactant forms sphere like agglomerates. At CMC2, the surfactant forms into cylindrical micelles. Further increment of surfactant concentration to 40% induces the cylindrical micelles to form hexagonal close packed liquid crystal. At around 80% surfactant concentration, lamellar structures are formed [68]. In conclusion, different surfactant concentration could engineer the mesoporous material to have different pore geometry.

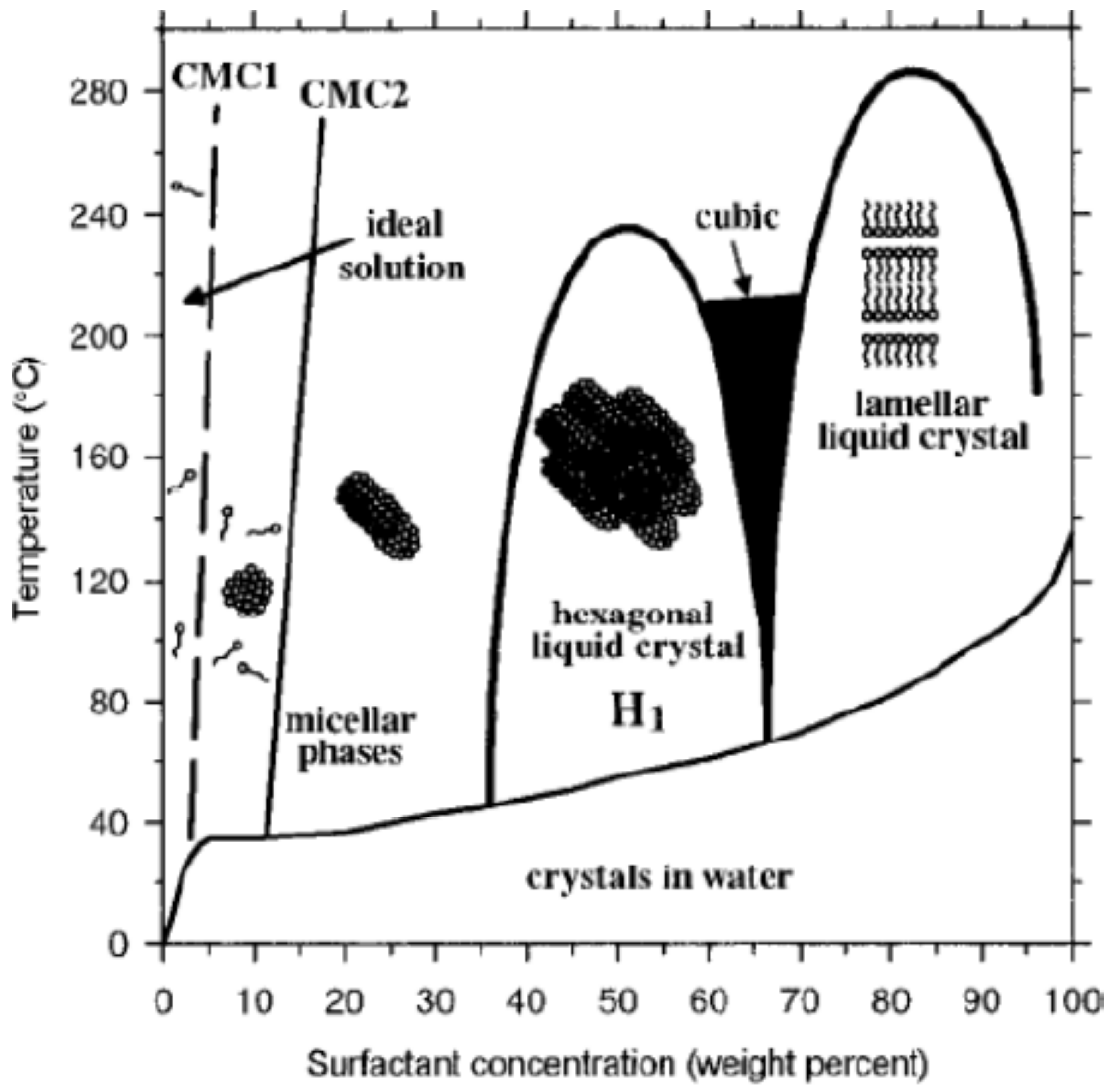

Fig. 6. A phase diagram of Cetyltrimethylammonium Bromide(CTAB) concentration and the change in mesophases. Reprinted with permission from N.K. Raman, M.T. Anderson, C.J. Brinker // Chem. Mater. 8 (1996) 1682, (c) 1996 American Chemical Society.

Other than that, the crystal structure could be engineered through variations of the spin-coating speed reported by Pan [69]. The mesoporous films are also reported to be reproducible on different surfaces with different fabrication method (dip coating or spin coating).On the other hand, Bandyopadhyaya [66] reported the geometry remains whereas there will be changes in the pore orientation on different surfaces. Nevertheless, Pan and Bandyopadhyaya above contradict each other in the sense that Pan [69] reported that variation of spin-coating speed changes crystal structures whereas Bandyopadhyaya [66] reported mesoporous films are reproducible with different fabrication methods. Based on the review, Bandyopadhyaya mentioned the experiment was just conducted at one spin-coating speed. There is no comparison between different speeds thus there is not enough evidence to prove the experiment is able to achieve a similar crystal structure on a different surface and through a different fabrication method.

The engineered mesopores of different pore sizes between samples are reported to not be affecting the mechanical properties for mesopores of the same material. Chemin [70] showed the mechanical properties are affected by density and the morphology. On the optical properties, Hutchinson [71] reported the refractive and absorption indices of 2D hexagonal mesoporous films depended on the porosity and pore shapes. As stated by Cheyssac [27], one of the optical properties of mesoporous silica is that it is a high scatterer of light. However, the exact characteristics of the scattering were not defined. In his other paper, Cheyssac [14] reported an angular distribution of the scattering characteristics by the mesoporous film shows an interference pattern.

4. CONCLUSION

It is discernible after reviewing many reports, that MRS is able to improve LEE from a light source. The MRS must be in the form of alternating silica and air arrays. The alternating silica shape could be any forms ranging from pyramids, cones, pillars or random shapes. It must also be perpendicular to the surface of the light source. To supplement the claim above whereby MRS could improve LEE, a simple regression of the MRS dimensions was plotted out showing an 80% R-squared which is a good fit. As such, we could conclude that MTF which is an MRS is possible to improve LEE.

ACKNOWLEDGEMENTS

A part of this study was supported by University Sains Malaysia (USM), Ministry of Education (MOE), Ministry of Science, Technology and Innovation (MOSTI), Osram Opto Semiconductors (Malaysia) Sdn Bhd & AUN/Seed-Net.

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