

Realizing High-performance Organic Photovoltaics based on Active Layer Regulation

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Abstract: This article is based on the regulation of the active layer material, the regulation of the receptor by the active layer, and the regulation of the thickness of the active layer, achieving the optimal design of semi-transparent organic photovoltaics. Specifically, an efficient ternary system of PM6: BTP-eC9: L8-BO was used as the active layer. By adjusting the ratio of the three to 1:1:0.5 and the thickness of the active layer to about 75nm, a semi-transparent device with PCE=7.64% and AVT=44.4% was obtained, with the optimal LUE=3.39%.

Keywords: Semi-Transparent Devices; Organic Photovoltaics; Forward Structure; Active Layer Optimization.

1. Introduction

With the rapid development of modern industrial society, traditional fossil fuels such as coal, natural gas, and oil can no longer fully meet the energy needs of industrial production. [1-4] In addition, due to the unsustainability of fossil fuels and their pollution of the global ecological environment, people urgently seek green, large-scale, and sustainable new energy to gradually replace these old energy sources. As the most important solar energy utilization technology, solar photovoltaic technology has made a leap forward in recent decades due to its effectiveness, reliability, greenness, and huge development potential. Currently, the mainstream solar cells in the scientific research community include: silicon crystal solar cells, perovskite solar cells, organic solar cells, etc. Organic solar cells use organic molecules as active layers to absorb light energy and convert it into directionally moving charge carriers. [5-7] It has the characteristics of adjustable absorption spectrum, lightweight, and flexibility, which is conducive to the preparation of semi transparent organic photovoltaics (ST OPVs). The main feature of this type of battery is that it can absorb the remaining light energy and convert it into electrical energy while transmitting as much visible light as possible. Therefore, the main use of semi transparent solar cells is to integrate them into buildings, such as urban high-rise buildings, greenhouses, household windows, and even car skylights. They can not only fully utilize the building area to achieve solar energy conversion, but also achieve normal lighting and crop growth through visible light. To improve the performance of semi transparent solar cells, it is mainly necessary to fully transmit visible light and fully absorb other light energy, and convert them into electrical energy. However, these two indicators require light transmission and light absorption, which conflict with each other. How to balance this contradiction and achieve the highest comprehensive efficiency is the biggest challenge in researching semi transparent solar cells. [8-11]

The structure of organic photovoltaics usually consists of glass bottom electrode transport layer 1 active layer transport layer 2 top electrode. Light usually enters from the bottom electrode, so the bottom electrode is required to be transparent. The transport layer is divided into an electronic transport layer (ETL) and a hole transport layer (HTL). [12-13] Taking

the electron transport layer as an example, its main function is to modify adjacent cathodes, facilitating the smooth transfer of electrons from the active layer to the cathode, while also hindering the reverse movement of holes. Similarly, the hole transport layer is also known as the interface layer. The structure of organic photovoltaics can be divided into traditional structure and reverse structure, where the bottom electrode of the traditional structure is the anode, that is, glass anode hole transport layer active layer electron transport layer cathode; The bottom electrode of the reverse structure is the cathode, that is, the structure is: glass cathode electron transport layer active layer hole transport layer anode. Both traditional and reverse structures typically use silver tin oxide (ITO) with excellent transparency and conductivity as the bottom electrode; The top electrode includes silver (Ag), aluminum (Al), gold (Au), etc., with Ag being the most commonly used. [14-16] The hole transport layer material in traditional structures is mainly PEDOT: PSS, while in reverse structures, molybdenum trioxide (MoO₃) is commonly used for the hole transport layer; The most commonly used electronic transport layer material in traditional structures is PDINN, while ZnO is commonly used in reverse structures. The active layer material usually uses two types of organic molecular materials, which have the characteristics of electron donating and electron accepting, respectively, called donor materials and acceptor materials. The two are mixed with each other through a solution, and then formed into a film through processes such as spin coating, scraping coating, and spraying, forming an interpenetrating structure. [15, 17-19] When sunlight is incident on the active layer, electron hole pairs, known as excitons, are generated. Excitons move to the interface between the donor and acceptor due to diffusion principles. Due to the energy level difference between the donor and acceptor and exciton dissociation, an endogenous electric field is formed due to the difference in work function between the cathode and anode. Electrons move directionally to the cathode, while holes move directionally to the anode, thereby generating photocurrent. [20-22]

