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# Q-Switched and mode-locked pulse generation with poly (9-vinylcarbazole) saturable absorber

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Poly (9-vinylcarbazole) (PVK) organic semiconductor material has been extensively investigated for photonic applications. Here, we demonstrate for the first time the utilization of PVK film as a saturable absorber (SA) to realize stable Q-switched and mode-locked fiber lasers. The modulation depth and the saturation power intensity of the prepared PVK film are 4% and 202 MW/cm$^2$, respectively. The proposed Q-switched laser operated at 1562 nm. The repetition rate increased from 60.10 kHz to 91.91 kHz, while the pulse width decreased from 6.00 µs to 3.43 µs, as the pump power was increased from 63.4 mW to 125.2 mW. The maximum pulse energy of 8.73 nJ was obtained at 125.2 mW pump power. Meanwhile, the proposed mode-locked laser operated at 1567 nm with 1.873 MHz repetition rate and 2.81 ps pulse width. The stable soliton mode-locked fibre laser was achieved by adding 100 m long single-mode fiber (SMF) in the cavity. At 187.0 mW pump power, the maximum pulse energy and average output power was obtained at 2.003 nJ and 3.751 mW, respectively. The fundamental frequency has a high signal to noise ratio of 63.10 dB, which indicates the excellent stability of the pulses. Our results contribute to the growing body of work studying the nonlinear optical properties of PVK that underpin new opportunities for pulsed laser technology.

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*Keywords:* Q-switching, Mode-locking, Organic semiconductor saturable absorber, Fiber laser

## 1. Introduction

Q-switched and mode-locked fiber lasers have become a mature technology, facilitating a broad range of applications, including laser material processing, medical surgery, optical communications and fiber sensing [1] [2] [3] [4]. Active and passive techniques can be used to generate pulsed lasers but passive schemes based on saturable absorbers (SAs) are preferable due to their simplicity of design, compactness, low cost and flexibility [5]. The SAs act as an optical switch that provides an intensity-dependent transmission without the implementation of expensive and complicated external modulators. Up to date, various types of saturated absorbing materials have been used and they include semiconductor saturable absorber mirrors (SESAMs), nanomaterials such as carbon nanotubes (CNTs), graphene, topological insulators (TIs), transition metal dichalcogenides (TMDs) and black phosphorus.

SESAMs are stable and flexible but expensive, complicated to manufacture and have limited operation wavelength [6-7]. SAs based on carbon nanomaterials such as CNTs and graphene are cheaper, easier to fabricate and have a moderate damage threshold [8]. However, CNT has a narrowband saturable absorption, weak stability, and low reliability [9] while Graphene has a low modulation depth and weak saturable absorption [10]. Topological insulators like $Bi_2Te_3$, $Bi_2Se_3$, and $Sb_2Te_3$, have narrow topological energy gaps, wideband saturable absorption, high damage threshold, large modulation depth and low saturable optical intensity [11-14]. Transition metal dichalcogenides like $WS_2$ and $MoS_2$ also have wideband wavelength operation and higher third-order nonlinearity properties. Besides, they are easy and cheap to fabricate [15-18]. Unlike other nanomaterials, black phosphorus has a layer-sensitive direct bandgap that changes from 0.35 eV (bulk) to 2 eV (monolayer layer). However, this material has poor stability in ambient atmosphere [19] [20].

Lately, organic materials that are environmentally friendly and bio-compatible have also been experimented with as SA materials. They are favorable since they do not pose any health risk to human. Poly(9-vinylcarbazole) (PVK) is an organic material that can endure a high temperature and have a good hole mobility [21]. PVK has a bandgap energy of about 3.4 eV and it has been used in the manufacturing of organic light-emitting diodes (OLEDs), transistors, non-volatile memory devices and solar cells [22]. In this work, a PVK thin film is prepared and used to construct an SA. The SA is inserted in an all-fiber laser cavity to generate a stable Q-switched and mode-locked pulses. The proposed laser generates a stable Q- switched pulse train at 1562 nm with a repetition rate of 60.10 kHz to 91.91 kHz and a corresponding pulse width of 6.00 μs to 3.43 μs as the pump power is increased from 63.4 mW up to 125.2 mW. Besides that, self-starting mode-locked pulse train at 1567 nm with a constant repetition rate of 1.866 MHz and the pulse width of 2.81 ps are observed as the pump power is increased from 125.2 mW up to 187.0 mW. To the best of our knowledge, this is the first demonstration of the using an organic semiconductor material as a Q-switcher and mode-locker.

