

# Plastic Materials for Modifying the Refractive Index of Contact Lens: Overview

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## Abstract

Contact lenses (CLs) are produced from optical polymers which recommended by experts to treat the refractive errors. Researchers are looking for ideal materials in order to improve the CLs optical qualities. This investigation gathers polymeric materials merits to choose the desired pure or doped polymer for the purpose of CLs manufacturing.

**Methods:** high performance CLs can be made of polymers with high transparency nanocomposite because of the need for optical plastic merits such as light weight, transparency, Biocompatibility, the ability to modify its refractive index and abbe number.

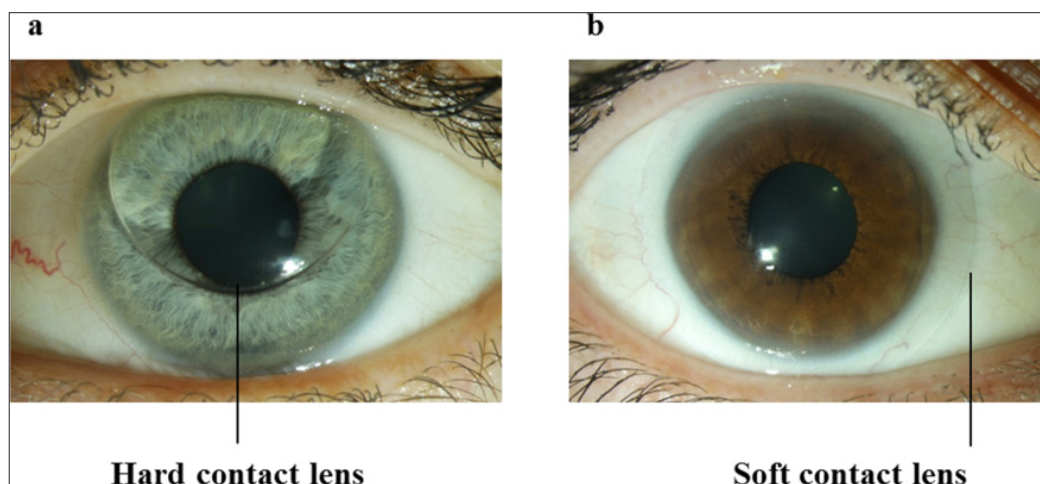
**Results:** TiO<sub>2</sub>-PMMA and TiO<sub>2</sub>-PHEMA nanocomposites can be considered the best candidate polymers in CLs fabrication due to its optical properties where it possesses RI higher than 1.52, vd of 30-54, transmittance higher than 92% and its flexibility and biocompatibility to the human eye tissues.

**Conclusion:** this article demonstrates the advancement of polymer properties utilized in optical applications focusing on RIs, vd and visible light transmittance of the optical plastics that can be utilized in the CLs manufacture or evolution of them.

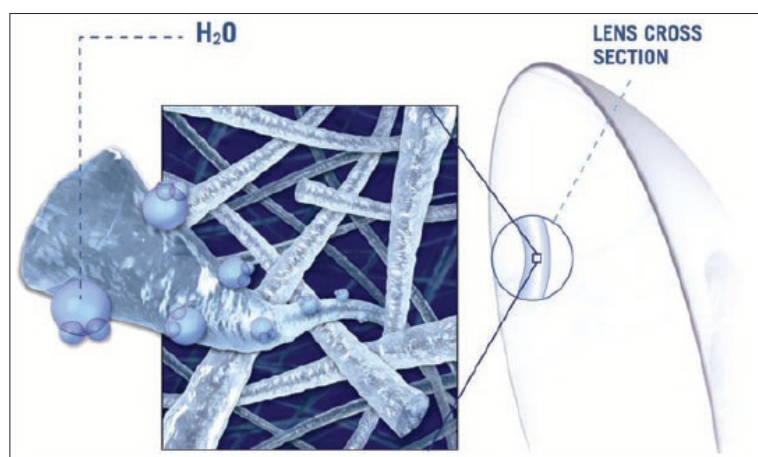
**Keywords:** Contact lens; Refractive index; Abbe number; Hybrid materials; Doped polymer