Translucent organic photovoltaics are organic photovoltaics that possess both light energy conversion and partial visible light transmission characteristics. To achieve transparency, it is usually required that both the active layer

and the top electrode are transparent. For the active layer, the absorption spectrum of the active layer can be regulated by changing the type and proportion of the receptor, thereby reducing the absorption of visible light and achieving this by reducing the thickness of the active layer. However, the energy conversion efficiency of the device will inevitably decrease. Therefore, for semi transparent organic photovoltaics, the contradiction between visible light transmission and conversion efficiency improvement is a difficult problem that needs to be carefully balanced. The most important evaluation indicators for semi transparent organic photovoltaics are the average visible light transmittance and energy conversion efficiency. To improve these two parameters, targeted design of the structure, materials, and processes of semi transparent organic photovoltaics is required. The bottom of semi transparent organic photovoltaics usually uses high transmittance ITO glass. The electron transport layer and hole transport layer have very little effect on light transmission due to their ultra-thin thickness and weak absorption. Therefore, optimizing the active layer is the key to improving its performance. [23-25]

This article is based on the regulation of the active layer material, the regulation of the receptor by the active layer, and the regulation of the thickness of the active layer, achieving the optimal design of semi transparent organic photovoltaics. Specifically, an efficient ternary system of PM6: BTP-eC9: L8-BO was used as the active layer. By adjusting the ratio of the three to 1:1:0.5 and the thickness of the active layer to about 75nm, a semi transparent device with PCE=7.64% and AVT=44.4% was obtained, with the optimal LUE=3.39%.

2. Device Preparation and Performance Characterization

2.1. Introduction

The active layer donor molecule used in this chapter is PM6, and the receptor molecules are BTP-eC9 and L8-BO. The molecular structures of the three are shown in the following figure. PM6 is a typical representative of high-performance wide bandgap polymer donor materials in organic photovoltaics. They can self assemble to form nanofiber structures during film formation, which helps to form a dual continuous fiber network active layer morphology conducive to hole and electron transport. Their absorption spectra are mainly located between 300-600 nm. BTP-eC9 is a high-performance non fullerene small molecule receptor material obtained by chlorine substitution of star small molecule receptor Y6. It can form a good blend film morphology and energy level matching with PM6, and the absorption spectrum range is mainly 600-850 nm. L8-BO is another Y6 derivative, characterized by the introduction of long branched side chains to enhance the interaction between alkyl side chains. L8-BO can not only act as a separate receptor to form a good dual fiber network morphology with PM6, but its absorption spectrum is similar to BTP-eC9, but its absorption peak is shifted by about 30nm relative to BTP-eC9. L8-BO can also be added as a third component to PM6: BTP-eC9 to improve the crystallinity and crystal size of the active layer film, thereby achieving better performance. At present, the conversion efficiency of binary organic solar cells based on PM6: BTP-eC9 as the active layer has exceeded 18%, while the corresponding three component PCE has exceeded 19%. When the ratio of PM6: BTP-eC9 is maintained at 1:1.2, the optimal PCE of the device is achieved due to the good

crystalline state between the donor and acceptor, as well as the maximum superimposed light absorption. However, this article takes ST OPVs as the research object and needs to comprehensively consider PCE and AVT, with the goal of maximizing LUE as the guiding principle. In theory, the influencing factors of AVT not only include transmittance, but also are related to the human eye response factor P of each band. When the visible light band is between 500-600 nm, $P > 0.4$. Therefore, reducing the content of PM6 to increase the transmission of 500-600 nm wavelength light, which greatly helps improve AVT, and increasing the content of BTP-eC9 to compensate for the PCE decrease caused by the transmission loss in the main absorption region are reasonable measures. Of course, an excessively imbalanced donor acceptor ratio (D: A) can lead to poor phase separation in the active layer and a decrease in carrier dissociation efficiency, resulting in more disadvantages than advantages. Therefore, it is necessary to regulate the donor acceptor ratio reasonably, and further consider the comprehensive impact of the addition of L8-BO on the AVT and PCE of the device.

