### Solar Photovoltaics: Fundamental Technology and Applications Prof. Kushal Soumitra Satapathi Department of Physics Indian Institute of Technology-Roorkee

# Lecture-24 Morphology Optimization of Organic Solar Cells

Welcome everyone, so the last 2 weeks we have been discussing about the organic solar cells every important varieties of the third generation solar cell. Now so for whatever we have learnt is that what are the different components of an organic solar cell, how to fabricate an organic solar cell and also the basic photo physics behind an organic solar cell. Now in the context of an highly efficient solar cell we have mention that the efficiency of a solar cell is a product function.

It depends upon lot of parameters, the first thing it depends upon excite on generation for that we mention in the last class that the lot of researchers are working to design low band gap polymers which can absorbs the light in a near IR range. Now the second stage in the solar cell application was the excite on diffusion and excite on dissociation. Now we have mention that this excite on diffusion and excite on dissociation that is related to the morphology of the thin films.

Now this morphology plays an very important role in determining the high efficiency in an organic solar cell. For example let us say you have a ideal material which can absorb the light in a near IR range like silicon kind of material or aluminum kind of material which has a band gap of 1.1 electron pole or 0.7 electron pole. But what will happen when I make the film, so there are lot of defects and traps in this material, this material is not well soluble.

So that it we do not get a bicontinuous phase separated percolated network throughout the film, so that means the morphology of this film is not good, so while you design a solar cell, so not only the low band gap materials or the proper energetics between the donor and acceptor but it is also important what is the morphology of the films. So today we will discuss about how the morphology of the films affect the device and what are the protocols to optimize the morphology of a device.

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# Morphology Optimizations of Organic Solar Cells

- The morphology of the photoactive layer in OPVs has strong influences on light absorption, exciton dissociation, charge transport, and charge recombination, so it determines their overall photovoltaic performance.
- Early-stage bulk-hetero junction (BHJ) layers were found to exhibit low exciton dissociation and charge transport efficiency, and thus much research was dedicated to optimizing the Nano scale BHJ morphology by controlling various morphological factors such as the phase separation and material crystallinity.
- New organic semiconductors and processing solvents have been designed and various treatments have been adopted with the aim of optimizing BHJ morphologies.

Now morphology optimizations of an organic solar cell, the morphology of the photo active layer OPVs has strong influence on light absorption, excite on dissociation, charge transport and charge recombination. So it determines the overall photovoltaic performance, early stage bulk-hetero junction layers were found to exhibit low excite on dissociation and charge transport efficiency and thus much research was dedicated to optimize the nano scale BHJ morphology by controlling various morphological parameters.

Such as the phase separation and material crystallinity, new organic semiconductors and processing solvents have been designed and various treatments have been adopted with the aim of optimizing BHJ morphologies. Now the first question is like what do we mean by an optimum morphology, so by optimum morphology we mean a bicontinuous phase separated percolated network by continuous.

Because throughout we want to see donor acceptant nano interface, phase separated we want to separate the donor phase and acceptor phase and also percolated. Because whenever we have a charge separation those electrons and holes needs to travel to the electrode. So the travel to the electrode through a percolating network, so that determines the optimum morphology.

Now there are lot of research has been ongoing to change the solvent to change the film, raising temperatures or like in a donor to acceptor ratio, so many so different things to optimize this morphology.

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Moreover for OPVs to be operationally stable, the optimized BHJ morphology must be maintained. So not only the charge transport but for the stability of the overall device the morphology also plays an important role. Because we have earlier mention that morphology determines the photo physics and if there is a polar degradation that can be always correlated to a bad morphology of the thin film.

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#### Morphology Control

#### \* Material Design for Morphological Control

- The materials used in OPVs affect the Nano scale morphology of the photoactive layer its light absorption, exciton dissociation, and charge transport abilities.
- The properties of the materials depend on their structural components (i.e. monomers, side chains, end groups) and on their physical properties (e.g., molecular weight, poly dispersity index, regio-regularity, and planarity).
- The materials must have a low band gap and a deep-lying highest occupied molecular orbital (HOMO) to broaden the light absorption range and the built-in potential.



The first is the material design for morphological control, the material used in OPVs affect the nano scale morphology of the photoactive layer, that is light absorption excite on dissociation and charge transport abilities. The properties of the materials depend on their structural components for example monomers, side chains, end groups etc. and on their physical properties for example molecular weight, polydispersity index, region-regularity and planarity.