## Introduction

The first material used in contact lenses CLs fabrication was heavy, non-wettable, rough and uncomfortable when wearing them and don't allow the oxygen to through to the cornea, so researchers resorted to employ plastic materials where they started with polymethylmethacrylate (PMMA) that had replaced by another polymer nano-composites because of unsuitability to wear because it prevent water and oxygen to transmit [1]. Scleral CLs were described by Feinbloom who proposed a scleral CL made of a glass part surrounded by a circular polymeric portion [2] which was developed and amended then by Tuohy [3]. These lenses materials were replaced by optical polymer due to their rough surface and impermeability to water or gases [4]. Several studies have discussed the bioavailable materials and the CLs brand properties [5,6]. Four main types of CLs are available for both spherical and cylindrical refractive error corrections rigid (hard), soft (hydrogel), rigid gas permeable and hybrid CLs. Rigid CLs made of PMMA [7,8] of RI: vd 1.49:59, light weight, high transparency, high resistant to scratching, a small size CLs and safe to the eye but it does not allow the permeability of water and oxygen (Dk) [9]. In contrast soft lenses are larger size than rigid lenses as illustrated in Figure 1 and made up of polymers that are capable of absorbing water (Figure 2) such as hydroxy-ethylmethacrylate (HEMA). Soft CLs have been patented by Wichterle [10,11], they are more comfortable for longer periods of usage [12,13]. CLs made of poly(2-hydroxyethyl) methacrylate (pHEMA) provides great hydrophilicity, but the oxygen permeability is not great [10,14]. Rigid gas permeable CLs (RGP) were introduced firstly in 1978. Made of material with molecules that allow the oxygen and carbon dioxide to transmit [15]. It was made up of a silicone-incorporated monomer to increase Dk. This process worked well where Chemists have developed these lenses by increasing the proportion of silicon content to increase the surface wettability and lens Dk [16]. Due to RGP CL rigidity, it causes corneal reshaping, which was useful for some post-surgery curing [17,18]. Another type of CLs namely hybrid CLs (gas permeable and HEMA copolymer composition) [19], they are designed in such a way that the central zone made of material of high oxygen permeability surrounded by hydrogel area [20].



**Figure 1:** Difference in hard and soft CLs size (a) Hard CL, (b) Soft CL [15].



**Figure 2:** Continuously wetting the material by siloxane molecules which bond and attract the surrounding water.

### Optical polymers used to improve the contact lenses optical properties

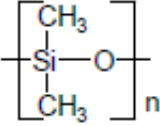
Investigation of optical polymers been the subject of different excellent review papers [21]. In the following text we discuss the development and improvement of polymers used in optical applications in terms of permeability, refraction and dispersion properties. A group of researchers has reached several types of optical plastics [22,23] that possess the basic optical characteristics such as RI, vd, light reflectance and transmittance; these polymers have (1.6-1.8) RI; they are Poly(arylene sulfide)s (PAS) (1.66-1.72) [24], transparent thin film made of  $\text{TiO}_2$  and poly(bisphenol-A and epichlorohydrin) (1.58-1.81) RI [25], polyimids derived from paradihydride, 4,40-[p-sulfonylbis-(phenylenesulfanyl)]-diphthalic anhydride (p-DPSDA) (1.7169 to 1.7420) [26], Polythiourethane (1.66-1.68), Polynaphthalen (1.68), Polyether ketone (1.65) and Polyvinyl carbazole (1.68) [27].

**Bis(mercaptomethyl)-dithiane (BMMD):** In 1997, Tsuyoshi Okubo and coworkers based on polyaddition and polymerization of (bis(mercaptomethyl)-dithiane) (BMMD) with diisocyanates and 2,5-bis (2-thia-3-butenyl)-1,4-dithiane (TBD) in bulk,

respectively to obtain a composition of (BMMD) based poly (Salkylcarbamate)s and poly(TBD) have (1.598-1.67) RI range value and vd range of (38.2-34.8) [28]. A developed thermoplastics made of Poly(thioester)s (PTE)s with sulfide and alicyclic series when it synthesized by (BMMD) with 1,3-adaman-tanedicarbonyl dichloride (ADC) to prepare Poly(BMMD/ADC) or cyclohexanedicarbonyl dichloride (CDC) to prepare poly(BMMD/ CDC); the polymer obtained with (RI:vd) of (1.6149:32.1) and (1.6217:30.0), respectively in the vision region [29].

