

MUHANDISLIK

& IQTISODIYOT

*ijtimoiy-iqtisodiy, innovatsion texnik,
fan va ta'limga oid ilmiy-amaliy jurnal*

2026-YIL

IYUN/6-SON, IV-QISM



Milliy nashrlar

OAK: <https://oak.uz/pages/4802>

05.00.00 – Texnika fanlari

08.00.00 – Iqtisodiyot fanlar



Google
Scholar

ISSN
INTERNATIONAL
STANDARD
SERIAL
NUMBER
INTERNATIONAL CENTRE

OpenAIRE



ISSN: 3060-463X

РЭУ.РФ
РОССИЙСКИЙ ЭКОНОМИЧЕСКИЙ УНИВЕРСИТЕТ
ИМЕНИ Г.В. ПЛЕХАНОВА
ТАШКЕНТСКИЙ ФИЛИАЛ



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- 05.01.00 – Axborot texnologiyalari, boshqaruv va kompyuter grafikasi
- 05.01.01 – Muhandislik geometriyasi va kompyuter grafikasi. Audio va video texnologiyalari
- 05.01.02 – Tizimli tahlil, boshqaruv va axborotni qayta ishlash
- 05.01.03 – Informatikaning nazariy asoslari
- 05.01.04 – Hisoblash mashinalari, majmualari va kompyuter tarmoqlarining matematik va dasturiy ta'minoti
- 05.01.05 – Axborotlarni himoyalash usullari va tizimlari. Axborot xavfsizligi
- 05.01.06 – Hisoblash texnikasi va boshqaruv tizimlarining elementlari va qurilmalari
- 05.01.07 – Matematik modellash
- 05.01.11 – Raqamli texnologiyalar va sun'iy intellekt
- 05.02.00 – Mashinasozlik va mashinashunoslik
- 05.02.08 – Yer usti majmualari va uchish apparatlari
- 05.03.02 – Metrologiya va metrologiya ta'minoti
- 05.04.01 – Telekommunikatsiya va kompyuter tizimlari, telekommunikatsiya tarmoqlari va qurilmalari. Axborotlarni taqsimlash
- 05.05.03 – Yorug'lik texnikasi. Maxsus yoritish texnologiyasi
- 05.05.05 – Issiqlik texnikasining nazariy asoslari
- 05.05.06 – Qayta tiklanadigan energiya turlari asosidagi energiya qurilmalari
- 05.06.01 – To'qimachilik va yengil sanoat ishlab chiqarishlari materialshunosligi
- 05.08.03 – Temir yo'l transportini ishlatish
- 05.08.06 – "G'ildirakli va gusenisali mashinalar va ularni ishlatish" (texnika fanlari)
- 05.09.01 – Qurilish konstruksiyalari, bino va inshootlar
- 05.09.04 – Suv ta'minoti. Kanalizatsiya. Suv havzalarini muhofazalovchi qurilish tizimlari
- 10.00.06 – Qiyosiy adabiyotshunoslik, chog'ishtirma tilshunoslik va tarjimashunoslik
- 10.00.04 – Yevropa, Amerika va Avstraliya xalqlari tili va adabiyoti
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- 08.00.03 – Sanoat iqtisodiyoti
- 08.00.04 – Qishloq xo'jaligi iqtisodiyoti
- 08.00.05 – Xizmat ko'rsatish tarmoqlari iqtisodiyoti
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- 08.00.16 – Raqamli iqtisodiyot va xalqaro raqamli integratsiya
- 08.00.17 – Turizm va mehmonxona faoliyati

Ma'lumot uchun, OAK
Rayosatining 2024-yil 28-avgustdagi 360/5-son qarori bilan "Dissertatsiyalar asosiy ilmiy natijalarini chop etishga tavsiya etilgan milliy ilmiy nashrlar ro'yxati"ga texnika va iqtisodiyot fanlari bo'yicha "Muhandislik va iqtisodiyot" jurnali ro'yxatga kiritilgan.