## 2. Preparation of PVK SA

The PVK thin film used to form the SA was prepared by the spin-coating technique as illustrated in Fig. 1. First, the organic PVK compound purchased from Sigma Aldrich was dissolved in chloroform solvent with a concentration of 10 mg/mL. The solution was stirred for about 15 minutes to ensure that all the composites were fully dissolved. A small volume of the prepared PVK solution was taken using a micro-syringe and dispensed on top of a glass substrate. Beforehand, the glass substrate was ultrasonically cleaned in distilled water, followed by acetone and ethanol. It was then purged with nitrogen gas to dry its surface. The spinning process was then executed at 2000 rpm for 40 s. During the spin rotation, the solution on the substrate experienced centrifugal force which spread it thinly on the substrate. Meanwhile, the solvent of the solution evaporated, leaving a PVK film with a thickness of approximately 80 nm. Then the film was soft-baked where the substrate was heated on a hot-plate at a temperature of the boiling point of chloroform for 15 minutes to enhance the solvent evaporation rate and ensure no solvent was left in the film.

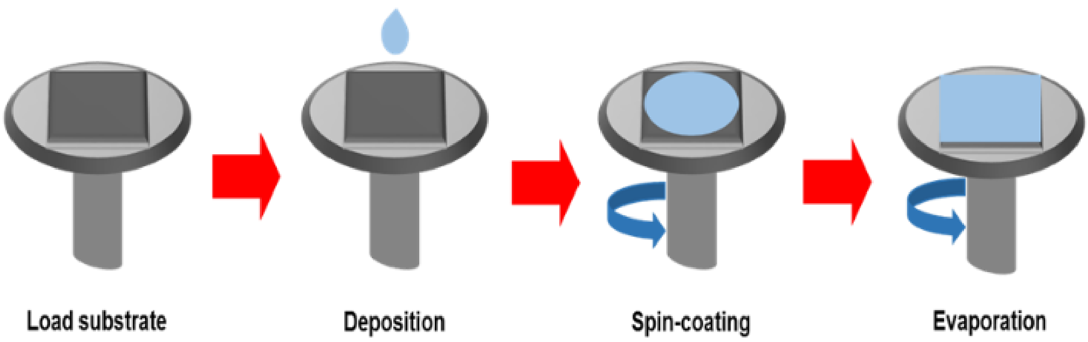
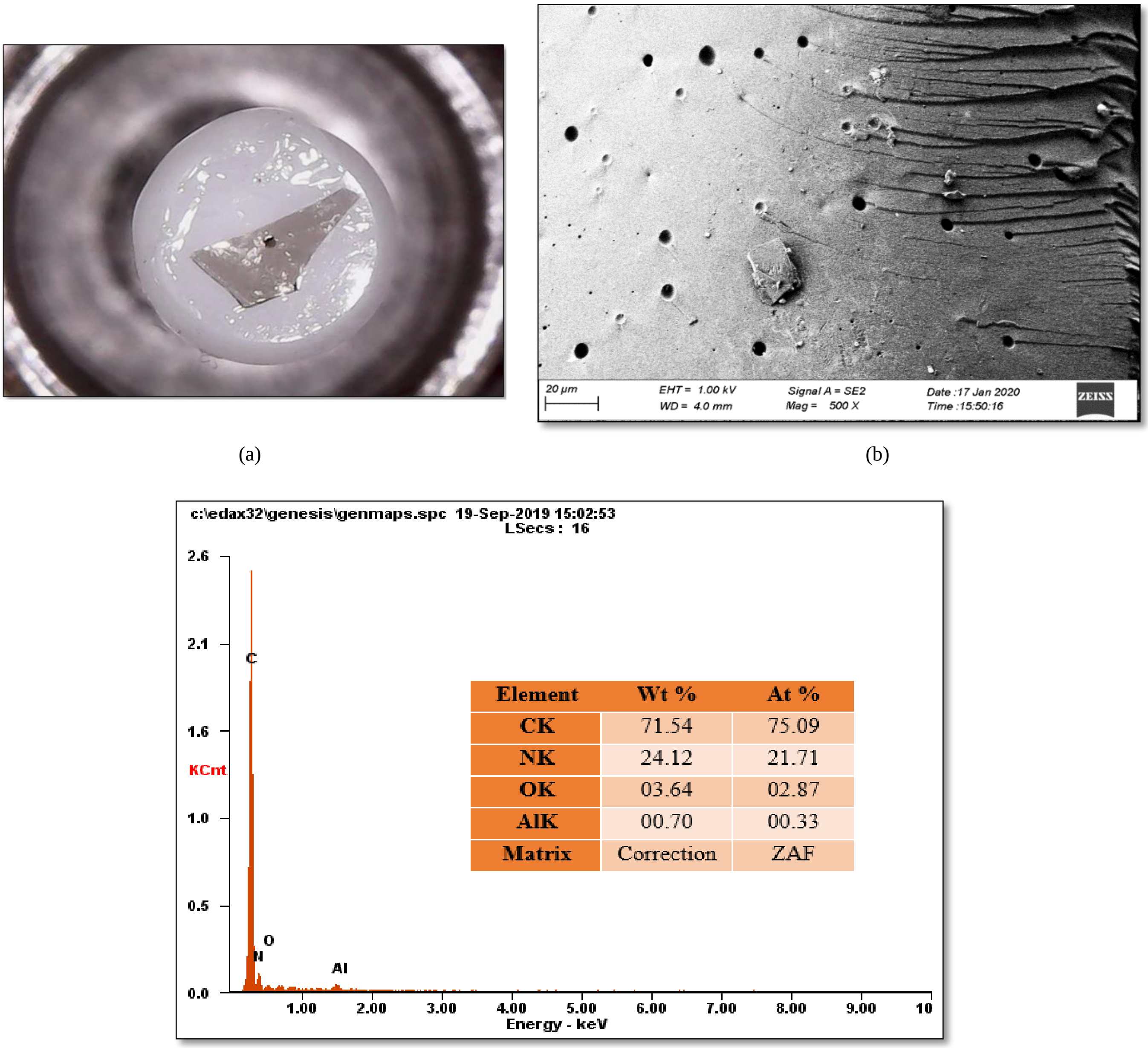