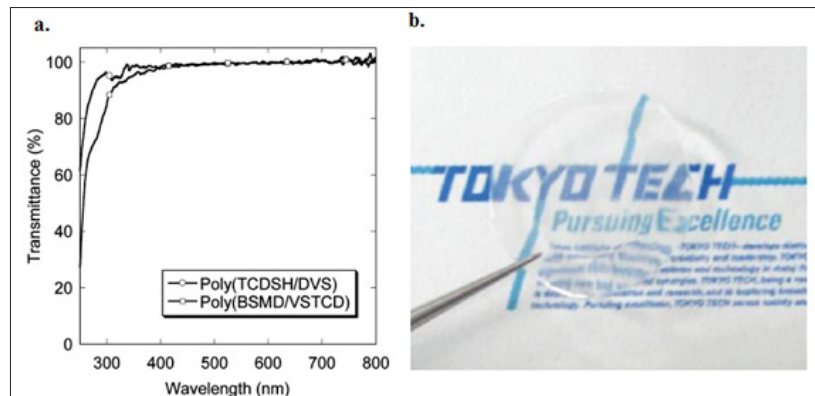
**Silicone based polymers:** Graded-RI AR coating can improve the optical properties of The used polymer as Ki-Chul [30] suggestion; three thin layers  $\text{SiO}_2$ ,  $\text{MgF}_2$ , and ITO were applied to form a polymer used in ophthalmic lenses; the resultant thin film exhibited (reflectance: lens RI) of (2.6%:1.60) and (2.7%:1.67) when using a of polymeric lens [30]. Polydimethylsiloxane (PDMS) (Table 1) of RI (1.375-1.404) and high gas transition has been used in hydrogel CLs that made of silicone as a main material [31]. Two drawbacks of (PDMS) based CLs; they are the high affinity for lipids and this type of lenses is non-wettable which leads to the patient's discomfort when wearing them in terms of vision and comfort [32,33].

**Table 1:** Chemical properties of high Dk PDMS.

Common Name	Polymer Abbrev.	Structure	Chemical Formula	Reference
Polydimethyl-siloxane	PDMS		$(C_2H_6OSi)_n$	[33]

The development of silicone hydrogels (long chains of Si derivatives, as seen in Figure 3) has been consumed extensively by CLs manufacturers because it has a role in increasing gas permeability through silicone channels up to four times the gas permeability in conventional hydrogel CLs and maintaining the water content of the CLs [34]. Improvement of this kind of lenses continued by Yuwen and coworkers; they have patented a silicone hydrogel CLs made of a polymerizable silicone and hydrophilic monomers

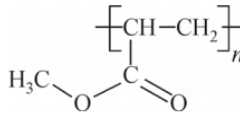
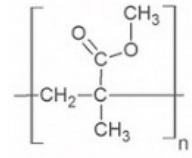
compound [35]. Then Scott and colleagues invented a silicone based high RI rigid gas permeable CL built up from a liquid crystal layer sandwiched between two rigid polymer layers [36]. The two previous CLs patents have an improved gas permeability and are appropriate to the eye tissues. Recently, two CLs made of silicone hydrogel and polyvinylpyrrolidone (PVP) to acquire a smooth water protected CLs valuable for individuals who are experiencing vision issues and working long timespans on PCs [37].

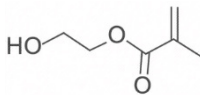
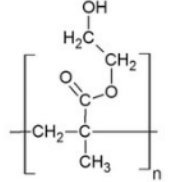
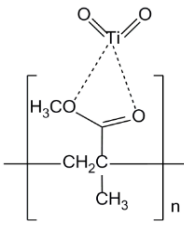
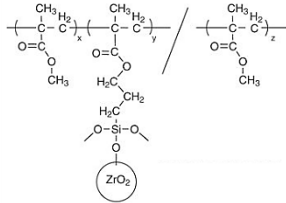
**Figure 3:** a: transmittance (%) of p(BSMD/VSTCD) and p(TCDSH/DVS), b: film of p(BSMD/VSTCD) [62].

