

Organic electro-optic polymer materials and organic-based hybrid electro-optic modulators

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Abstract: High performance electro-optic modulator, as the key device of integrated ultra-wideband optical systems, have become the focus of research. Meanwhile, the organic-based hybrid electro-optic modulators, which make full use of the advantages of organic electro-optic (OEO) materials (e.g. high electro-optic coefficient, fast response speed, high bandwidth, easy processing/integration and low cost) have attracted considerable attention. In this paper, we introduce a series of high-performance OEO materials that exhibit good properties in electro-optic activity and thermal stability. In addition, the recent progress of organic-based hybrid electro-optic devices is reviewed, including photonic crystal-organic hybrid (PCOH), silicon-organic hybrid (SOH) and plasmonic-organic hybrid (POH) modulators. A high-performance integrated optical platform based on OEO materials is a promising solution for growing high speeds and low power consumption in compact sizes.

Key words: organic electro-optic materials; modulator; organic-based hybrid modulator; heterogeneous integration

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1. Introduction

Electro-optic modulators (EOMs) convert signals from the electrical to the optical domain, which are at the heart of high-capacity optical communication, broadband microwave photonics, sensing, and quantum technologies^[1–4]. To fulfill the requirements of booming information technologies, such as big data, internet of things (IoT), cloud-based services, and autonomous vehicles, as well as the artificial intelligence/machine learning (AI/ML), EOMs should be able to chip-scale integrate with electronics at high-density^[1, 3]. However, EOMs have long suffered from the substantial size mismatch with electronic circuits together with issues of energy efficiency, bandwidth, optical loss, drive voltage, and signal distortion. Tremendous efforts have been made in the past few decades to optimize the performance of EOMs and improve the integration with electronics^[1, 5]. Recent advances of silicon photonics provide dense integration of complex photonic functionalities, using mature technology and process toolsets from complementary metal–oxide–semiconductor (CMOS) electronics^[6–8]. Due to lack of intrinsic second-order non-linearity, silicon photonics rely on the plasma dispersion effect that derives from carrier injection by application of a voltage^[4, 9]. This plasma perturbs the transmission of light and thus effects modulation. Unlike Pockels modulators, plasma dispersion modulators do not have a linear dependence of voltage and refractive index change Δn (thus, they show chirp). Most recently, various heterogeneous approaches that combine Pockels-type electro-optic (EO) materials with dielectric or plas-

monic waveguide structures have been explored to introduce better solutions for silicon devices^[10–12].

Currently used materials for modulators include thermo-optic materials^[13], III–V semiconductor (electrical absorption materials, e.g., GaAs, InP)^[14], inorganic crystal materials (lithium niobite)^[15], liquid crystals (LC) and organic electro-optic (OEO) materials. The process of refractive index change for thermo-optic materials and LCs is relatively slow and they cannot be used in high-speed EO modulation. Crystalline inorganic materials, which generally have low optical loss and good stability, are widely used for a long time in fiber optic networks, especially the thin-film lithium niobite on insulator (LNOI) shows the good performance^[16, 17]. However, due to the relatively low EO activity, the EO modulators based on lithium niobate have long propagation length and large footprints, which might be difficult to integrate with high-density. III–V semiconductors modulate light via changing the wavelength of an optical resonance caused by an applied electric field. During electro-absorption modulation, both index of refraction and absorbance change, which perhaps leads to the unwanted phenomenon of “chirp”, altering the signal waveform together with high optical loss near the half-maximum of absorption.

OEO materials are of great interest because they offer high EO coefficients, fast response speed, and extremely high bandwidth^[9, 18–27]. The Pockels effect of OEO materials comes from the conjugated π -electron system of nonlinear optical (NLO) molecules, which has a few femtoseconds phase relaxation time and enables the theoretical bandwidth more than 15 THz. Significant improvements in device performance have been made by silicon-organic hybrid (SOH)^[28–37] and plasmonic-organic hybrid (POH)^[11, 38–44] Pockels effect modulators, which have been proven to be effective photonic plat-

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forms for both analog and digital applications. OEO materials generally have larger EO coefficients (300–500 pm/V), and have a potential to achieve higher EO coefficients (>1000 pm/V) based on the theoretical predictions^[3, 9, 45]. The large EO coefficients are critical for low insertion loss, best energy efficiency, and the highest bandwidth operation. The EO coefficient r_{33} of OEO materials is proportional to combination of chromophore hyperpolarizability (β), electric field poling-induced acentric order of the chromophores ($\langle \cos^3\theta \rangle$), and chromophore number density (ρ_N). The half-wave voltage-length parameter $V_{\pi}L$ for an EO modulator is inversely proportional to n^3r_{33} . The past two decades of research and development have demonstrated many effective molecular engineering strategies, such as side-chain engineering, self-assembly, multi-chromophore dendrimers, site-isolation, chromophore blending, and control of chromophore shape, to increase $\rho_N\langle \cos^3\theta \rangle$ (increasing ρ_N by eliminating the inactive polymer host, while maintaining good poling efficiency)^[46–59]. State-of-the-art POH modulators have already demonstrated ultra-high electro-optical bandwidths (> 500 GHz)^[41], small device footprints < 10 μm^2 , energy efficiencies ≤ 70 aJ/bit^[31, 42], and voltage-length parameters ($V_{\pi}L$) of ≤ 50 V $\cdot\mu\text{m}$ with existing high-performance OEO materials^[12, 60]. To continually shrink the $V_{\pi}L$ values, POH and SOH modulators require novel device designs and engineering as well as improved EO activity.

This paper reviews the recent advances in OEO materials and organic-based hybrid EO modulators. To get groundbreaking performance, it is necessary to carry out synergistic innovation from rational design of OEO materials to device engineering and advancements in communication systems. Theory-guided design has been proven to be a very effective and highly efficient strategy to get high-performance chromophores. The combination of experimental and theoretical studies affords better understanding of structure-property relationships, as well as the prospects for implementation in nanoscale photonic devices. The theory-guided advances in OEO materials greatly promote the development of SOH and POH devices. In summary, the design paradigms for both materials and devices have changed dramatically over the past several years, and this review summarizes and discusses the importance of these changes.

2. Organic electro-optic (OEO) materials

Organic-EO materials are generally composed of two parts: the first part is the polymer that provides the substrate, which determines the refractive index and thermal properties of the material, the second part is the active chromophore that performs the EO conversion function^[5].

The influencing factors of the EO performance is as shown in Eq. (1):

$$r_{33} = 2\beta(\omega, \epsilon)\rho_N\langle \cos^3\theta \rangle g(\omega, n), \quad (1)$$

where $g(\omega, n)$ is the modified Lorentz–Onsager factor taking into account the dielectric properties of the environment surrounding the chromophores. n is the refractive index of the material at the operational wavelength.