*Fig. 1. Spin-coating technique of PVK:PVA thin film (color online)*

Finally, the PVK film was peeled from the glass substrate and a small portion of it was cut and placed onto a fiber ferrule as shown in Fig. 2(a). It was then mated with another clean ferrule via a fiber adaptor to form a SA device. The surface morphology of the PVK film was characterized by using a Field Emission Scanning Electron Microscope (FESEM). Fig. 2(b) shows the FESEM image, which exhibits a smooth surface with few large particles. It was also analyzed with energy dispersive spectrometry (EDS) to identify its main elements. The results in Fig. 2(c) shows that the film contains 71.54 % Carbon, 3.64 % Oxygen, 24.12% Nitrogen, and 0.70 % Aluminium.




(a) (b)

(c)

*Fig. 2. (a) PVK on the ferrule (b) FESEM image (c) EDS profile (color online)*

The linear optical absorption profile of the PVK film is shown in Fig. 3(a), which indicates an absorption loss of around 3 dB at 1550 nm. It was obtained by launching a broadband light source (Anritsu, MG922A) into the film and measuring the transmitted light using an Optical Spectrum Analyzer (OSA: Anritsu, MS9710C, 0.6 – 1.75 µm) with a resolution of 0.02 nm. Measurement of the saturable absorption of the PVK SA was carried out using a balanced twin-detector as illustrated in Fig. 3(b). The homemade mode-locked laser operating at 1557.7 nm with a repetition rate of 1.885 MHz and a pulse width of 3.62 ps was used as the pump light source. After passing through an adjustable attenuator (Thorlabs EVOA1550A), the input was split using a 50:50 fiber coupler; where half of the beam was used for detecting absorption and the other half for monitoring as a reference signal. By adjusting the attenuator, the transmitted power as a function of the incident optical power was recorded via photodetectors PD1 and PD2 (Thorlabs DET01CFC). Optical power meter (Thorlabs PM 100D) was used to measure the output power for nonlinear absorption measurement. With increasing peak power intensity, the transmittance of the SA tends to a constant, confirming saturable absorption. The nonlinear absorption obtained is plotted in Fig. 3 (c). The PVK film possesses a saturable absorption of 4%, a non-saturable absorption of 78%, and a saturation intensity of 202.07 MW/cm$^2$. This confirms its ability to convert continuous-wave into a train of short pulses in an EDFL cavity.