The materials must have a low band gap and a deep lying highest occupied molecular orbital HOMO to broaden their light absorption range and the built-in potential.

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The carrier mobilities of the PV materials must be controlled to reduce charge recombination during charge extraction and to increase fill factor ok. So I mean since we have electrons and holes we have to different kinds of mobility, electron mobility and hole mobility. And this kind of charge carrier mobility should be such that it can reduce the recombination rate because if there is a recombination then that is a loss in the circuit.

And also we should enhance the charge extraction and since all of these is related to the performance of a device which is reflected by the film factor, so the overall objective of increasing charge carrier mobility is also to increase the fill factor. Second is that to enhance the morphologies of the blend films PV materials require processability that is solubility in organic solvents and miscibility with other organic semiconductors.

Now as I mention that we develop a excellent organic semiconductor which absorbs the light in the near IR range. But that material is not soluble in most of the organic solvent, so what that material will do if we wanted to make a solution processable thin films. So that is why the solubility is very very important and also whatever the material which we have designed that should also mixed well with the acceptor material, so the miscibility also an important parameter.

Solubility can be achieved by modifying the functional side chains of the material by breaking the planarity of other interactions in the molecule structure and by varying the molecular weight of the polymer to impede self-aggregation. Now why the solubility is affected, if there are lot of aggregation then there will be like you know problem in the solubility. Now it is worthwhile to mention that the most of the time you can check the solubility by an experimental method called dynamics light scattering which is abbreviated as DLS.

DLS stands for dynamic light scattering, now here what usually is done a laser beam falls on the material and now let us say I have a solvent and I dissolve some kind of material inside it. So the material undergoes a random Brownian motion, now whenever light beams falls on it so material will diffract that light. Now if I put it detector at a 90 degree angle to the solution so the detector will count the photon beam which is reflected or scattered from this particle.

Now from this time gap between the successive particles we can calculate the hydrodynamic radius or the particle diameters of the solvent. Now if most of the cases if the hydrodynamic radius is very very small which you usually absorb in the case of solution but if the hydrodynamic radius is very very large and it is very very poly disperse. So you will see that your solution is not a solution rather it is a dispersion.

Now this solubility usually is achieved by adding some side groups, for example let us say I have C60, this is not soluble in organic solvent but if I put a side group then we have PCBM then this is soluble in organic solvent. Similarly like you know if the polymer structure is such that it interact with each other and aggregates, so then our swab should be like to destroy the structure in such a way the planarity is maintained.

So it would destroy the structure in such a way the aggregation can be prevented, so these are all the thumb rules for increasing the solubility of a material. To achieve the appropriate morphology for efficient exciton dissociation and charge transport some intermediate level of miscibility of the active layer components is required, yeah. As you mention that miscibility with the acceptor layer is also important, so not only the donor solubility but how does it mix with the acceptor that actually determines the phase separation.

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The donor acceptor composition, the ration of the donor polymer to the acceptor significantly influence the electrical properties of the blind film. The morphology of the active layer and the photovoltaic performance of the associated cell. For example in earlier days people use to use P3HT and PCBM at 1:1 wet ratio but if you make a device using 1:1 ratio or if an make a film with 1:1 ratio and if you make a film with 1:2 and 1:4 ratio a P3:PCBM.

You will see that in the latest case 1:4 ratio the morphology is way different then 1:way ratio. So that is why the 1:4 ratio is preferred to fabricate a device because it leads the high efficiency why because it gives a very good morphology. A good phase separated by continuous network which is often proved by atomic force microscopy or A film an optimized donor acceptor ratio in the BHJ film guarantees the high crystallinity of components bicontinuous percolation and interconnected pathways.

These are very very important terminologies, so basically a good morphology implies a high crystallinity of the components bicontinuous percolation and interconnected pathways for the charge carriers and enables the optimizations of the carrier dissociation, transport and extraction. The morphology and cell efficiency of the MDMO-PPV to PC60BM BHJ blend systems can be controlled by adjusting the concentration of PC60BM.

As I mention that if I take the polymer:PCBM ratio as 1:1 or if I take a ratio 1:4 the morphology of the resultant films will be way different. And the efficiency can also be controlled just by simply changing the donor to acceptor ratio.