Adjusting the thickness of the active layer is a necessary task for constructing high LUE semi transparent organic photovoltaics. In general, the thicker the active layer, the more it absorbs incident light, the higher the exciton generation rate, and the more carriers are generated. This results in a higher short-circuit circuit density of the device, allowing it to achieve higher PCE. However, at the same time, due to a decrease in visible light transmission, the AVT of the device will decrease. Therefore, it is necessary to adjust the thickness of the active layer to achieve the optimal balance between AVT and PCE.

The device structure constructed in this section is ITO/PEDOT: PSS/PM6: BTP-eC9/PDINN/Ag. When regulating the active layer, other experimental conditions should be kept consistent. This structure belongs to the traditional forward structure, with ITO as the anode and light incident from the ITO surface.

2.2. Device Preparation Process

(1) Cleaning of glass/ITO, quartz sheets, and glass sheets

Use glass detergent, deionized water, acetone, and isopropanol as cleaning agents to clean in order in an ultrasonic cleaning machine. The cleaning time for ultrasonic cleaning is 15 minutes. Blow dry the washed ITO glass substrate with a nitrogen gun, and then. Pre-cleaned ITO glass substrates were treated with a UV ozone cleaner for 15 minutes to increase the work function and surface area of ITO

Immersion.

(2) Hole transport layer

PEDOT: PSS is used as a hole transport layer, and PEDOT: PSS solution is spin coated on ITO at a speed of 4000 rpm. On the glass, spin coat for about 30 seconds, then anneal at 150 °C in air for 15 minutes, with a film thickness of 30 nm.

(3) Active layer

Dissolve in chlorobenzene at a ratio of PM6: BTP-eC9: L8-BO=1:1:0.5 (w/w), with a total concentration of 20 mg ml⁻¹. Then 0.5% vol of 1,8-diiodooctane (DIO) was added as an additive, and the solution was stirred at 65 °C for 2 hours to completely dissolve. In a glove box filled with nitrogen, the active layer was spin coated onto the PEDOT: PSS layer at 5000rpm for about 40 seconds, and then annealed at 100 °C for 10 minutes to obtain a ternary active layer film with a thickness of about 75nm.

(4) Electronic transport layer

PDINN was used as an electron transport layer and dissolved in methanol solution to prepare a concentration of 1.0mg ml⁻¹. Spin coat the active layer at a speed of 3000rpm for 30 seconds to form an electron transport layer with a thickness of 10nm.

(5) AgNWs electrode

For the top electrodes, original AgNWs, AgNWs-PD (10:1) and AgNWs-PC (3000 rpm) are prepared according to the same way of AgNWs films fabrication.

2.3. Characterization Method

Use TU-1810 UV visible spectrophotometer to measure the absorption spectrum of the active layer and the transmission spectrum of the device (300-1000 nm). Use a probe-based surface profilometer to test the thickness of each layer.

Measure the J-V curve of the device using a computer-controlled Keithley 2400 light source under AM 1.5G illumination (100 mW cm⁻²). The measurement unit and calibrated solar simulator (Enli Technology Co., Ltd.) were used, and no bias voltage or light homogenization process was applied before J-V measurement. Use a standard silicon secondary reference cell cover with KG-5 filter to calibrate the illumination intensity, and provide a value of 100 mW cm⁻² during testing. All J-V curves are tested by scanning the J-V measurement values in the forward direction (from short circuit to open circuit) with a step size of 0.02V, and the delay time is set to zero.

The measurement of External Quantum Efficiency (EQE) spectra was carried out using the QE-R3011 spectral response measurement developed by Enlitech Co., Ltd. equipped with standard silicon diodes.

Use an ellipsometer to test the refractive index (n, k values) of each layer of the device (Glass/ITO/PEDOT: PSS/active layer/PDINN/AgNWs), with a testing range of 370-1000nm and an incident angle of 65.17°.