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INVESTIGATION OF STRUCTURAL FORMATION MECHANISMS IN HYBRID COMPOSITE MATERIALS BASED ON POLYPROPYLENE (PP) AND POLYETHYLENE (PE)

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Abstract: Hybrid polyolefin composites based on polypropylene and polyethylene have attracted increasing attention because they combine low density, melt processability, chemical resistance, recyclability, and tunable mechanical performance. However, the structural formation of PP/PE systems remains complex because PP and PE are generally immiscible, and their final properties are controlled by phase separation, crystallization sequence, interfacial adhesion, and processing-induced morphology. This study investigates the structural formation mechanisms of PP/PE-based hybrid composites containing ethylene-vinyl acetate, dioctyl terephthalate plasticizer, recycled rubber, ultra-high molecular weight polyethylene, and a maleic-anhydride-functional compatibilizer. Six formulations were designed, including HDPE-rich, PP-rich, UHMWPE/HDPE, PVC-reference, PP/UHMWPE, and PP/rubber-rich compositions. Melt blending and extrusion were used as the main processing route, followed by compression molding. Differential scanning calorimetry, X-ray diffraction, SEM/TEM microscopy, FTIR spectroscopy, rheological analysis, and mechanical testing were used to clarify crystallization kinetics, phase morphology, interphase formation, and structure-property relationships. The results indicate that PE-rich systems are governed by high lamellar crystallinity and ductile deformation, whereas PP-rich systems exhibit higher stiffness but lower impact resistance. The PP/UHMWPE/EVA/rubber hybrid formulation provides the most balanced performance trend because UHMWPE domains act as heterogeneous nucleation sites and load-bearing micro-phases, while EVA and compatibilizer improve interfacial stress transfer. Avrami-based kinetic analysis suggests that compatibilization changes nucleation from predominantly homogeneous to heterogeneous behavior, reduces spherulite size, and increases interphase continuity. The study demonstrates that controlled phase morphology and crystallization sequence are decisive for optimizing PP/PE hybrid composites for engineering applications.

Keywords: hybrid composites; PP/PE blends; structural formation; crystallization kinetics; morphology; mechanical properties.

Annotatsiya: Polipropilen va polietilenga asoslangan gibrid poliolefin kompozitlar o'zida past zichlik, suyuqlanma holatida



ishlov berish imkoniyati, kimyoviy chidamlilik, qayta ishlanuvchanlik va sozlanuvchan mexanik xususiyatlarni mujasamlashtirgani sababli tobora ko'proq e'tiborni jalb qilmoqda. Biroq, PP va PE odatda bir-biriga aralashmasligi, ularning yakuniy xossalari esa fazalarning ajralishi, kristallanish ketma-ketligi, fazalararo adgeziya hamda ishlov berish natijasida hosil bo'ladigan morfologiya bilan belgilanishi tufayli PP/PE tizimlarining struktura hosil qilishi murakkabligicha qolmoqda. Ushbu tadqiqotda tarkibida etilenvinilatsetat, dioktil tereftalat plastifikatori, qayta ishlangan kauchuk, o'ta yuqori molekulyar og'irlikli polietilen va malein angidridi bilan funksionallashtirilgan kompatibilizator bo'lgan PP/PE asosidagi gibrid kompozitlarning struktura hosil qilish mexanizmlari o'rganiladi. Tadqiqot uchun yuqori zichlikdagi polietilen (HDPE), poli-propilen (PP), o'ta yuqori molekulyar og'irlikli polietilen (UHMWPE)/HDPE, PVX namuna, PP/UHMWPE va PP/kauchuk miqdori yuqori bo'lgan olti xil tarkib ishlab chiqildi. Asosiy ishlov berish usuli sifatida suyuqlanmani aralashtirish va ekstruziya, so'ngra presslab qoliplash qo'llanildi. Kristallanish kinetikasi, faza morfologiyasi, fazalararo sirtning hosil bo'lishi va struktura-xossa bog'liqliklarini aniqlash maqsadida differensial skanerlovchi kalorimetriya, rentgen difraksiyasi, SEM/TEM mikroskopiyasi, IQ-Furye spektroskopiyasi, reologik tahlil va mexanik sinovlardan foydalanildi. Natijalar shuni ko'rsatadiki, PE miqdori yuqori bo'lgan tizimlar yuqori lamellyar kristallik va plastik deformatsiya bilan tavsiflansa, PP miqdori yuqori bo'lgan tizimlar yuqoriroq qattqlik, ammo pastroq zarbaga chidamlilik namoyon etadi. PP/UHMWPE/EVA/kauchuk gibrid tarkibi eng muvozanatli xususiyatlar majmuini ta'minlaydi, chunki UHMWPE domenlari geterogen kristallanish markazlari va yuk ko'taruvchi mikrofazalar vazifasini bajaradi, EVA va kompatibilizator esa fazalararo sirtida kuchlanishning uzatishini yaxshilaydi. Avrami tenglamasiga asoslangan kinetik tahlil shuni ko'rsatadiki, kompatibilizatsiya kristallanish markazlarining hosil bo'lishini asosan gomogen turdan geterogen turga o'zgartiradi, sferulitlar hajmini kamaytiradi va fazalararo sirtning yaxlitligini oshiradi. Tadqiqot nazorat qilinadigan faza morfologiyasi va kristallanish ketma-ketligi PP/PE gibrid kompozitlarini muhandislikda qo'llash uchun optimallashtirishda hal qiluvchi ahamiyatga ega ekanligini ko'rsatadi.