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Varying the compositions of a blend system can change the crystalline features of the components. So basically how the acceptor molecule will crystallize or aggregates in presence of the donor material that can be dominated or that is determined by their weight ratio. In a BHJ film with P3HT and the acceptor with di-n-propylated dithienylsilole unit, a persistent glassy solid forms over a range of hypoeutectic compositions with respect to P3HT.

So this blend system requires a higher acceptor ratio than the other to form electron percolation paths in the crystalline acceptor network.

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Second important parameters is solvent engineering, now if we change one solvent to other solvent what basically we are changing the dielectric constant. Now you know that dielectric constant is related to the polarity of the solvent. So basically when we change from one solvent to another solvent we change the polarity of the solvent. Now if the polarity of the solvent is different, then for the same material solubility will be different so that is why the solvent choice is very very important for making a good thin film.

Now the morphology of an active layer varies dramatically with the solvent that is used for it is depositions. The solubility and volatility of the solution system depend on the processing solvent, the dependence can be associated with dramatic changes in the solution state molecular confirmation and in the kinetics of drying film. So what do we mean by the kinetics of the drying film, let us say I have a P3HT polymer and I am spin coating this polymer from chloroform solvent.

And in another case I am spin coating it from dichlorobenzene solvent, now chloroform has a very boiling point 65 degree Celsius. So at some temperature it will start evaporating. Now the way the film P3HT film will dry if I use a chloroform as a solvent and if I use the dichlorobenzene as a solvent will be way different. So that is why the kinetics of the drying films will be different and if that kinetics is different, then the way the polymers chains will be packed that will be also be different.

That means the morphology will also be different. Mixture of the solvents can also used to fine tune the active layer morphology very often instead of 1 solvent we use 2 different solvent. Now these 2 solvents can be miscible to each other or they can be immiscible to each other. If the 2 solvents are immiscible to each other then one is called the anti-solvent to the others, now use of the anti-solvent also helps to crystallize the film. Solvent engineering strategy is important for the control of the morphology of active layers.

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### Solvent Engineering

#### Effects of Solvent Selection

- The morphology of a solution-processed organic active layer depends critically on the solvent used.
- The degree of crystallization and the phase separation of the film are strongly affected by the choice of the solvent.
- The evaporation rate of the solvent determines the time required for the film to dry. If the solvent resides for a long time in the film, then there is sufficient time for the donor and acceptor molecules to migrate to form a thermodynamically stable morphology.

Now what are the effects of solvent selection, the morphology of a solution processed organic layer depends critically on the solvent used. The degree of crystallizations and the phase separation of the film are strongly affected by the choice of the solvent. The degree of crystallization and the phase separation and I said that as the solvent is different the dielectric constant is different if the dielectric constant is different then the polarity is different.

Now we can choose 2 solvent where the dielectric constant difference of the polarity difference is small. And we can choose 2 solvent where the dielectric constant difference or the polarity difference is very very large. In this 2 case the morphology evolutions will be completely different because the way the materials will crystallize and the degree of the drying the film which can lead to the phase separation will be completely different in this 2 different case.

The evaporation rate of the solvent determines the time required for the film to dry if the solvent resides for a long time in the film. Then there is sufficient time for the donor and acceptor molecule to migrate to form a thermodynamically stable morphology. So basically if we want the donor and acceptor to interconnected between each other to make a interconnected network, then what will happen.

Then we we want the solvent to stay in the system for a more time, so obviously we should not choose a solvent whichever lower boiling point. So we should choose a solvent which have a higher boiling point and that exactly it was observed while making P3HT PCBM device if you use a temperature of 65 degree Celsius boiling point solvent. For example if you make a P3HT PCBM blend device from chloroform and on the other hand if you make it from a dichlorobenzene which has a much higher boiling point and the chloroform the film morphology is way better than the chloroforms.

Because now the possibility of getting a thermodynamically stable morphology is much higher in the case 1, 2 dichlorobenzene in comparison to the chloroform.

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If evaporation is completed in a very short time, the fast removal of solvent can immobilize the donor and acceptor molecule and the morphology can become kinetically frozen in some intermediate state yeah. So if the solvent evaporates very very rapidly, so before it reaches a

thermodynamically equilibrate state it will be frozen in some intermediate state, it will be kinetically frozen in between some intermediate state.