**Polymethacrylates:** Include, polymethyl acrylate (PMA), PMMA, HEMA and pHEMA. Hybrid PMMA-dioxide nanocomposites, focusing mostly on PMMA-SiO<sub>2</sub> [38], PMMA-TiO<sub>2</sub> [39,40], PMMA-ZrO<sub>2</sub> [41] (Table 2 & 3). Trialkoxysilane-capped PMMA-titania (PMMA-TiO<sub>2</sub>) hybrid thin film has been prepared with concentration range T0-T100 to obtain RI range of (1.49- 2.010) and vd range of (36.5-14). The results showed that the RI range increased from (1.505 to 1.867) as the tintania concentration increase from 2.9 to 70.7wt% while the vd exhibited an opposite behavior [42]. An organic

polymer PMMA/ZrO<sub>2</sub> prepared by Otsuka [43] to get polymerized material of above 95% visible light transmittance and increment in RI to 1.534 when applying 38.8wt% ZrO<sub>2</sub> concentration, PMMA/ZrO<sub>2</sub> used to improve optical devices merits [43]. Hybrid PMMA-ZrO<sub>2</sub> obtained by sol-gel deposition method resulted highly transparency ~90% and 1.57 RI at 532nm [44]. A great RI boost up to 1.780 obtained by employing nanohybrid material (titania-PMMA) and transparent thin film in visible region that decreased vd to 31.8 when TiO<sub>2</sub> concentration was 40wt% [45,46].

**Table 2:** Chemical structures and formulas of high-RI and high- vd optical polymers used for contact lenses fabrication.

Common Name	Polymer Abbrev.	Chemical Formula	Structure	Reference
Polymethacrylates	PMA	$(C_4H_6O_2)_n$		[53,54]
Polymethyl-methacrylate	PMMA	$(C_5O_2H_8)_n$		[55]

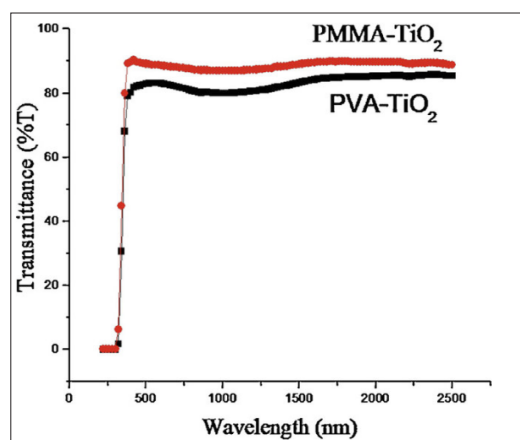
Hydroxy ethyl methacrylate	HEMA	$C_6H_{10}O_3$		[56]
Poly (2-hydroxyethyl methacrylate)	pHEMA	$(C_6H_{10}O_3)_n$		[33]
Titania-PMMA	TiO <sub>2</sub> -PMMA	TiO <sub>2</sub> - $(C_5O_2H_8)_n$		[45,57]
Poly (methyl- methacrylate)/ zirconium oxide	PMMA-ZrO <sub>2</sub>	ZrO <sub>2</sub> - $(C_5O_2H_8)_n$		[43]

**Table 3:** The optical characteristics of high-RI and high-*vd* polymers used in contact lens applications.

Polymer abbrev.	Transmittance (%)	RI	<i>vd</i>
PMA	90	1.4793	60
PMMA	92	1.448-1.50	59
HEMA	97	1.453	-
pHEMA	90	1.5119	54
TiO <sub>2</sub> -PMMA	89	1.780	31.8
ZrO <sub>2</sub> -PMMA	95	1.534	-
TiO <sub>2</sub> -pHEMA	90	1.527	54

TiO<sub>2</sub> NP can rise the polymer RI up to 2 and the transmittance to 90% through the visible light [47]. PVA-TiO<sub>2</sub> and PMMA-TiO<sub>2</sub> nanocomposites exhibited light transmittance between 80 and 90% and 1.6 and 2.3 RI [48] as illustrated in Figure 4. Hendrik [49] have investigated a flexible TiO<sub>2</sub>-pHEMA hydrogel that high transparency in the visible range when it is applied in optical applications, high *vd* of 54 and RI=1.527 with an excellent transparency ~90% transmittance [49,50], this nanostructure is therefore an attractive material in optical application that required high visual performance. PVA hydrogel material was the topic of investigation in the 1990s [51]. In addition, polyethylene glycol (PEG) surface coating has invented by Imafuku to improve the Si-based CLs hydrophilicity [52]. Low cost, high wettability and bioavailability

PVA hydrogel make it relatively important to hydrogel CLs industry compared with silicone and HEMA hydrogels [53-57].