To activate the macroscopic EO effect, the organic chromophores need to be poled in the polymer network by heating them close to their glass-transition temperatures T_g to in-

duce the alignment while applying an external electric field. An active push-pull chromophore contains a p-conjugated bridge which is end-capped with a donor and an acceptor (D- π -A). To optimize the molecular first hyperpolarizability β of the EO chromophores, the electron density distribution needs to be fine-tuned through chemical modification of molecular constituents. The approaches for improving molecular β generally include using stronger donor/acceptor combination, extending conjugating system, or low-aromatic π -bridge, and so on. With the enhancement of molecular β , however, the molecular dipole moments rapidly increase, induce the strong anti-parallel aggregation between chromophores and make it very difficult to get acentric order under electric field poling. The chromophores with high- β need to be well-engineered with enough steric hindrance to inhibit unwanted aggregations. There are many structure-engineering strategies that have been proved effective to isolate the chromophores with each other, such as side-chain engineering, dendritic structure, and so on. Our group designed a series of high-performance chromophores with stronger electron-donors that also serve as the bulky groups to isolate with each other, as shown in Fig. 1.

Furthermore, 1,1,7,7-tetramethyl-julolidine was proved to provide not only the strong electron-donating ability but also the excellent steric hindrance in our group in Fig. 1 (Chromophore representative: WJ1, A and A2)^[21, 22, 24, 51, 52, 59]. WJ1 was synthesized using a facile route to get a high yield (36%)^[52]. The conjugating structure of the chromophore was protected by the tetramethyl groups, thiophene ring and chlorohexyloxy group, of which the steric effect and electron-donating ability enhance the molecular nonlinearity. Due to the greatly weakened dipole-dipole interactions, WJ1 can be doped in amorphous polycarbonate (APC) at a higher concentration (40 wt%) with no terrible dipole-dipole aggregations compared with most of the reported guest-host EO polymers. The best r_{33} value of electric-poled WJ1/APC films is 337 pm/V, which is comparable with some outstanding materials. CLD- and FTC-types chromophores were also synthesized using 1,1,7,7-tetramethyl-julolidine as the donor. FTC-type chromophore A2 showed a relatively high r_{33} value of 223 pm/V in APC films by 25 wt%^[21].

The chromophores using 4-(diarylamino)phenyl as donor have demonstrated significantly enhanced thermo- and photostability compared to their 4-(dialkylamino)phenyl analogues^[61]. Due to the weak electron-donating ability and strong π - π stacking interactions, however, these chromophores have poor performance in achieving large r_{33} in poled films. The r_{33} values derived from these chromophores are lower than lithium niobate. Dalton's group and Jen's group have designed many chromophores with stronger 4-(diarylamino)phenyl donors by introducing alkoxy groups into donor, or replacing the phenyl-ring by heteroaromatic rings that could increase donor strength and reduce aromaticity and ionization potential^[61, 62]. We designed a new electron-donor bis-(4-diarylamino)phenyl group based on bis-(4-alkoxyphenyl)phenyl group by replacing alkoxy groups with alkylamino groups. The bis-(4-alkylamino)phenyl-amino donor exhibits stronger electron-donating ability than bis-(4-alkoxyphenyl)phenyl-amino donors. In Fig. 1, the chromophore T3 showed the best r_{33} value of 95 pm/V which was 1.6-fold of

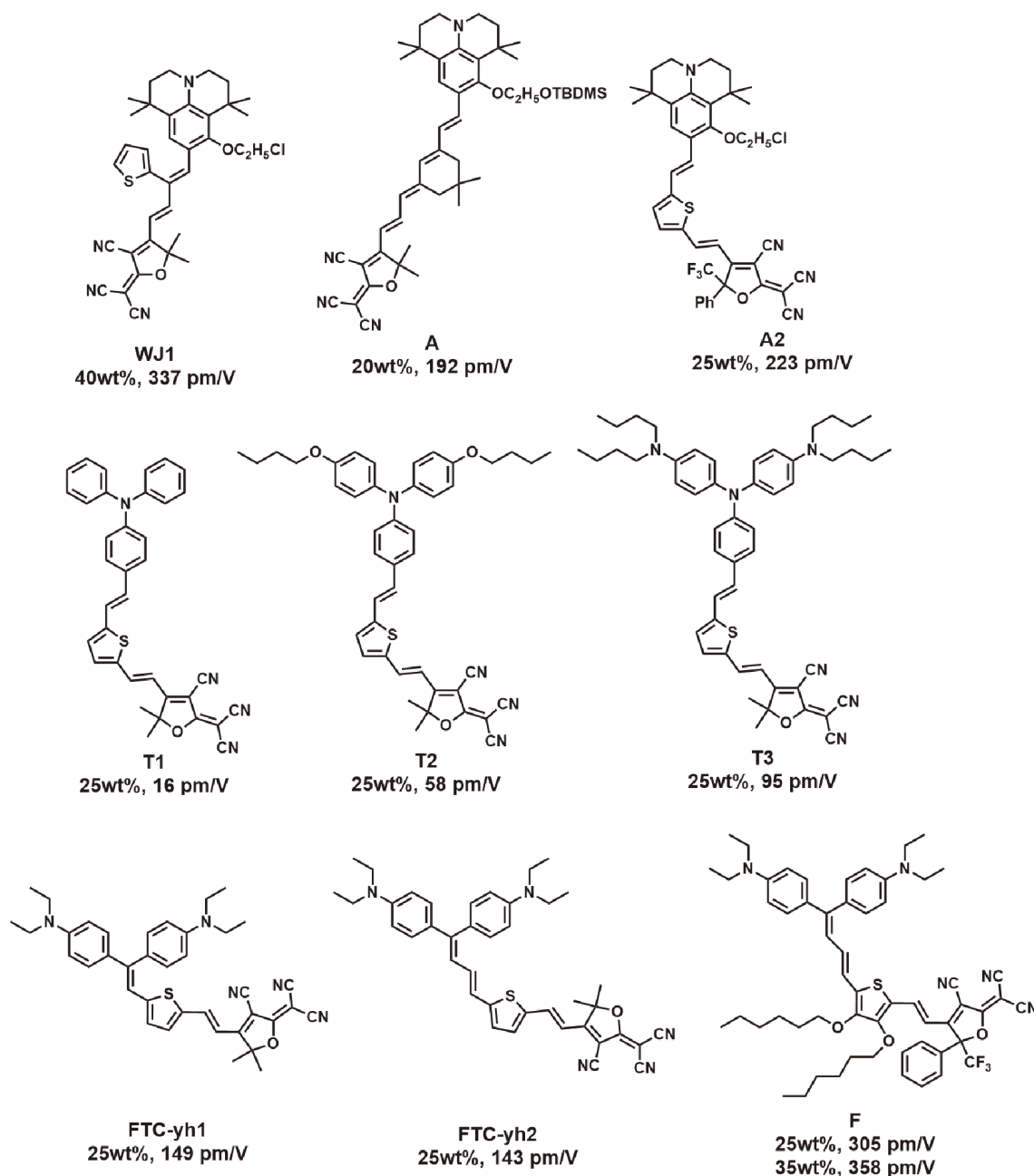


Fig. 1. EO chromophores with stronger electron-donors.