So probably the complete transformation of bikinetics or phase separated bicontinuous network which we really like to achieve that cannot be achieved by using a very low boiling point solvent. These are phenomena are well illustrated by the case P3HT, a film that is spin coated from CF contains P3HT nanocrystallites with size less than 20 nano meter that are dispersed in a disordered phase and have poor interconnectivity.

In contrast a film spin coated from DCB dichlorobenzene CF stands for a chloroform, dichlorobenzene develops P3HT crystals with typical size in the 80 to 100 nanometer that form through the growth and interconnection of the crystallities. So basically we choose the same material P3HT, if we spin coat the material from the chloroform we get a crystallities size less than 20 nano meter.

But if we make use the same material from a DCB or dichlorobenzene solvent we usually get a crystallite size 80 to 100 nano meters and that is because of the growth and interconnection of the crystallities. These differences in the crystal growth of P3HT have been attributed to the different evaporation speeds of the 2 solvent. As I mention that the why the 2 solvents evaporates that is completely different.

If their boiling point is different then one of the solvent will lead faster than the other solvent, so if the solvent leads very very in a faster way. So they will not give the enough time for the material to crystallize what to grow larger crystal grain. Whereas if the solvent evaporates from it is slow then the kinetics of the crystal growth is become more control and you get a larger crystallities.

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The next important what is the solvent additives, the term solvent additives is used when the solvent added to produce a mixed solvent system has both a high boiling point and a selective solubility for the donor and acceptor molecules. The high boiling point or low volatility of the solvent additives means that the solvent additive persists for a long time in the film during the development of it is morphology.

As again like you know we mention that the strategy should be to choose a high boiling point or low volatile solvent. So this has been added in addition to our solvent to make sure we get a better morphology and this is called solvent additives. So in addition to the solvent from which we are fabricating the film we can also add an extra solvent which is called solvent additives and that extra solvent has the property that it has a high boiling point and low volatility, then it always leads to a better quality film.

The solvent additive interact with the organic semiconductor molecules within the active layer film and these extended interaction alters the films nanostructures.

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Now non solvent morphology modifier, this is a very important terminology. A third component can be incorporated into binary blend system to modify the morphology of the photo active layer ok. Let us say I use chloroform as one of the solvent and I use some additive because I know that chloroform vaporize very rapidly so that to get a larger crystal grain I have added an additive. But what I can also do, I can also add another solvent a third solvent which dielectric constant is way different than the chloroform, then it is called the anti-solvent of the chloroform.

So the third component remains in the BHJ film and affects the morphology in contrast to the solvent additive which is removed after it modifies the morphology. So after the solvent additive has modified the morphology then we remove it but the solvent anti-solvent that it remains in the system. Third component material include organic semiconductor, organic insulator and block copolymer and can be a second donor or acceptor, an energy cascade linker or an alloy of a donor and an acceptor.

Depending on the optoelectronic properties and the location of the third component in the blend. So basically where you wanted to have this third component where is at a donor positions or where is at a acceptor positions where the energetic should be there, what will be the packing fraction. So depending all this factor we can choose this third component, the third component acts as a morphology modifier. Proper use of morphological modifier controls the domain size which is very important, crystalline ordering of defect sites in the blend films. We do not need lot of defects. If there are lot of defects state is there that means there are lot of trap states is there that leads to the electron whole recombination, so that is why to minimize this thing we add this third solvent, well.

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As I mention that there are several other techniques is there for example post-treatment, posttreatments can be usually performed on deposited active films. Thermal annealing and solvent annealing and polar solvent post-treatment methods are some of the popular post treatment methods. These methods can be used to optimize the morphology of the polymer acceptor BHJ structures, now what is thermal annealing.

Let us say I have this substrate on which I made these P3HT PCBM film again we take this example of this conventional material P3HT PCBM right. So now after making the film so I wanted to enhance or I want to ensure the crystallinity of the film. So one way of increasing the crystallinity is the heating the film, now the heating of the film is called sintering, now we can sinter it and we could sinter it at a temperature we should have a higher than the boiling point of the solvent.