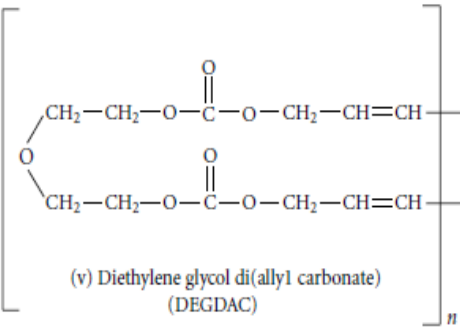


**Figure 4:** Transmittance (%) of PVA- TiO<sub>2</sub> and PMMA-TiO<sub>2</sub> nanocomposite films [48].

**Polycarbonate (PC):** In 2009 The studies concerned to modify the lightweight polycarbonate (PC) lenses (clarity ~90%) from diethylene glycol di(allyl carbonate) (DEGDAC) based (polythiol) (Table 4), the resultant (PC) of improved RI (1.49-1.66) and decreased *vd* range between (26-37) [58]. Five decades ago, polycarbonates were commonly used in spectacle lenses fabrication which holds high scratching resistance, but it is hard to use in CLs, despite the trails that were done to develop them. In 2018, Halah A [59] was tested Allyl carbonate polymer material as CL [59], which is usually known as CR-39. The CR-39 characterized by RI of 1.49 and *vd* of 58-59.

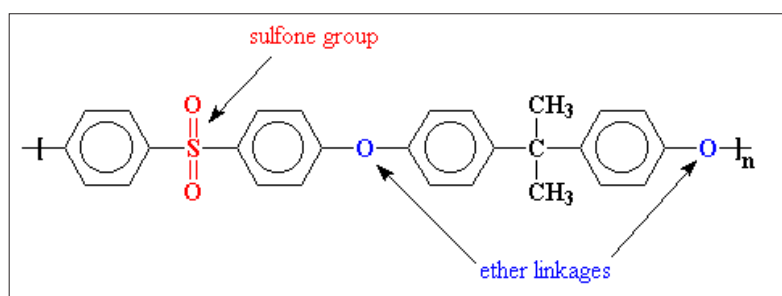


**Table 4:** Chemical structure and formula of DEGDAC.

Common Name	Polymer Abbrev.	Chemical Formula	Structure	Reference
Diethylene glycol di (allyl-carbonate)	DEGDAC	C <sub>12</sub> H <sub>18</sub> O <sub>7</sub>	 <p>(v) Diethylene glycol di(allyl carbonate) (DEGDAC)</p>	[58]

**Sulfur-containing polymers:** Sulfur compounds such as polysulfide, polythiol, episulfide groups are used in CLs fabrication due to their optical properties. Polymers containing a sulfur atom such as 2-(tricyclo-[5,2,1,0<sup>2,6</sup>]-3-decenylthio) ethyl methacrylate (DTEMA) appears high transparency (transmittance 91%) and have a RI and vd of 1.557 and 47.4, respectively. When it converted into a copolymer, the vd value degraded as the RI increased, according to the added polymer and its monomers concentration. When bromine DTEMA/ bromine 2,2-bis(3,5-dibromo-4-methacryloyloxyethoxy-phenyl) propane (Br<sub>2</sub>DTEMA/Br<sub>4</sub>BMEPP) copolymer

is prepared RI, vd and transmittance have reached to 1.608, 30.4 and 90%, respectively [60]. In 2010 by Yasuo [61] and researchers have improved the optical properties of copoly (thioether-sulfone)s (Figure 5) with a polymeric composition of sulfide, sulfone, and alicyclic, the later materials were equipped by copolymerization technique of 2,5-bis(sulfanylmethyl)-1,4-dithiane (BMMD) and cyclohexane-1,4-dithiol (CHDT) with divinyl sulfone (DVS) based on the Michael polyaddition the resultant polymer called Poly (BMMD/CHDT-DVS) had (1.6512-1.6022) RI range and (42.6-50.6) vd range [61].