T2 performance^[18].

A series of Y-type chromophores using Michlers ethylketone as the donor was first reported by our group, which had strong electron-donating ability and large steric hindrance, as shown in Fig. 1^[54, 58, 63–65]. The molecular conformation, including the hindrance of the donor and π -bridge, effectively isolates the chromophores and weakens the dipole–dipole interactions. Density functional theory (DFT) was used to calculate the HOMO–LUMO energy gaps and first-order hyperpolarizability of these chromophores. These chromophores showed better thermal stability with their decomposition temperatures all above 220 °C. Most importantly, the high molecular hyperpolarizability of these chromophores can be effectively translated into large EO coefficients in poled polymers due to the bulky donor structure. The poled films of FTC-yh1 and FTC-yh2 displayed r_{33} values of 149 and 143 pm/V at the concentration of 25 wt% in amorphous polycarbonate (APC)^[63]. To improve EO activity

based on FTC-yh1, two long alkoxy chains were introduced into thiophene-bridge to effectively isolate with other chromophores, and using stronger electron acceptor CF_3 -Ph-tricyanofuran (CF_3 -Ph-TCF) rather than TCF can significantly improve the β value. The poled films of 25 wt% and 35 wt% chromophore F in APC exhibited 305 and 358 pm/V respectively which were comparable with high-performance CLD-type chromophores^[54].

In recent years, the research focus has been shifted from electrically poled polymer composite materials to high-number-density neat (pure) chromophore materials that yield r_{33} larger than 500 pm/V in thin films^[66]. Moreover, theory has led the way to utilization of OEO materials with controlled lattice dimensionality (e.g., two-dimensional or Bessel lattice materials). The dipole-dipole aggregations can be harnessed by well-engineered molecular configurations. Dalton's group tried to do the poling for the neat JRD1 and YLD124 films that were prepared without EO inactive polymers, as shown in

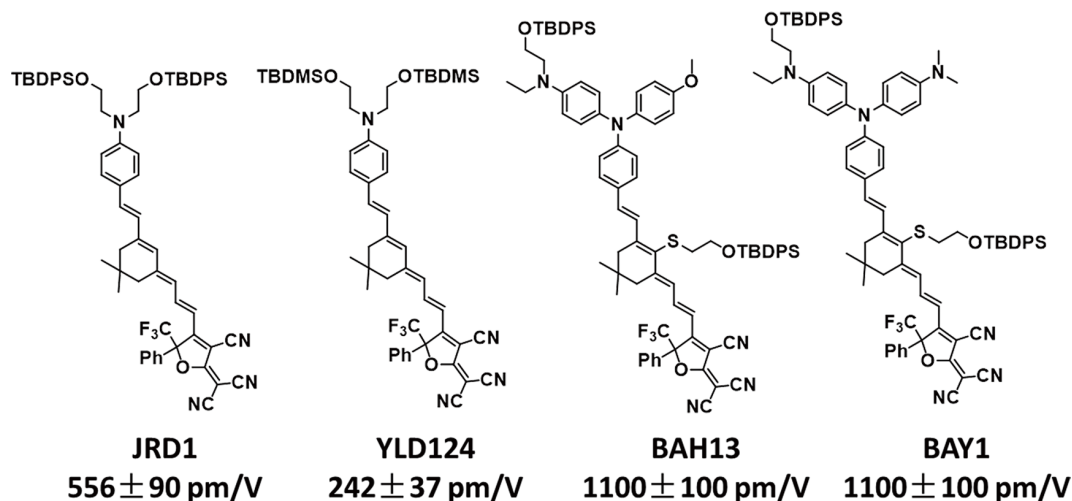


Fig. 2. High performance chromophores for neat film poling.

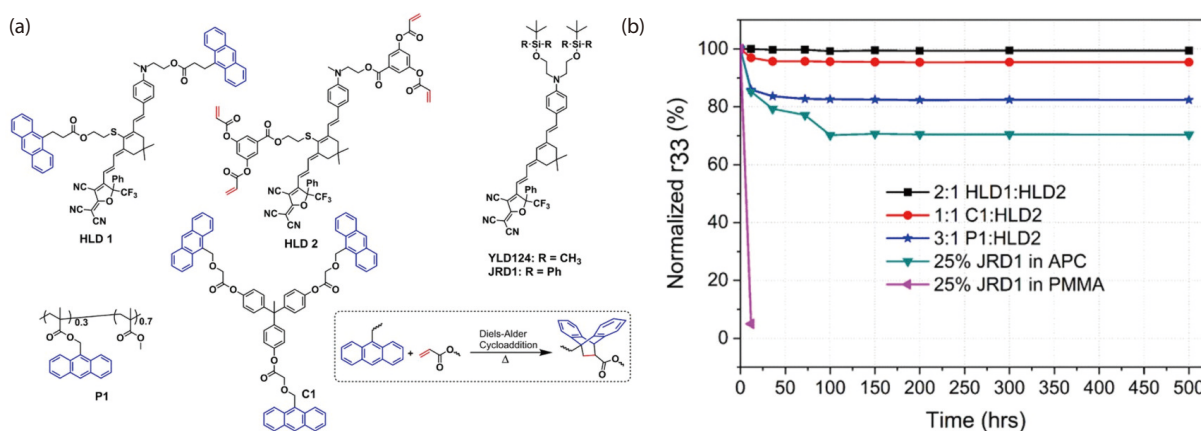


Fig. 3. (Color online) (a) Chemical structure for chromophores HLD1, HLD2, cross-linker C1, and polymer P1. (b) Temporal stability of the poled films HLD1/HLD2, HLD2/C1, HLD2/P1, JRD1/APC, and JRD1/PMMA at the curing temperature of 85 °C. Reproduced with permission from Ref. [27]. Copyright 2020, American Chemical Society.

Fig. 2^[56, 66]. Due to the greatly reduced electric resistance of neat chromophore films in poling, large leakage current was observed that will cause huge poling field loss. To overcome this problem, many kinds of dielectric materials have been used as charge blocking layers (CBLs) by inserting between EO layer and electrode^[66, 67]. By using CBLs, the poled JRD1 film exhibited greatly enhanced r_{33} value of 556 pm/V^[66].