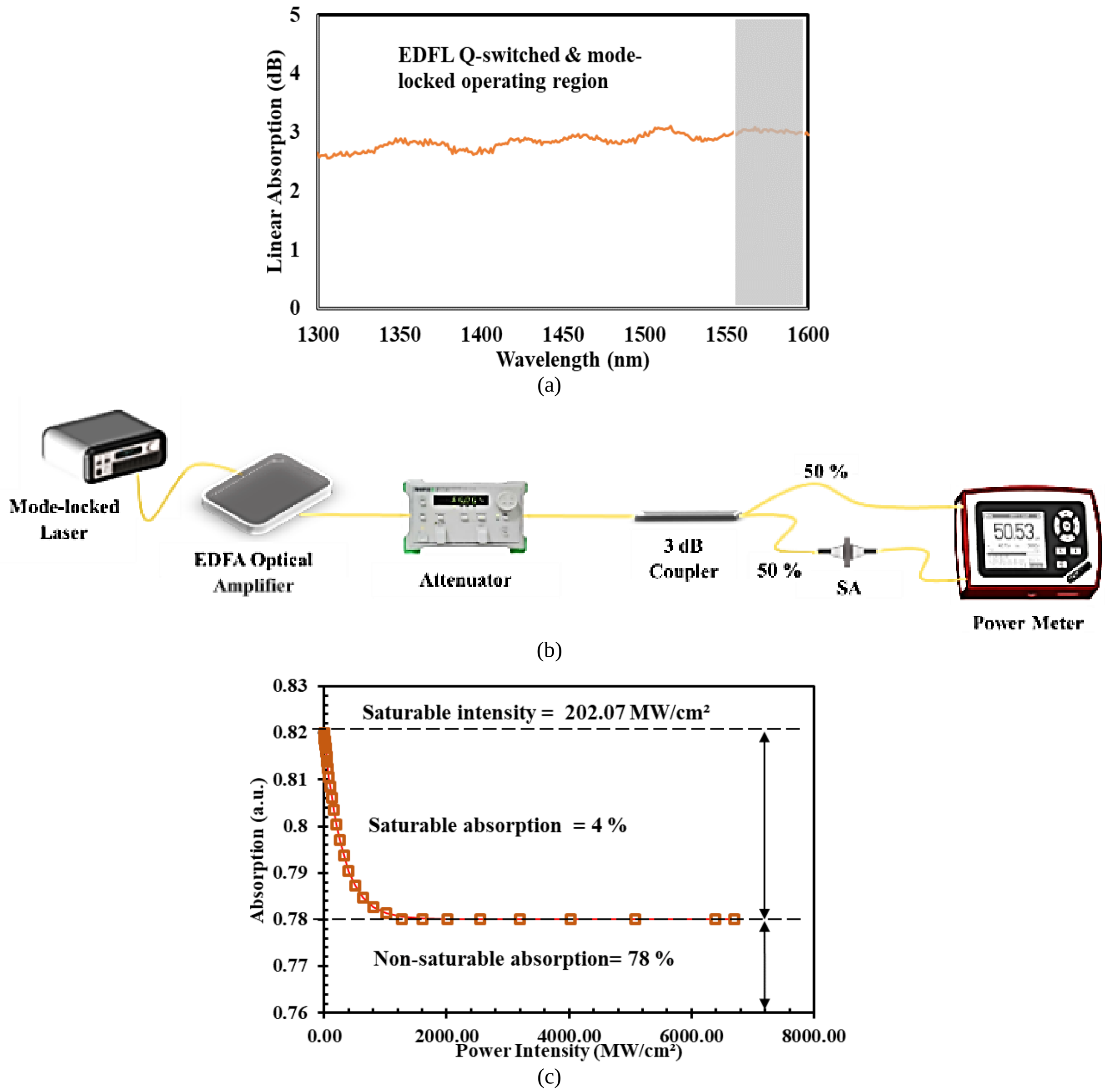



*Fig. 3. (a) Linear absorption profile of PVK thin film (b) Nonlinear optical measurement (c) Nonlinear absorption profile of PVK thin film (color online)*

## 3. Laser configuration

The PVK SA can be used to generate both Q-switched and mode-locked fiber lasers. For Q-switched fiber laser, a 2.4 m long Erbium-doped fiber (EDF) is used as the gain medium. The EDF has a peak absorption of 23.9 dB/m at 980 nm and a cut-off wavelength of 900 nm. The numerical aperture of the EDF is 0.24 while its core and cladding diameters are 5.1 and 125.4 µm, respectively. Its peak absorption is 41.1 dB/m at 1530 nm. Besides the SA and EDF, other components that make up the ring cavity include, a 980/1550 wavelength division multiplexer (WDM), a polarization-independent optical isolator and an output coupler as shown in Fig. 4. The EDF is pumped by a 980 nm laser diode through the WDM. The isolator ensures a unidirectional propagation of the laser. The output coupler with a splitting ratio of 10:90 is used to take out 10% of the laser for analysis in time and frequency domains. The 10% output is further divided by a 50/50 fiber coupler to enable two simultaneous measurements.

For mode locked laser, a 100 m long single-mode fiber (SMF) is added to manage the dispersion regime and increase the nonlinearity of the cavity. Thus, the total length of the cavity increases to ~111 m. The performance of the Q-switched and mode-locked fiber laser is measured using an optical spectrum analyzer (OSA: Anritsu, MS9710C, 0.6 – 1.75 µm), an autocorrelator, an oscilloscope (GWINSTEK: GDS-3352), and a radio frequency (RF) analyzer (Anritsu MS2683A) with a high-speed photodetector. The autocorrelator is also used to measure the precise temporal characteristics of the mode-locked laser as the resolution of the oscilloscope is limited within microsecond to nanosecond.