**Figure 5:** PES get their name because of existence of sulfone groups and ether groups in their back bone chain.

Due to Suzuki and colleagues proposal, a flexible, highly transmittance thermoplastic disulfanyl-tricyclodecane/divinyl sulfone p(TCDSH/DVS) of (n=1.6052 and vd=48.0) and 2,5-bis(sulfanyl-methyl)-1,4-dithiane bis(vinylsulfone) tricyclodecane p(BSMD/VSTCD) of (RI=1.6228 and vd=45.8) [62]. Tapaswi [63] and coworkers have reached a flexible transparent film made of 2,2'-Bis(thiophenyl)-benzidine (BTPB) and 2,2'-bis(4-chlorothio-phenyl)-benzidine monomers (BCTPB) were prepared and synthesized to achieve the aromatic polyimide (BPDA/BCTPB) of (1.7282) RI and transmittivity of ~83% [63]. The gained film useful in lens materials other optical applications required high transparency and flexibility (Figure 3).

Bin [64] were utilized Alumina nanoparticles (NPs) to get low dispersion materials; they prepared 1,3-bis(2-hydroxyethyl-sulfonyl)-2-propanol/2,2'-thiodiglycolic acid/ alumina (BHESP/TDGA/Al) which provided (RI=1.580, vd=55) and 2,2'-thiodiglycolic

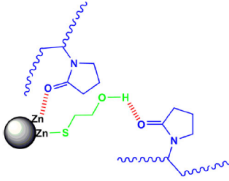
acid/ phthalic acid/ alumina (BHESP/PA/Al) of (RI = 1.631, vd=53) nano-composites [64]. Carbonylsulfide (COS), eugenol-based glycidyl ether (EGE) and guaiacol-based glycidylether (GGE) were used by Zhang and coworkers to prepare the copolymers EGE/COS, GGE/COS, and EGE/ GGE/COS; these polymers appeared high-RI>1.58 and vd range of (32.5 - 40.4) [65]. Tang and a group of researchers have intended a novel high-RI (1.707) and transparent (transmissivity of 89%) polymer named episulfide-thiol has prepared from episulfide synthesis which in turn is derived from 9,9-bis(4-glycidyoxyphenyl)fluorine (BGF) and 4,4-thiodibenzenethiol (TBT) to get the high optically transparent polymer episulfide-thiol [66].

**Zinc based polymers:** According to Sung [67], UV-block hydrogel CLs (78~90% transmittance in visible region) can be produced by inserting Zinc oxide (ZnO) NP in addition to vinyl pyridine to MMA, HEMA and N-vinylpyrrolidone (N-VP) blend

to take out (1.429~1.450) RI polymer from this polymerization process [67]. A group of researchers have come up with hydrogel nano-composite a chain of ZnS/PVP/PDMS (Table 5), (30-60)

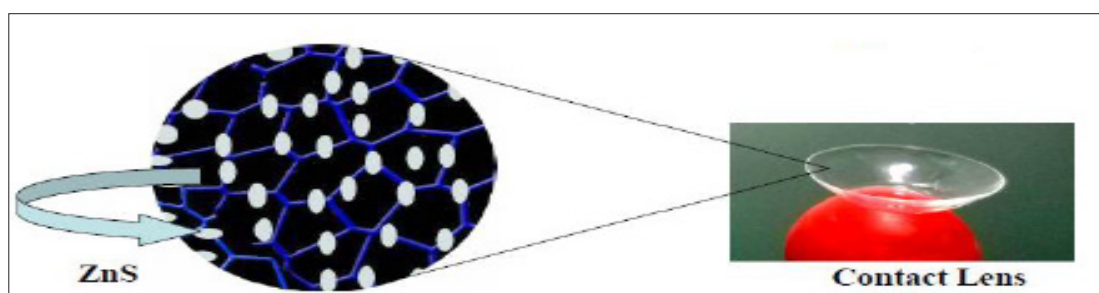
wt% concentration of ZnS NP has exploited to realize a transparent nano-composite, (1.58-1.70) and (1.38-1.46) RI in the dry and hydrated states, respectively [68].