Theoretical predictions suggest that the performance of OEO materials can be considerably greater than 1000 pm/V. Recently, significant progress has been made in theory-guided design for OEO chromophores that permits screening of many structures by evaluating their calculated hyperpolarizabilities, band gaps, and dipole moments, and finding materials optimized for multiple design criteria^[68, 69]. Dalton's group designed and synthesized several new chromophores with hugely improved molecular β and r_{33} values. As shown in Fig. 2, BAY1 and BAH13 exhibited the maximum r_{33} values close to 1100 pm/V. Titanium oxide and hafnium oxide were used as the CBLs. The EO performance of BAH13 was also evaluated by POH devices in which BAH13 could greatly reduce the $V_{\pi L}$.

To address the challenge of long-term and high-temperature stability of OEO materials, a crosslinkable EO system consisting of two chromophores, HLD1/HLD2, that can be electric field poled and then thermally crosslinked in situ has

been used to form a stable system, as shown in Fig. 3(a). A maximum of r_{33} of $290 \pm 30 \text{ pm/V}$ was obtained in a cross-linked film and greater than 99% of the initial r_{33} value was maintained after heating to 85 °C for 500 h in Fig. 3(b)^[27].

Though achieving very large r_{33} values of about 1000 pm/V in bulk thin film, the EO performance of OEO materials in POH or SOH devices is about 200 or 400 pm/V through the calculation of device's half-wave voltage. In the future, the research will focus on how to improve the EO performance in SOH, POH devices or other type nano-photonics.

3. Organic-based heterogeneous hybrid electro-optic devices

3.1. Photonic crystal-organic hybrid (PCOH) modulator

Photonic crystals are a type of artificial functional material that was first proposed in the late 1980s, which are formed by periodic arrangement of dielectric materials with different dielectric constants in space. If a line defect is introduced into a photonic crystal, then a photonic crystal optical waveguide (PCW) can be generated.

In 2008, Brosi *et al.* proposed a slot-photonic crystal slow-light phase modulator with a length of only 80 μm , a bandwidth of 78 GHz and a transmission speed of 100 Gbit/s^[70]. As shown in Fig. 4(a), the slow-light phase modulat-

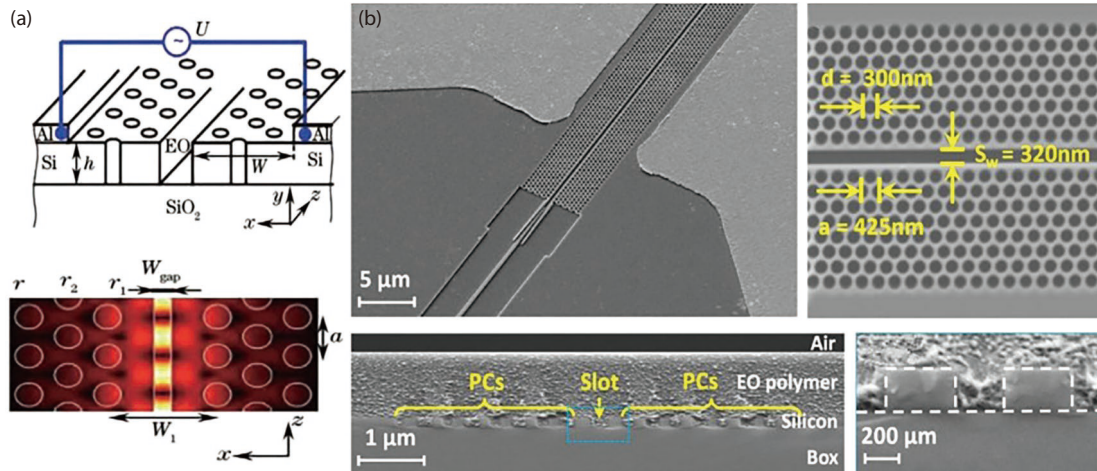


Fig. 4. (Color online) (a) Schematic of the slot-photonic crystal slow-light phase modulator and dominant electric field component E_x at quasi-TE mode. Reprinted with permission from Ref. [70], Copyright 2008, The Optical Society. (b) Scanning electron microscopy (SEM) images of the fabricated device. Reprinted with permission from Ref. [73], Copyright 2016, The Optical Society.

or consists of slotted photonic crystal waveguides (150 nm) filled with nonlinear OEO materials which form the interaction region. Due to the slow-light effect of the slot-photonic crystal waveguide, the velocity of the light field group in the modulation region decreased, the interaction time between the light field and the microwave electric field is significantly increased, and then the length of the modulator is reduced. In 2010, Chen's group demonstrated a PCOH modulator integrating silicon photonic crystal (PC) slot waveguide (75 nm) and AJ-CKL1/APC electro-optic polymer. An ultra-efficient EO modulation based on a half-wave voltage length product $V_{\pi}L = 0.56$ V·mm and an effective electro-optic coefficient $r_{33} = 132$ pm/V is obtained by using slow-light effect and strong field constraints in a slot waveguide^[71]. A new EO polymer-filled silicon slot PC waveguide modulator was reported and the structural diagram is shown in Fig. 4(b). The half-wave voltage V_{π} of the PCOH modulator is 0.97 V, and the half-wave voltage length product $V_{\pi}L$ is 0.291 V·mm and then the 3dB bandwidth is raised to 15 GHz in 2016^[72, 73].

3.2. Silicon-organic hybrid (SOH) modulators

Silicon photonics offers great potential for its CMOS compatibility with electronics and low-cost optoelectronic integration technologies. In recent years, various silicon-based active and passive optical waveguide devices have developed rapidly, and silicon photonics is a promising platform for highly integrated photonic chips due to its high-index-contrast and the compatibility with the mature complementary. However, silicon has nearly no electro-optic effect, so the traditional silicon modulators mainly rely on free carrier dispersion effect to achieve electro-optic modulation. Therefore, it is difficult for silicon modulators to achieve 100 GHz bandwidth and eliminate the nonlinear response^[17].

Integrating the excellent electro-optic materials into a mature silicon photonics platform is an effective means to overcome bandwidth and loss limits that stem from the physics of the plasma dispersion effect. SOH modulator combines with silicon-on-insulator (SOI) waveguides and the functional organic nonlinear materials with large r_{33} to realize the large bandwidth and low half-wave voltage ultra-compact modulator. Around 2011, a SOH phase modulator was first demonstrated by the Christian Koos and Juerg Leuthold's group at a

data rate of 42.7 Gbit/s with a widely open eye diagrams^[74]. In the SOH device, the silicon slot waveguide by increasing the doping concentration of the silicon strips can realize low resistivity, and the EO bandwidth is increased according to the corresponding RC time constant. As shown in Fig. 5(a), the electro-optic polymer M1 consists of chromophores dispersed in APC with the electro-optic coefficient of 70 pm/V, and the slot width of 120 nm and the 1.7 mm long phase shifting section is adopted in the device with a V_{π} of 5.3 V. The SOH phase modulator was reported by the same group with the frequency response of more than 100 GHz by using a device as short as 500 μm , a highly conductive electron accumulation layer and an improved gate insulator in Fig. 5(b)^[10]. The results indicate that the RC time constant is not a speed limitation of the device at 100 GHz.