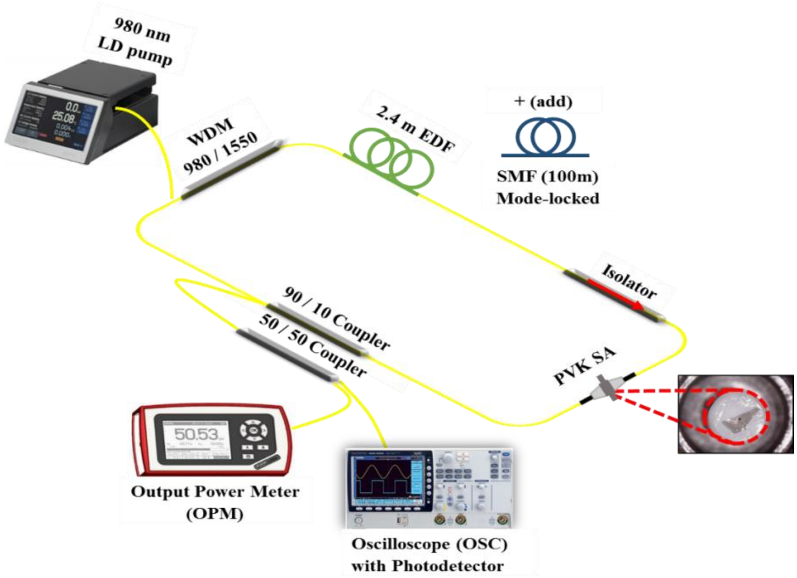



*Fig. 4. The configuration of the Q-switched and mode-locked fiber laser with EDF as a gain medium and PVK thin film as a SA (color online)*

## 4. Q-switched laser operation for PVK SA

Without the PVK SA, the EDFL operated in continuous wave (CW) mode. With the PVK SA, Q-switched pulses were generated when the input pump power reached 63.4 mW and beyond. The output pulse trains at three different pump powers are shown in Fig. 5. At 63.4 mW pump power, the Q-switched laser had a repetition rate of 60.10 kHz and pulse width of 6.00 µs as shown in Fig. 5 (a). As the pump power was increased to 96.4 mW, the repetition rate rose to 76.92 kHz, while the pulse width contracted to 4.20 µs as shown in Fig. 5 (b). Fig. 5 (c) shows the maximum repetition rate of 91.91 kHz and the minimum pulse width of 3.43 µs of the Q-switched laser at the maximum pump power of 125.2 mW. The output pulse train was stable until the pump power reached 125.2 mW, beyond which it became unstable.