3. Results and Discussion

3.1. Proportion Control of Active Layer

Table 1 shows the performance parameters of ST OPVs devices with different D:A proportion of active layers. As shown in Figure 1a,1b, 1c, based on the J-V curve of the device, it can be analyzed that as the content of donor PM6 in the active layer of the device decreases, the open circuit current density of the device decreases significantly. Moreover, when the ratio of PM6: BTP-eC9 reaches 1:2.2, the VOC, JSC, and FF values of the device all decrease, especially JSC and FF values, which decrease to 10.75 mA cm⁻² and 59.5%, respectively. This may be due to the poor phase separation of excitons caused by excessive BTP-eC9 content when forming a blend film with PM6, resulting in poor exciton generation and separation efficiency. Ultimately, the PCE of the device decreases from 7.95% to 5.63%, a decrease of about 29.2%. From the normalized absorption curve of the active layer, it can be seen that the absorption peaks of the active layer are mainly at 625nm and 830nm, with valleys at 445nm and 680nm. The peak values of the transmittance curves of each device are roughly 450nm and 675nm, with valleys at 625nm and 830nm, respectively. The peaks and valleys of the absorption curve and transmittance curve complement each other perfectly, indicating that the absorption of light by the active layer is a key factor affecting the transmittance of the device, and also proving the rationality and reliability of the data. Observing the changing trend from different D: A ratios, it can be concluded that as the proportion of PM6 decreases, the device's absorption of visible light decreases, which indeed leads to an increase in the device's transmittance. When D: A decreases from 1:1.2 to 1:2.2, the AVT of the device increases from 34.92% to 48.58%, with an increase of about 39.1%. Finally, the device based on PM6: BTP-eC9=1:1.5 achieved an AVT of 44.08% at a PCE of 7.36%, obtaining the highest LUE value of 3.14%. Therefore, PM6: BTP-eC9=1:1.5 is used as a reference for subsequent real conditions.

Table 1. The parameter of ST-OPVs based on different PM6: BTP-eC9 (D: A) proportion with 75 nm active layer and 50 nm AgNWs

D:A1(: A2)	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)	AVT (%)	LUE (%)
1:1.2	0.80±0	14.99±0.13	62.87±2.82	7.95 (7.63±0.32)	34.92	2.78
1:1.5	0.80±0.01	13.63±0.19	62.87±3.71	7.36 (7.02±0.34)	44.08	3.24
1:1.8	0.80±0.01	12.86±0.19	62.20±3.4	6.82 (6.44±0.38)	45.28	3.09
1:2.2	0.80±0.01	10.75±0.43	59.50±3.4	5.63 (5.23±0.36)	48.58	2.74

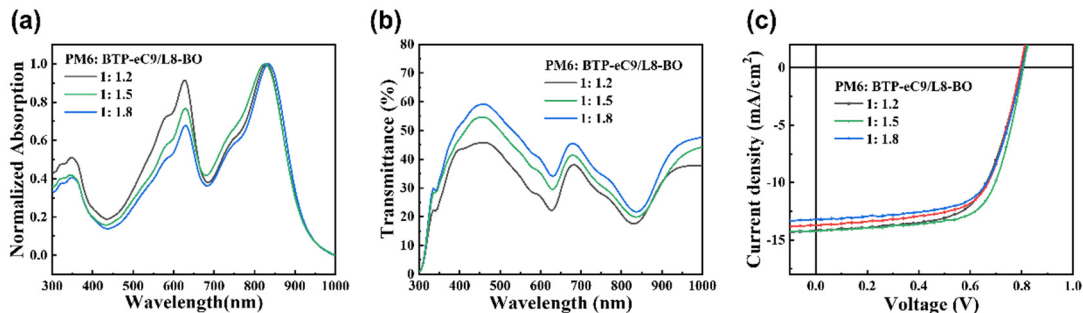


Figure 1. (a) Absorption curve, (b) transmittance curve and (c) J-V curve of ST-OPVs based on different PM6: BTP-eC9 proportion with 75 nm active layer and 50 nm AgNWs

3.2. Control of Active Layer Thickness

Table 2 shows the various performance parameters of ST OPVs under different thicknesses of active layers. By combining the J-V curve and transmittance curve, it can be clearly seen that as the thickness of the device decreases, the absorption of incident light by the active layer decreases, resulting in a gradual decrease in the JSC of the device. At the same time, the transmittance of each wavelength band of the

device increases, making the AVT of devices with thinner active layers higher. In addition, the open circuit voltage V_{OC} and fill factor FF of the device remain basically unchanged, so the PCE of the device will decrease with decreasing thickness. The final device achieved the best balance between AVT (=44.08%) and PCE (=7.31%) at an active layer thickness of 75nm (corresponding to a spin coating speed of 5000r), with a LUE of 3.22%.