So let us say if I use chlorobenzene or if I use chloroform, so I should heat at a temperature which will be higher than the boiling point of the solvent. So that even the there is some residual

solvent stage in the systems after this process that will evaporate and the crystallization process will complete. So when we heat the P3HT PCBM film at 80 degree Celsius and if we heat at 120 degree Celsius the morphology of the film will be way different.

And most of the cases to get an optimum morphology and a optimum crystallites the 120 degree Celsius works better than 80 degree centigrades, that is why we usually sinter the film after fabricating the P3HT PCBM blend. Now this annealing is done by the presence of the heat that is why it is called thermal annealing, similarly you can anneal the film in the presence of the solvent vapor also that is called solvent annealing.

For example let us say I have made a P3HT film using dichlorobenzene as a solvent now to enhance the morphology what we can do we can take the chloroform and we can saturate it beyond it is paper spacer and then we can expose the film with the saturated paper spacer of this solvent. So what will do this will plasticize or passivate the trap states and it will enhance the morphology of the film.

And also you can use polar solvent as a post treatment methods, these methods can be use to optimize the morphology of the polymer acceptor bulk hydrogen cell structures. They induce the formation of a highly crystalline domain consisting of the donor polymer interconnections between the donor and the acceptor and particle space separation that improves excite on dissociation charge transport and charge collection.

As a result the associated solar cell has a high power conversion efficiency, now if the morphology is good obviously the charge dissociation or excite on dissociation, excite on transport and excite on collections or the charged collection all of the factors will be optimized and if all of the factors is optimized then efficiency will automatically will increased.

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# **Post-Treatment**

- Thermal annealing can be used to modify the morphology of the active layer. The applied heat energy induces phase separation and changes in properties such as the nano morphology and crystallinity of the donor: acceptor blend; these properties are closely related to the optoelectronic properties of the associated PV cell.
- Solvent vapor annealing is also known as solvent annealing and is achieved by confining a photoactive film and a processing solvent together in a closed system.
- Solvent vapor annealing can be a good method for the preparation of a wellaligned BHJ structure because the solvent vapor in the closed system slows the drying process and thus provides enough time for control of the morphology of the active film.



Some of the post treatment process which will discuss here for example thermal annealing that can be use to modify the morphology of the active layer. The applied heat energy induce phase separation and changes in properties such as the nano morphology and crystallinity of the donor, donor acceptor blend. These properties are closely related to the optoelectronic properties of the associated solar cell.

Solvent vapor annealing is also known as the solvent annealing and is achieved by confining a photoactive film and a processing solvent together in a closed system. Solvent vapor annealing can be a good treatment for preparation of a well aligned BHJ structure because the solvent vapor in the closed system slows the drying process and they provides enough time for control of the morphology of the active film.

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Solvent vapor annealing also improves the crystallinity nano scale phase separation and extent of the interconnected donor acceptor network. These change can increase the whole mobility current density and power conversion efficiency of solar cell. Polar solvent treatment which is also a popular method for post treatment use a polar solvent such s ethanol or methanol the morphology of the donor acceptor active layer can be tailored by spin coating the polar solvent on to the active layer or by soaking it with the polar solvent.

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Now the another aspect of this is order nano structures within the photoactive layer, now this selection inverse appropriate that intentionally introduce well-ordered nanostructures within the active layer to improve the photovoltaic performance of the OPVs. Now whenever we talked

about the photoanode I say that like you know we have use the pad geometry but we can also use curvatures in this things, we can also make different kind of patterns on the substrate.

Now one of this objective of the pattern let us say I want to make a waveguide kind of structure, so then I can increase the light confinement. If electric field enhancement is there then there will be more charge separations and there will be more short-circuit current in the devices. The main purpose of the introduction of such nanostructure is to build well organized pathways for charge transport, so that the charge carriers can be collected at electrodes without several loss.

Fabrication of the ordered nanostructure has usually been achieved by exploiting molecular selfassembly and nanostructured templating. In the following we discuss the 2 most widely studied ordered nanostructures, ordered BHJ structures and 1D PNW structures.

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Molecular orientation engineering at the donor acceptor interface, the molecular orientation of the organic semiconductor in electronic device critically affects their characteristics. The face-on orientation of organic semiconductors molecule should be respect to the substrate is favorable in general for charge transport in the OPVs. The optical properties of organic semiconductors such as optical transition and excite on diffusion can be highly anisotropic and strongly dependent on molecular orientation.