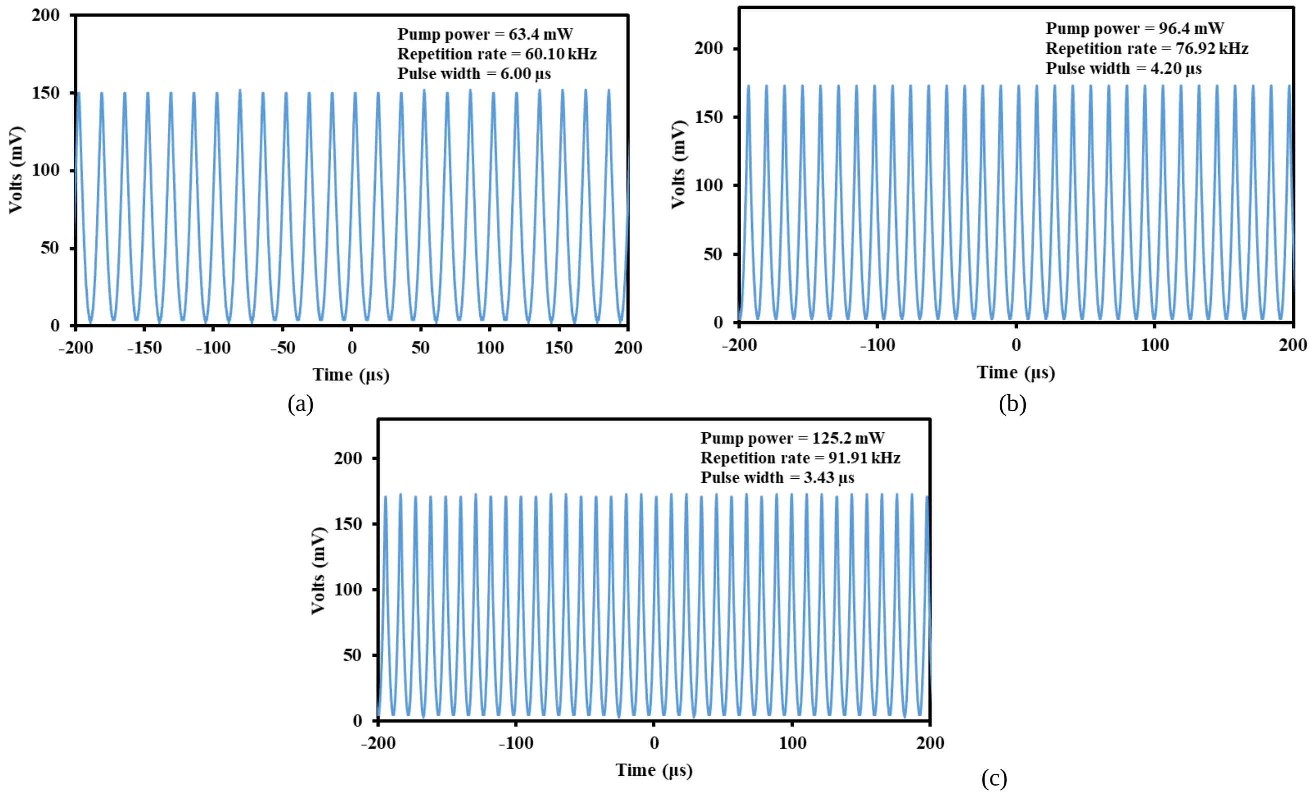



*Fig. 5. Q-switched output pulse train using PVK SA at incident pump powers of (a) 63.4 mW, (b) 96.4 mW, and (c) 125.2 mW*

Fig. 6(a) shows the optical spectrum of the Q-switched EDFL at the pump power of 125.2 mW. The peak lasing is at 1562 nm with a 3 dB bandwidth of 0.5 nm. Due to the SA, the cavity loss increases and shifts the operating wavelength slightly to get more gain to offset the loss. Fig. 6 (b) shows the RF spectrum of the Q-switched EDFL at the input pump power of 125.2 mW to check the stability of the pulse operation. Fig. 6 (c) shows the enlarged spectrum depicting the fundamental frequency at 92.61 kHz with a signal-to-noise ratio (SNR) of 51.14 dB which confirms the pulse stability. We also increased the pump power up to the maximum power of 300 mW to investigate the damage threshold of the SA. When the pump power was decreased below 125.2 mW the stable Q-switched pulses returned. This shows the PVK SA has a good damage tolerance as there was no thermal damage caused by the high input pump power.

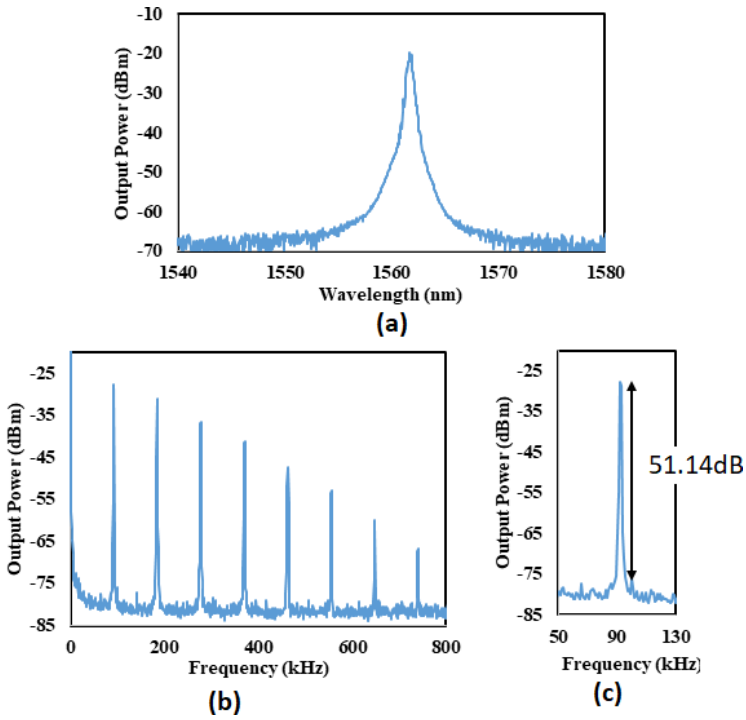



*Fig. 6. Spectral and temporal characteristics of the PVK SA based Q-switched laser at 125.2 mW pump power (a) Optical spectrum (b) RF spectrum with 800 kHz span (c) Enlarged RF spectrum with 80 kHz span showing the fundamental frequency*

Unlike mode-locked laser which has a fixed repetition rate determined by the cavity length, Q-switched laser has a variable repetition rate. Fig. 7(a) shows the pulse repetition rate and pulse width of the Q-switched laser as functions of the incident pump power. The repetition rate is observed to increase from 60.10 kHz to 91.91 kHz with the increment of incident pump power from 63.4 mW to 125.2 mW. However, the pulse width decreases from 6.00 µs to 3.43 µs. Fig. 7(b) summarizes the relations between the output power or single pulse energy and the pump power. Both output power and pulse energy increase as the incident pump power rises. The output power increases almost linearly with the pump power and the maximum recorded output power is 0.802 mW at the pump power of 125.2 mW. The pulse energy varies from 5.26 nJ to 8.73 nJ, with the maximum value of 8.73 nJ achieved at 125.2 mW pump power.