Low energy consumption is a problem that must be considered. In 2015, the power consumption of a 1-mm-long SOH modulator was reduced to 0.7 fJ/bit, and the device was able to be operated at data rates of up to 40 Gbit/s^[31]. In Ref. [34], a SOH modulator with very low half-wave voltage (0.21 V) in a 1.5-mm-long device was reported (Fig. 5(c)). Organic chromophore JRD1 as a cladding, and the maximum n^3r_{33} amounts to 2300 pm/V with r_{33} of 390 pm/V was achieved in the SOH device. To overcome the RC limitation of SOH modulator, BaTiO₃ with high- κ can avoid the resistive slab and form a large coupling capacitor between the optical slot waveguides. A bandwidth of 76 GHz and a V_{π} of 1.3 V are achieved in the 1-mm-long SOH capacitively coupled modulator (Fig. 5(d))^[75].

The reported results showed that the poled polymer film can still maintain good electro-optic activity in the cryogenic region of 7 K^[76]. However, the thermal stability and long-term reliability of OEO materials have always been concerned because thermal relaxation of chromophores will happen when its working and storage temperature are higher than T_g . It illustrates that the thermal stability of the OEO materials seems to have been solved in the following work. As shown in the Fig. 5(e), the SOH modulator combined an ultra-high T_g of 172 °C EO polymer with silicon rib waveguides. The modulator showed 200 Gbit/s PAM4 modulations, and feature excellent reliability at ambient temperatures up to 110 °C^[77].

Silicon slot waveguides filled with OEO polymers ($r_{33} =$

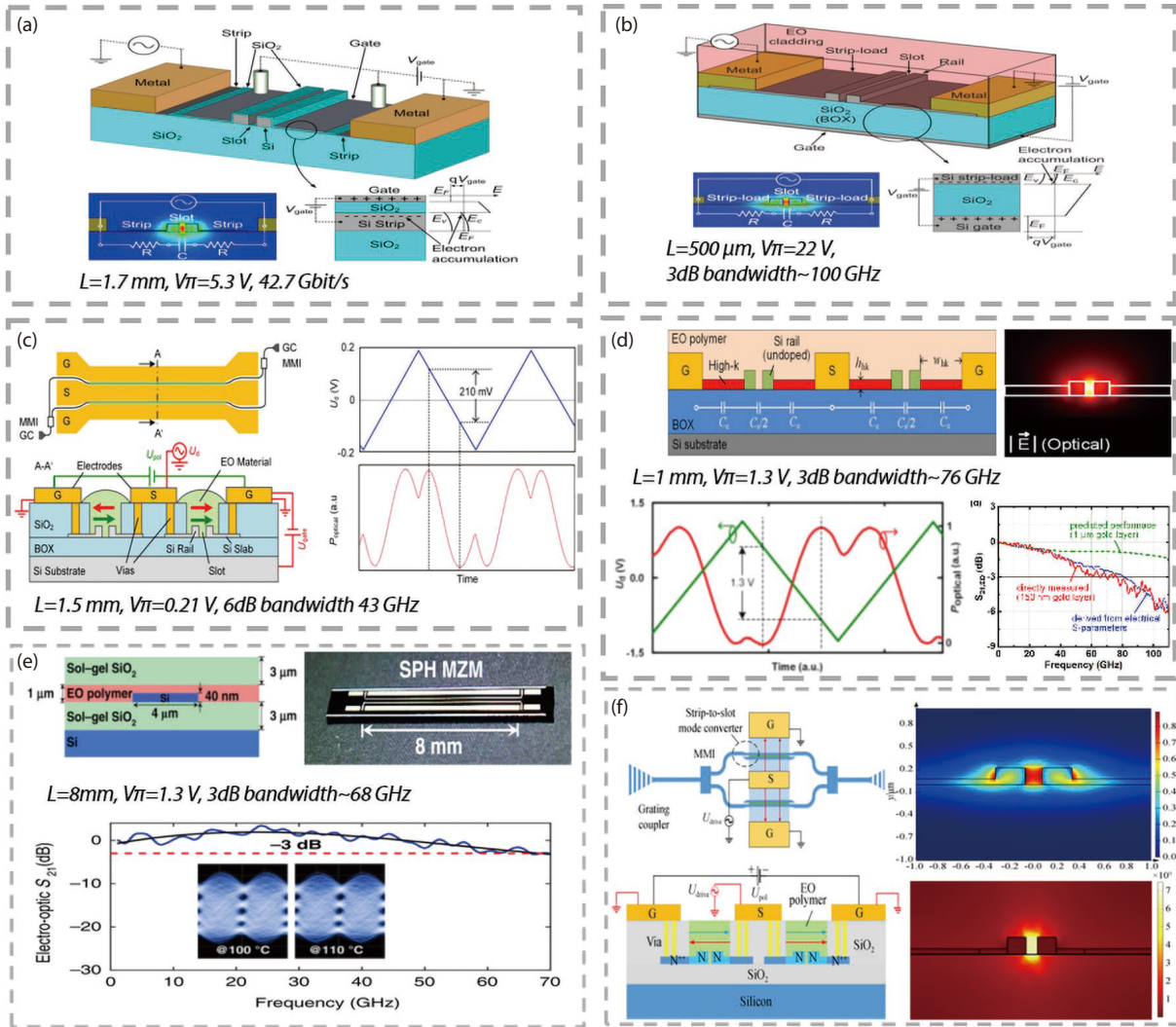


Fig. 5. (Color online) (a) The SOH phase modulator with an SiO₂ film on top of the silicon strips which cover with the gate electrode. Reproduced with permission from Ref. [74]. Copyright 2011, Optical Society of America. (b) 100 GHz SOH phase modulator. Reproduced with permission from Ref. [10]. Copyright 2014, Nature Publishing Group. (c) Ultra-low half-wave voltage of 0.21 V SOH MZM. Reproduced with permission from Ref. [34]. Copyright 2018, Optical Society of America. (d) Capacitively coupled SOH MZM with high-κ slotlines. Reproduced with permission from Ref.[75]. Copyright 2021, Optical Society of America. (e) High-temperature-resistant SOH MZM working up to 200 Gbit/s over 100 °C. Reproduced with permission from Ref. [77]. Copyright 2020, Nature Publishing Group. (f) The structure of SOH MZM by optimizing the strip-to-slot mode converter. Reproduced with permission from Ref. [78]. Copyright 2020, Optics and Precision Engineering.