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# Molecular Orientation Engineering at the Donor– acceptor Interface

- When monolayer graphene inserted at the anode interface as an epitaxial template for the growth of highly oriented pentacene crystals in the lying-down orientation. This orientation significantly increases light absorption and the exciton diffusion length, and thereby increases the efficiency of photon harvesting by the pentacene layer over that with a layer in the standing-up orientation.
- It has been found that the orientation of molecules at the donor-acceptor interface also has a strong influence on the photocurrent generation and interfacial energetics of OPVs.



When monolayer graphene inserted at the anode interface as an epitaxial template for the growth of highly oriented pentacene crystals in the lying down orientation. This orientation significantly increase light absorptions and the excite on diffusion length and thereby increase the efficiency of the photon harvesting by the pentacene layer over that with a layer in the standing-up orientation.

So we can make it either by a lying down orientation or a by a standing-up orientation, but if we put a layer of graphene on the perylene or on the pentacene in a lying down orientation then the charge carrier mobility and the charge collection efficacy because of the larger excite on diffusion length will be much higher. And that will always lead to the higher (()) (26:44) and higher efficiency, it has been found that the orientation of molecules at the donor acceptor interface also has a strong influence on the photocurrent generation and interfacial energetics of OPVs.

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The next important aspect is the morphological stability it is highly desirable that solar cell must be morphologically stable. Several critical factors influence the stability of OPVs including thermal degradation, diffusion of components, chemical reactions between components and photo degradation. Crosslinkable donors and acceptors, side chain modification, incorporation of metal nanoparticles into the active layer. And removal of residual solvents and additives of the factors use to improve the morphological stability of the OPV.

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Thermal degradation is a major source of morphological instability, the use of non solvent additives can improve the thermal stability of the BHJ films. Now if you have a temperature variations over the from the winter to the summer or even in the summer like in a firm 30 degree

to 50 degree Celsius. Because of this temperature variations some of the organic materials are not very much stable.

So to prevent that we use some kind of solvent additive like acetyl butyl pyridine or any kind of solvent additives. The modification of the molecular structures of donor polymers can also suppress the thermal degradation of their associated BHJ films. The presence of cross linkable components in BHJ films can prevent other components from diffusing from the original matrix and increase morphological stability.

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# Conclusion

- The ideal morphology is necessary for high power conversion efficiency and stability in organic photovoltaics and this ideal morphology can be achieved through the strategies mentioned above.
- optimized morphologies are not stable for long periods because of processes such as the molecular diffusion of the organic components and molecular degradation through exposure to environmental conditions. Therefore, further research is required into the fabrication of long-lasting OPVs that can resist morphological degradation under diverse environmental conditions.
- By optimizing their morphologies, high performing OPVs with PCEs above 15% can be obtained in the near future for their commercialization to advance the OPV market.

The ideal morphology, so basically is necessary for high power conversion efficiency and stability in organic photovoltaics and this ideal morphology can be achieved through the strategies mentioned above. So for conclusion like what you have learn that and in a bulk heterojunction solar cell we need a bicontinuous phase separated percolated network and that is an optimum morphology.

An optimum morphology is always related to the excite on diffusion and excite on dissociations, if the morphology is optimum then the photo physics will also be good and we will get a larger JSC value and larger VOC value. We have discuss some of the protocols or some of the decent strategies for improving the morphology and improving the efficiency of the device. Optimized

morphology are not stable for long periods because of processes such as the molecular diffusion of the organic components.

And molecular degradation through exposure to environmental conditions therefore further research is required into the fabrication of the long lasting OPVs that can resist morphological degradation under diverse environmental conditions. By optimizing their morphologies high performing OPVs with PCE above then 15% can be obtained in near future for their commercialization to advance the OPV market.

Now most of the time the high efficiency in organic solar cell is related to the good morphology and a good photo physical properties. So if we can improve the morphological properties, so the photo physics will automatically will improve and we will get a high efficiency and high stable organic solar cell. There are lot of again research papers or review papers on the morphology optimizations and there are some books is also there.

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Recent advances in morphology optimization for organic photovoltaics by Kilwon Cho that is a very important reference. Similarly for a more fundamental understanding you can refer to the solar photovoltaics book by Chetan Solanki. But there are also lot of excellent review papers and research articles on the morphology optimization things, thank you so much.