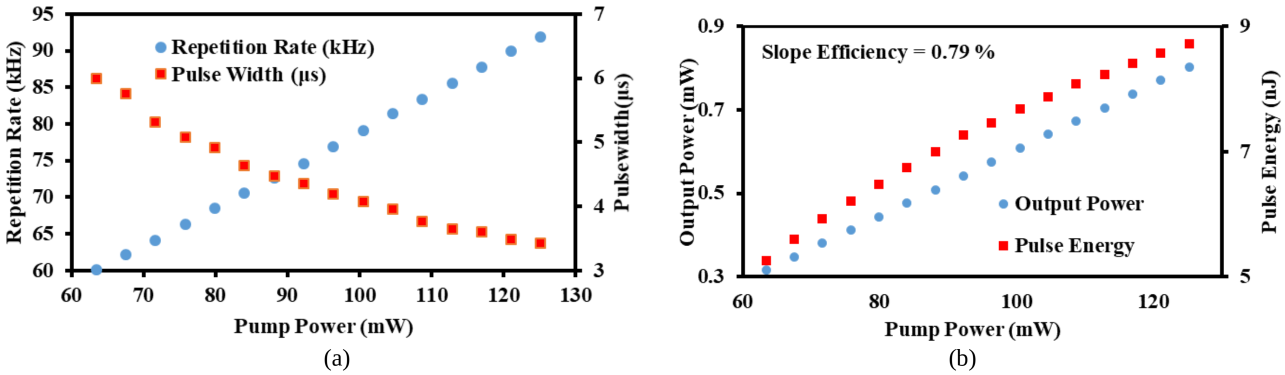



*Fig. 7. Relationship (a) Repetition rate and pulse width (b) Output power and pulse energy with incident pump power of the Q-switched EDFL (color online)*

## 5. Mode-locked laser operation for PVK SA

The laser starts to generate CW at the pump power of 63.4 mW. Soliton mode-locked laser was observed at the fundamental repetition frequency of the cavity (1.866 MHz) when a 100 m long SMF was added into the cavity. A stable self-starting mode-locked pulses were obtained within a pump power range from 125.2 mW to 187.0 mW. Fig. 8(a) shows the spectral profile of the output laser, centered at 1566.95 nm with a peak power of -12.91 dBm and full width at half maximum (FWHM) of approximately 0.97 nm. The spectrum displays soliton Kelly sidebands at 1564.50 nm and 1569.95 nm, demonstrating the formation of conventional soliton due to the balance between self-phase modulation (SPM) and dispersion effect inside the cavity. The soliton mode-locking was achieved in the anomalous dispersion regime.