150 pm/V) in a SOH Mach Zehnder modulator (MZM) were reported in internal research group. According to the simulation results, $V_{\pi}L$ of 0.45 V·mm and bandwidth of 70 GHz could be achieved^[78]. The overall structure and the cross-section structure of the modulator are shown in Fig. 5(f). The strip-to-slot mode converter has a cone-shaped coupling efficiency of 99.5%, which greatly reduces the loss caused by mode conversion.

3.3. Plasmonic–organic hybrid (POH) modulators

Highly compact nanometer optoelectronic integrated chips require smaller device sizes. However, the mode size of traditional dielectric waveguide is more than half of its wavelength length due to the diffraction limit. Surface plasmon photonics are a means of providing ultra-compact and high-speed components. Surface plasmon polariton (SPP) devices have developed rapidly, such as SPPs waveguide, SPPs modulator, SPPs sensor and so on. Plasmonic–organic hybrid modulators have attracted much attention in recent years which combine many advantages of the large RF band-

width of a travelling wave modulator, the compactness of a photonic-crystal modulator, and the energy efficiency of a ring modulator.

The size variation of the various organic-based hybrid modulators (all-organic, SOH, and POH) is shown in Fig. 6(a)^[11]. All-organic thin film OEO devices consist of upper and lower cladding layer and core OEO layer which are all made of polymer. The three layers are sandwiched between top and bottom electrode. To achieve low half-wave voltage, devices of 1–2 cm in length are typically required, because the electrode separations of greater than 7 μm (thickness of three-layer films)^[79]. Although all-organic devices achieved significant drive voltage and bandwidth, they still suffered from a large footprint for integration of electronics and photonics. As shown in Fig. 6(b), in the SOH modulator, the electrical field drops only across the silicon slot waveguide filled with the OEO materials, and this allows strong overlap of optical and RF electric fields, reducing device length to a few hundred micrometers^[79]. POH modulator uses metal-slot waveguides

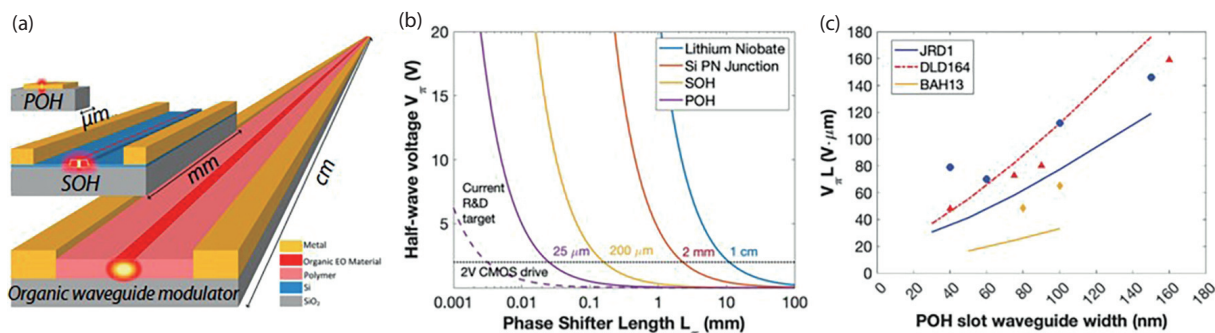


Fig. 6. (Color online) (a) The relative size of all-organic, SOH and POH modulator. Reproduced with permission from Ref. [1]. Copyright 2017, American Chemical Society. (b) Variation of halfwave voltage (V_{π}) with electrode length/device length (L) for various types of devices. Reproduced with permission from Ref. [79]. Copyright 2021, American Chemical Society. (c) Comparison measured (symbols) and computationally predicted (lines) $V_{\pi}L$ values for JRD1, DLD164, and BAH13 organic OEO materials at 1550 nm in a POH MZM. Reproduced with permission from Ref. [69]. Copyright 2022, Royal Society of Chemistry.