**Table 5:** Chemical characteristics of ZnS/PVP.

Common Name	Polymer Abbrev.	Chemical Formula	Structure	Reference
Zinc sulfide/ poly (N-vinyl- pyrrolidone)	ZnS/PVP	ZnS-(C <sub>6</sub> H <sub>9</sub> NO) <sub>n</sub>		[75]

Incorporation of PVP and moisturizing agents into Soft CLs has changed the surface wettability. These agents may result increased comfort and enhanced the stability of tear film [69,70]. Zinc also used extensively to prepare NP antibacterial coatings applied on CLs surfaces [71], also the hybrid ZnO/PMMA polymer provides a colorless film of (1.6) RI which can be considered useful in such applications [72]. ZnS NPs have employed to obtain polymerized nano-composite grafted with ZnS, where a copolymerization process with a mixture of N,N-dimethylacrylamide (DMA) and

HEMA (Table 5) or glycidyl methacrylate (GMA) or methyl methacrylate (MMA), the produced polymer exhibits a transmittance above 92% in visible region and RI range of (1.652 and 1.751) after changing ZnS concentration from 33.4 to 50.3wt% [73]. Then Peili [74] has suggested a ZnS-inorganic hydrogel CLs (Figure 6) by copolymerizing ZnS-polymerizable group with HEMA monomer to get RI with a range of (1.38 -1.45) as the ZnS concentration varied from (30 to 60)wt% [74,75].

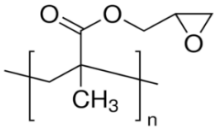


**Figure 6:** ZnS-inorganic hydrogel CL [74].

**Poly (Glycidyl Methacrylate) (PGMA):** High RI can be achieved by Poly(glycidyl methacrylate) (PGMA) (Table 6) modification as Huinan [76] proposal; this is done by using a PGMA's selenide chain, The concentration of selenium has been varied by a certain percentages, the resultant polymer has RI of (1.719) and  $v_d$  decreased to (11.3) [76]. Optical Polymers helped researchers to improve CLs performance as Jung [77] and his colleague did for PDMS-CLs [77]. They have developed it for the purpose of

preventing its adsorption of protein by submerging the lens in deby poly(ethylene glycol) meth-acrylate (PEGMA) [78]. PEGMA is a low-cost polymer and it is easy to incorporate with other chemical components. Industrialization aspects need to be considered to achieve the required configurations in a new CLs generation, where, increasing PEGMA concentration in silicone hydrogels affected the lens properties such as increasing brittleness above 20%.

**Table 6:** PGMA chemical structure and formula.

Common Name	Polymer Abbrev.	Chemical Formula	Structure	Reference
Poly (glycidyl methacrylate)	PGMA	(C <sub>7</sub> H <sub>10</sub> O <sub>3</sub> ) <sub>n</sub>		[79]

**Polymers based on phosphorus:** In order to rise up the RI of phosphorus compounds, Phosphonic dichlorides and diols used in synthesis with polyphosphonate; the gained polymers characterized by high optical transparency, high RI (1.60-1.65) and vd of (22.4-29.9) [79]. Ryoyu [80] have developed novel high RI materials (1.626-1.687) and vd (21.3–27.5) consisting of poly(thiophosphonate)s, which were synthesized by polycondensation of phenylthio-phosphonic dichloride with various bisphenols [80].

## Conclusion

Initially at least, rigid CLs are providing much less comfort than soft lenses. There were several attempts to use RGP CLs for extended wear. The success of RGP CLs due to their rigidity, causing corneal reshaping, which was beneficial for some post-surgery treatment. The main contraindications for both hard and soft CLs are the allergy and dry eye. Selecting a suitable Polymers and material types that can be used in CL manufacture must be characterized by specific optical properties such as oxygen transmissivity, adaptability with human eye tissues, high RI and high vd gas permeability, high transparency within the visible spectrum and scratching resistance to get the desired image quality when wearing them. Doping with TiO<sub>2</sub> NPs satisfies the requirements that must be met in the desired plastic polymer such as TiO<sub>2</sub>-PMMA and TiO<sub>2</sub>-PHEMA optical polymers to improve the CLs optical quality.

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