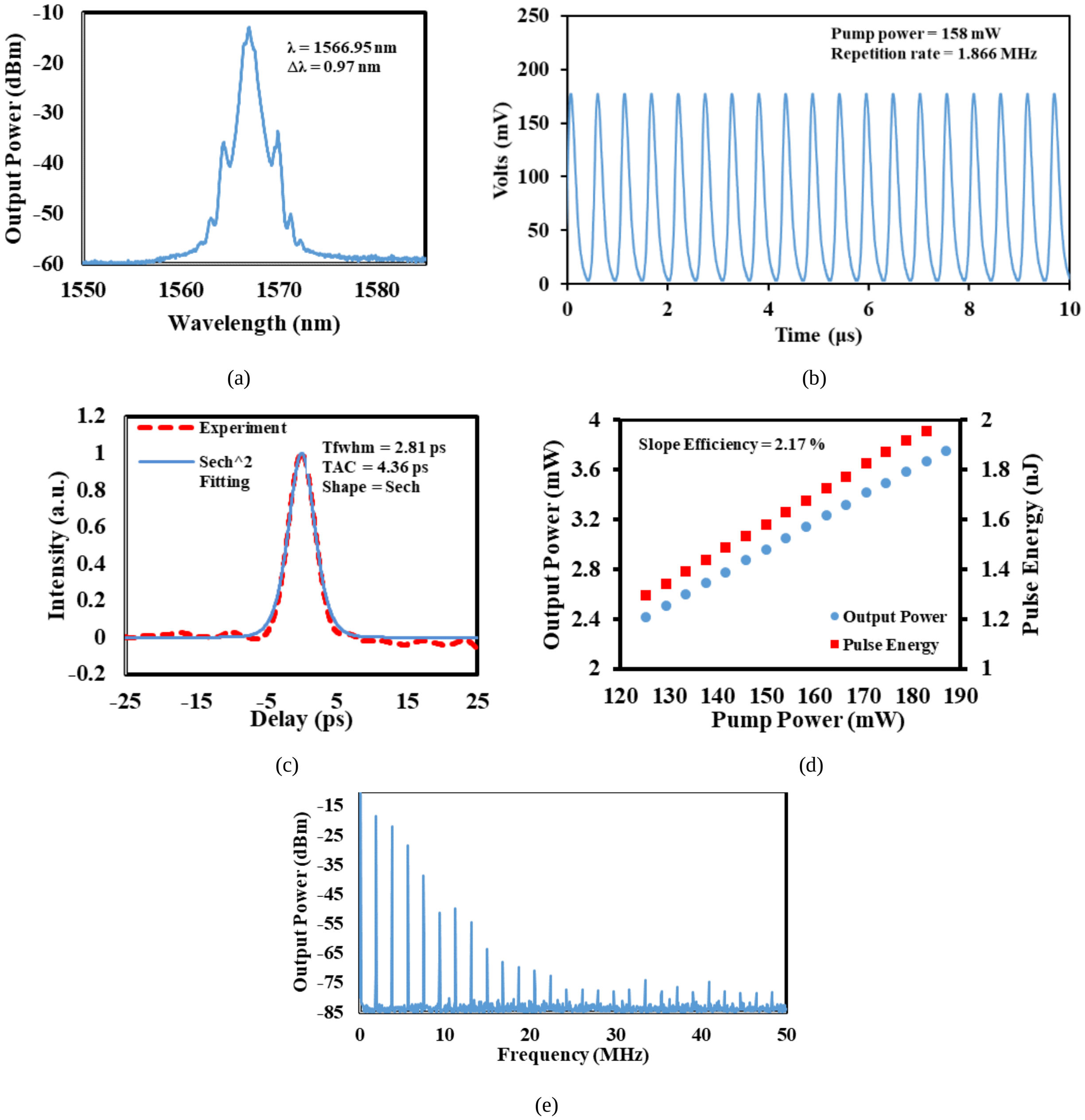



*Fig. 8. Mode-locking performances with PVK SA (a) OSA's spectrum (b) Typical pulse train (c) Autocorrelator trace (d) The output power and the pulse energy versus the input pump power (e) RF spectrum within a span of 50 MHz (color online)*

Fig. 8(b) illustrates a typical pulse train at the pump power of 158 mW, with a fundamental cavity repetition rate of 535.91 ns corresponding to a total cavity length of 111 m. The time interval between two pulses corresponds to the fundamental repetition frequency of 1.866 MHz. The autocorrelation instrument was used for the measurement of the pulse width of the soliton laser. Fig. 8(c) shows the experimental autocorrelation trace alongside a $sech^2$ fitting. The measured pulse width is 2.81 ps and the time-bandwidth product (TBP) is calculated at around 0.3329. The theoretical value of the TBP for $sech^2$ profile of mode-locked pulses is 0.315, and thus the result indicates that the pulse is a chirped pulse. From the experiment, as the pump power increased from 125.2 mW to 187.0 mW, the repetition rate remained at around 1.866 MHz, which confirms that the laser operates in the mode-locking state. Fig. 8(d) shows the linear relationships between the output power and pulse energy versus the input pump power. As the pump power was increased from 125.2 mW to 187.0 mW, the output power grew from 2.415 mW to 3.751 mW with a slope efficiency of 2.17 %. The corresponding pulse energy also increased from 1.294 nJ to 2.003 nJ. Fig. 8(e) shows the RF spectrum at the pump power of 187 mW with a span of 50 MHz. From the RF spectrum, we can see that the harmonic signals produced are more than 10 and the signal-to-ratio (SNR) is 63.10 dB with the fundamental frequency of 1.866 MHz. The mode-locking pulse train was stable.

## 6. Conclusion

In summary, stable passively Q-switched and mode-locked EDFLs have been successfully demonstrated using a PVK SA acting as both a Q-switcher and mode-locker. The modulation depth and the saturation power intensity of the prepared PVK film are measured at 4% and 202 $MW/cm^2$, respectively. A stable Q-switched laser operating at 1562 nm was achieved with the addition of the PVK SA in the laser cavity. Increasing the pump power from 63.4 to 125.2 mW caused the repetition frequency of the pulses to rise from 60.10 to 91.91 kHz, while the pulse duration reduces from 6.00 µs to 3.43 µs. The maximum single pulse energy of 8.73 nJ was obtained at the pump power of 125.2 mW. It is noted that a stable soliton mode-locked fibre laser was also realized by adding 100 m long SMF in the EDFL cavity. At the maximum pump power of 187.0 mW, the laser generated a pulse train with a repetition rate of 1.866 MHz, pulse energy of 2.003 nJ, pulse width of 2.81 ps and output power of 3.751 mW. The mode-locked laser had a high SNR of 63.10 dB, which indicates an excellent stability of the pulses. The high SNR ratio and long-term stability prove that the PVK SA is a good Q-switcher and mode-locker. This work introduces a new application of PVK organic semiconductor material to nonlinear photonics, emphasizing the application of a PVK SA in Q-switched and mode-locked fiber lasers.

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