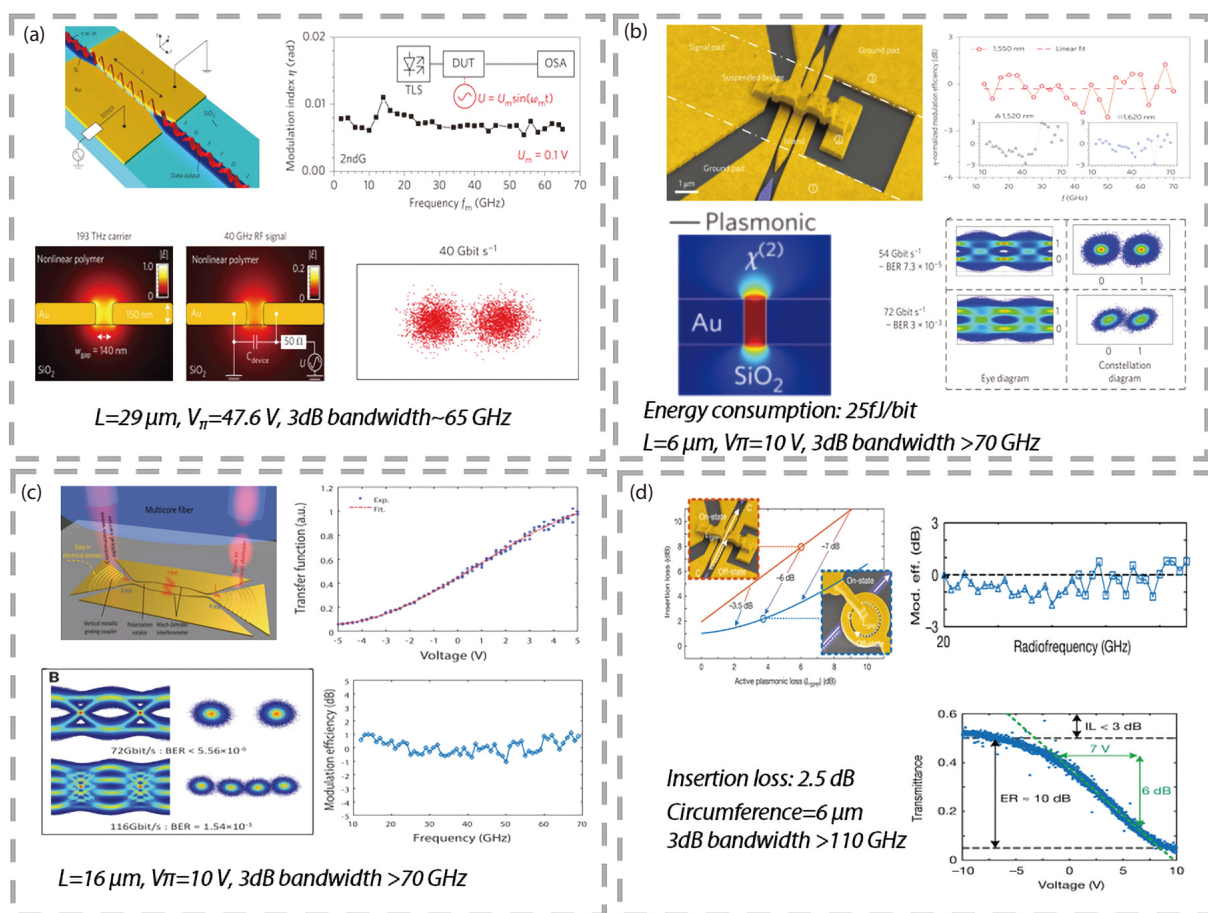


Fig. 7. (Color online) (a) A high-speed POH phase modulator designed and fabricated. Reproduced with permission from Ref. [11]. Copyright 2014, Nature Publishing Group. (b) POH MZM with metal-insulator-metal plasmonic slot waveguide. Reproduced with permission from Ref. [12]. Copyright 2015, Nature Publishing Group. (c) All-plasmonic MZM using a single metal layer without the silicon waveguide. Reproduced with permission from Ref. [38]. Copyright 2017, Nature Publishing Group. (d) Low-loss plasmonic electro-optic ring modulator. Reproduced with permission from Ref. [40]. Copyright 2018, Nature Publishing Group.

rather than silicon slot waveguides and exploits stronger confinement of light for surface plasmon polaritons, thereby reducing device lengths to a few tens of micrometers and electrode spacing to less than 50 nm. For the 2 V CMOS drive, the phase shifter lengths for lithium niobate and Si PN junction are 1 cm and 2 mm, while phase shifter lengths for SOH and POH are only 200 and 25 μm , respectively. Therefore, the SOH and POH showed the small footprint for the CMOS process-compatible chip-scale integration. $V_{\pi}L$ values from 40 to

50 $\text{V}\cdot\mu\text{m}$ have been reported (symbols) in JRD1 and DLD164 for POH devices in Fig. 6(c), and even lower values based on demonstrated improvements in new materials[69].

In 2014, J. Leuthold's group first reported a new POH phase modulator, and the structure diagram is shown in Fig. 7(a)[11]. Light enters the plasmon waveguide from the silicon waveguide and propagates in the surface plasmon (SPP) mode. The gap is filled with OEO materials, and the phase modulation is realized by applying voltage to the metal. The

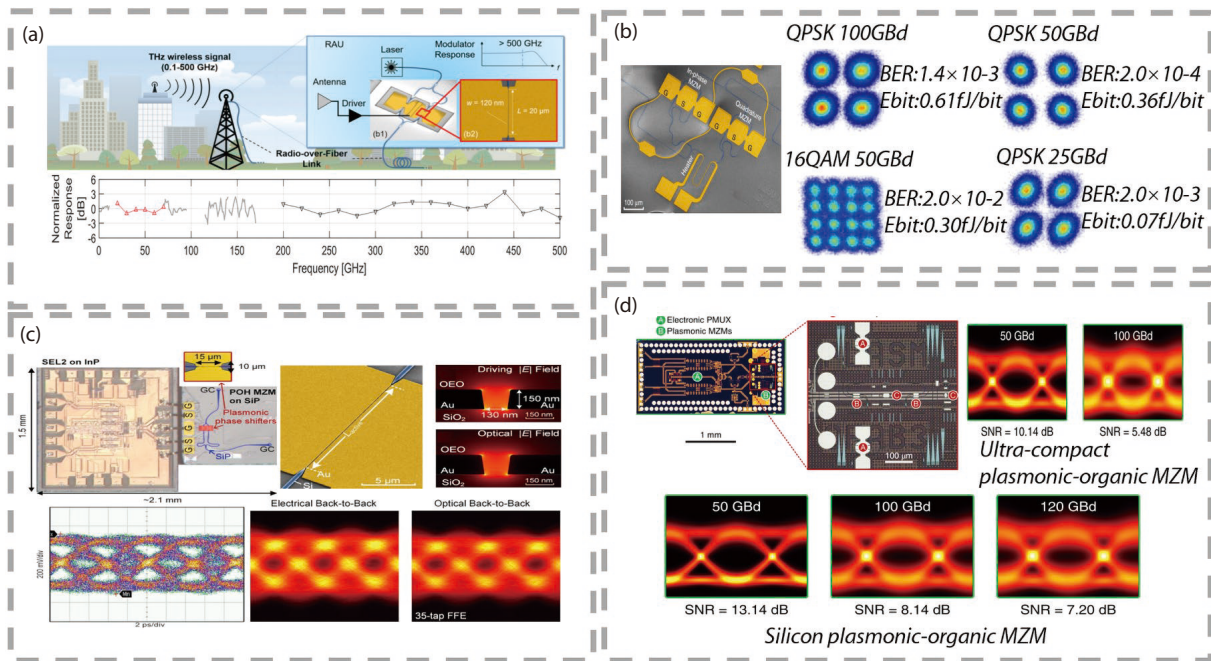


Fig. 8. (Color online) (a) Beyond 500 GHz POH MZM used for sub-THz microwave photonics. Reproduced with permission from Ref. [41]. Copyright 2019. American Institute of Physics. (b) 222 GBd on-off-keying transmitter based on POH MZM. Reproduced with permission from Ref. [80]. Copyright 2020. Optical Society of America. (c) Compact IQ electro-optic modulator operated with sub-1-V driving electronics. Reproduced with permission from Ref. [42]. Copyright 2019. Nature Publishing Group. (d) Symbol rates 100 GBd monolithically integrated electro-optical transmitter based on POH MZM. Reproduced with permission from Ref. [44]. Copyright 2020. Nature Publishing Group.

device is only $29 \mu\text{m}$ long, operates up to 40 Gbit/s, and has a frequency response of at least 65 GHz. Next, a POH intensity MZM was demonstrated with a bandwidth of 70 GHz in 2015, and the scanning electron microscope image of the MZM component is shown in Fig. 7(b). The device has a $V_{\pi}L$ of 0.06 V·mm and an energy consumption of 25 fJ/bit, which is attributed to the filling of the slot with a polymer called DLD-164^[12].

In contrast to the previous two works, the all-plasmonic MZM without any silicon waveguide is reported in Fig. 7(c)^[38]. All the elements (grating couplers, splitters, polarization rotators and active section) are included in a single metal layer in this POH MZM. The length of phase shifter is $16 \mu\text{m}$, and the half-wave voltage is 10 V. The frequency response of the device exhibits no speed limitation up to 70 GHz and 116 Gbit/s PAMs modulation formats.