Table 2. The parameter of ST-OPVs based on different active layer thicknesses with PM6: BTP-eC9 = 1: 1.5 and 50 nm AgNWs.

Active layer thickness (nm)	V_{OC} (V)	J_{SC} (mA/cm ²)	FF (%)	PCE (%)	AVT (%)	LUE (%)
110	0.81±0	17.23 ±0.46	63.60 ±4.10	9.52 (8.89±0.60)	31.38	2.99
90	0.78±0.02	15.78 ±0.60	63.60 ±2.16	8.33 (7.82±0.49)	37.26	3.10
80	0.80±0.01	14.25 ±0.47	63.66 ±1.63	7.41 (7.25±0.12)	41.46	3.07
75	0.80±0.01	13.43 ±0.19	65.87 ±0.71	7.31 (7.12±0.15)	44.08	3.22
70	0.80±0	12.74 ±0.36	62.47 ±0.61	6.67 (6.33±0.18)	45.89	3.06
65	0.79±0.01	12.34 ±0.18	64.46 ±1.23	6.53 (6.25±0.17)	46.65	3.05

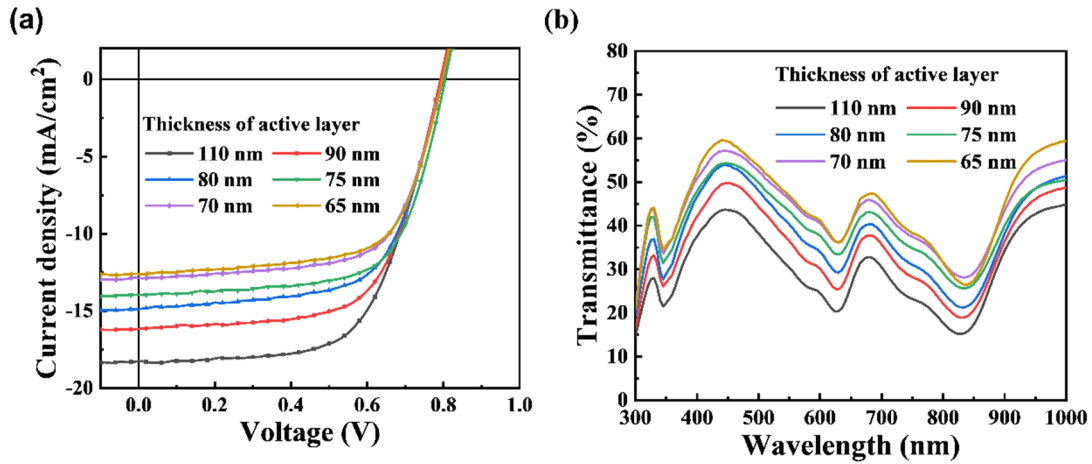


Figure 2. J-V curve (a) and transmittance curve (b) of ST-OPVs based on different active layer thicknesses with PM6: BTP-eC9 (1: 1.5) and 50 nm AgNWs.

3.3. Ternary Strategy

In order to compare the performance of binary and ternary components while maintaining consistent donor and acceptor concentrations, we selected PM6: BTP-eC9=1:1.5 as the control binary semi-transparent device with the best performance, and replaced 1/3 of BTP-eC9 with L8-BO, i.e. PM6: BTP-eC9: L8-BO=1:1:0.5 as the experimental group of ternary semi-transparent devices, while keeping the concentration of PM6 in the active layer unchanged and the total concentration unchanged. The performance parameters of the device are shown in **Table 3**. Specifically, compared to the two components, the V_{OC} of the three components with L8-BO added increased from 0.81V to 0.82V, the current density increased from 13.63mA cm⁻² to 13.82mA cm⁻², and the FF value increased from 66.08% to 67.20%. Increasing the PCE of the device from 7.36% to 7.64%. The absorption