The electron motion generates heat through ohmic losses for plasmonics, which has led to a broad view that plasmonics is too lossy for optical transmission. To reduce the intertion loss of POH modulator, a plasmonic electro-optic ring modulator was reported in the Fig. 7(d). The ohmic losses can be bypassed by using “resonant switching”. The intertion loss of the device is only 2.5 dB which can be reduced by more than 6 dB relative to non-resonant MZM device. The bandwidth of the plasmonic resonator is beyond 110 GHz^[40].

Plasmonics is indeed a viable path to an ultracompact, highest-speed, and low-cost technology modulator that can be used in a wide range of fields. The applications of POH modulators have been carried out. Terahertz wireless applications demands modulators with frequency responded to the sub-terahertz range, high power handing, and very low nonlinear distortions. Beyond 500 GHz, V_{π} of 3 V, and high linearity $25 \mu\text{m}$ -long POH modulator was experimentally demonstrated, which can achieve a 222 GBd on-off-keying transmit-

ter in the Figs. 8(a) and 8(b)^[41, 80]. The POH modulator shows good performance, not only in the digital signals but also in the sub-terahertz radio-over-fiber analog optical link. The POH modulator has high linearity with a third-order input intercept point (IIP3) of 18.9 dBm. Meanwhile, an analog RoF link over 100 GHz bandwidth (220–325 GHz) was used to prove the capability of direct terahertz-to-optical-to-terahertz conversion of the POH modulator, and the experiment represents the link has a flat response over the complete 220–325 GHz frequency range. Based on these results, POH modulators have potential applications in the general field of microwave photonics, 5G wireless communications, antenna remoting, internet of things, sensing and more analogy links for the sub-terahertz range.

Heni *et al.* demonstrated an in-phase/quadrature (IQ) POH modulators, and the active region was $4 \times 25 \mu\text{m} \times 3 \mu\text{m}$ with sub-1-V driving electronics. As shown in Fig. 8(c), the devices exhibit low electrical energy consumptions of 0.07 fJ/bit at 50 Gbit/s, 0.3 fJ/bit at 200 Gbit/s, and 2 fJ/bit at 400 Gbit/s with organic chromophore HD-BB-OH/YLD124 filling the slot^[42]. The IQ POH modulator can be used in long-haul and short-haul communications.

Recently, a symbolic rate over 100 GBd single-chip electro-optical transmitter that combines advanced bipolar CMOS with silicon plasmonic was reported. This is a great progress, because it addresses key challenges in single-chip integration through the co-design of electronics and plasmonic layers in the Fig. 8(d)^[44]. The HLD1/HLD2 organic material in the plasmonic modulator shows very excellent electro-optic and thermal-stability performance. The POH modulator based on the above organic materials can operate stable data modulation at temperatures above 112 °C.

The calculated capacitance of POH modulator is approximately several fF, so the RC bandwidth can break through the

Table 1. The MZM on various EO platforms with operating principle of Pockels effect. The best result reported is given in parenthesis.

Platform	3-dB EO bandwidth (GHz)	V_{π} (V)	Footprint (mm)	Loss (dB/cm)
SOH	>60 (76 ^[75])	0.21 V ^[34]	<1	~20 (22 ^[83])
POH	>100 (500 ^[41])	4.8 V ^[24]	<0.02	~500 (400 ^[12])
PCOH	78 ^[70]	0.94 ^[73]	~0.3	~200 ^[73]
LNOI	>67 (110 ^[81])	1.4 ^[81]	5–20	~0.3
LN/Si	70 (106 ^[82])	5.1 ^[15]	>5	~1 (0.98 ^[15])

limitation of RC time constant and be much greater than 1 THz. Meanwhile, the Pockels effect is based on the reorganization of π -electrons within a nanometer-long molecule of OEO materials and is estimated to be on the timescale of femtoseconds or attoseconds. The Pockels bandwidth will be much greater than 1 THz. Therefore, the POH modulator has a great space for the ultra-large bandwidth.

4. Conclusion

The recent developments of OEO materials and organic-based hybrid modulators were reviewed. The outstanding characteristic of OEO materials is their design flexibility which makes OEO materials have high electro-optic activity, fast response speed, high processability, good thermal stability and low cost. Meanwhile, the OEO material is flexible and compatible with many processes (CMOS, silicon photonics) without bonding. The defect for both datacom and telecom system is the lack of performance, specifically as it relates to very high speeds and very low power consumption. Heterogeneous integration based on the organic materials is a very promising solution for high-speed optical modulators and communication system.

Table 1 presents various performance for the high-speed modulators with operating principle of Pockels effect in SOH, POH, PCOH, LNOI, and LN/Si (LN film bonded to SOI platform). The linear Pockels effect can offer intrinsically good linearity. SOH has the lowest half-wave voltage (0.21 V) because of the large EO coefficient ($r_{33} = 390$ pm/V) of OEO polymer in the device. The bandwidth of SOH is intermediate between POH and thin film lithium niobite based modulators. POH modulators have the highest bandwidth and the smallest footprint, which make them promising materials for ultra-high speed and low energy consumption integrated system on a chip (SOC). Compared with the LNOI, POH modulators show the large propagation losses which can be solved through waveguide structure and manufacturing process^[40]. PCOH modulators are less studied, perhaps because they need to fabricate complex photonic crystals. The LNOI modulator exhibits ultra-low propagation losses, but may be difficult to achieve low driving voltage for CMOS-level and small size simultaneously owing to the fixed r_{33} .

It is reasonable to find suitable applications for different materials because discrete device applications will likely be based on low optical loss materials such as lithium niobite and silicon. For the application of chip-scale integration of electronics and photonics, the requirement of small size close to μm and CMOS-level driving voltage less than 1 V will likely favor OEO materials. OEO materials are suitable for the satellite and space telecommunications because they may be used to develop small size, lightweight, excellent efficiency and radiation resistance devices^[79]. The range of applications for electro-optics is broad, besides the high-capacity telecommunica-

tions and ultra-bandwidth microwave photonics, electro-optics can be used in Lidar, sensor, computing, THz and other areas.

In the future, new OEO materials can be designed and synthesized to optimize the EO performance, refractive index, optical loss and EO thermal-stability, and offer many potential possibilities for high-performance EO modulators. SOH modulator shows low $V_{\pi}L$ and energy consumption, and the aim is to improve the bandwidth and maturity of processing technology. A POH device is a highly efficient modulator to achieve unprecedented performance (very high speed, low power consumption) and compactness, and designing the new waveguide structure and optimizing preparation process should be attracted attention to reduce optical loss. More and more research institutions, universities and companies (e.g., Light-wave Logic) have already studied the organic EO materials devices, and high-performance optical circuits and system could be expected.

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