curve of the receptor film alone reveals the key reason for the increase in device current density: L8-BO has a more purple shifted absorption than BTP-eC9, which complements the absorption of PM6 and BTP-eC9. Therefore, adding L8-BO to the PM6: BTP-eC9 mixture increases the absorption coefficient of the ternary mixture, which helps to increase light harvesting and thus improve photocurrent. The absorption curve of the active layer indicates that the ternary active layer has higher absorption in the near red region (650-800nm) than the binary component, confirming the correctness of the above statement. In addition, the three components have almost negligible reductions compared to the two components (AVT decreased from 44.8% to 44.4%). The energy level arrangements of the three materials are shown in **Figure 3**, where L8-BO has a lower highest occupied molecular orbital (HOMO) energy level of -5.72 eV compared to BTP-eC9, and in terms of the lowest unoccupied

molecular orbital (LUMO) energy level, compared to BTP-eC9 (-3.98 eV), L8-BO's LUMO (-3.90 eV) shifts upward. The well-arranged energy levels formed by PM6, L8-BO, and

BTP-eC9 can ensure effective exciton dissociation and charge transfer.

Table 3. The parameter of ST-OPVs based on ternary or binary system of active layer with 75-nm active layer and 50-nm AgNWs.

PM6: BTP-eC9:L8-BO	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)	AVT(%)	LUE(%)
1:1.5:0	0.81 (0.80±0.01)	13.28 (13.14±0.19)	65.08 (62.37±3.71)	7.16 (6.72±0.28)	44.8	3.21
1:1:0.5	0.82 (0.815±0.005)	13.82 (13.60±0.22)	67.2(64.7±2.5)	7.64 (7.18±0.44)	44.4	3.39

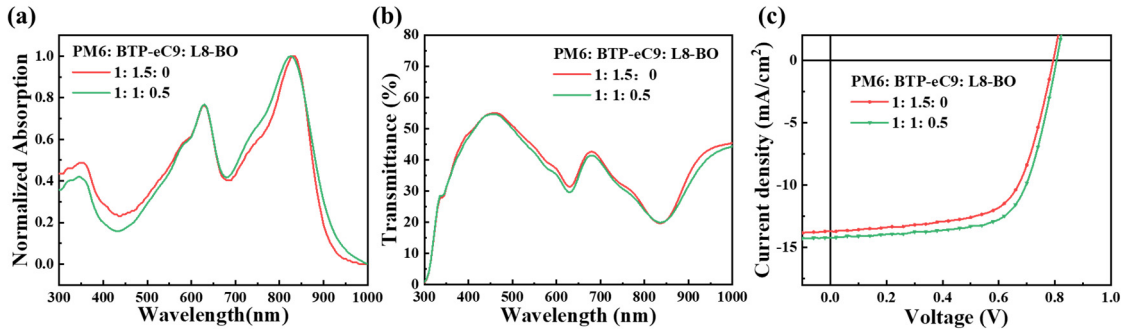


Figure 3. (a) J-V curve, (b) EQE curve, (c) transmittance curve of ST-OPVs based on ternary or binary system of active layer with 75-nm active layer and 50-nm AgNWs.

4. Summary

All devices in this chapter are based on traditional structural design, namely glass/ITO/active layer/DINN/AgNWs thin film, where the active layer adopts a stable and efficient PM6: BTP-eC9 (: L8-BO) system (Figure 3b). In order to optimize the performance of ST OPVs through active layer regulation, we comprehensively optimized the PCE and AVT indicators of the device from three perspectives: the ratio of active layer to receptor (D: A), the thickness of the active layer, and the ternary strategy. The conclusions are as follows:

(1) When the total concentration of the receptor remains constant while the PM6 content increases and the BTP-eC9 content decreases, the visible light absorption of the device decreases, the PCE decreases, the transmittance increases, and the AVT increases. Finally, when the PM6: BTP-eC9=1:1.5 is used, the PCE of the device is 7.36%, the AVT is 44.08%, and the highest LUE value of 3.14% is obtained.

(2) When the thickness of the active layer of the device decreases, the absorption of full band light by the device decreases, resulting in an increase in PCE and a decrease in AVT. Finally, the device achieved the best balance between AVT (=44.08%) and PCE (=7.31%) at an active layer thickness of 75nm (corresponding to a spin coating speed of 5000r), with a LUE of 3.22%.

(3) When using L8-BO instead of a small portion of BTP-eC9 to form a three-component device, the crystal morphology of the active layer is improved, and the device has improved in VOC, JSC, and FF values. Finally, based on the three component ST-OPV, when PCE=7.64%, AVT=44.4%, and the best LUE=3.39% are obtained.

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