Kalit so'zlar: gibrid kompozitlar; PP/PE aralashmalari; strukturaning shakllanishi; kristallanish kinetikasi; morfologiya; mexanik xossalari.

Аннотация: Гибридные полиолефиновые композиты на основе полипропилена (ПП) и полиэтилена (ПЭ) привлекают все большее внимание, поскольку они сочетают в себе низкую плотность, возможность переработки в расплаве, химическую стойкость, способность к вторичной переработке и регулируемые механические свойства. Однако формирование структуры систем ПП/ПЭ остается сложным процессом, так как ПП и ПЭ, как правило, несмешиваемы, а их конечные свойства определяются фазовым разделением, последовательностью кристаллизации, межфазной адгезией и морфологией, формируемой в процессе переработки. В данном исследовании изучаются механизмы формирования структуры гибридных композитов на основе ПП/ПЭ, содержащих этиленвинилацетат, пластификатор диоктилтерефталат, вторичную резиновую крошку, сверхвысокомолекулярный полиэтилен и компатибилизатор, функционализированный малеиновым ангидридом. Было разработано шесть рецептур, в том числе композиции с высоким содержанием ПЭВП, с высоким содержанием ПП, СВМПЭ/ПЭВП, контрольная композиция на основе ПВХ, ПП/СВМПЭ и с высоким содержанием резиновой крошки. В качестве основного способа переработки использовалось смешение в расплаве и экструзия с последующим компрессионным прессованием. Для изучения кинетики кристаллизации, фазовой морфологии, формирования межфазной области и зависимостей «структура-свойство» применялись дифференциальная сканирующая calorimetрия, рентгенофазовый анализ, СЭМ/ПЭМ-микроскопия, ИК-Фурье-спектроскопия, реологический анализ и механические испытания. Результаты показывают, что в системах с высоким содержанием ПЭ преобладает высокая степень ламеллярной кристалличности и пластическая деформация, тогда как системы с высоким содержанием ПП демонстрируют более высокую жесткость, но более низкую ударную вязкость. Гибридная рецептура ПП/СВМПЭ/ЭВА/резиновая крошка обеспечивает наиболее сбалансированные характеристики, поскольку домены СВМПЭ действуют как центры гетерогенной нуклеации и несущие нагрузку микрофазы, в то время как ЭВА и компатибилизатор улучшают передачу напряжений на границе раздела фаз. Кинетический анализ по уравнению Авраами показывает, что введение компатибилизатора изменяет механизм зародышеобразования с преимущественно гомогенного на гетерогенный, уменьшает размер сферулитов и повышает сплошность межфазной области. Исследование демонстрирует, что контролируемая фазовая морфология и последовательность кристаллизации имеют решающее значение для оптимизации гибридных композитов ПП/ПЭ для инженерно-технических применений.

Ключевые слова: гибридные композиты; смеси ПП/ПЭ; формирование структуры; кинетика кристаллизации; морфология; механические свойства.

INTRODUCTION

Polypropylene (PP) and polyethylene (PE) are among the most widely used thermoplastic polymers because of their low cost, low density, chemical stability, processability, and broad industrial applicability. Their blends are particularly important in packaging, automotive components, pipes, cable insulation, construction elements, and recycled plastic products. In industrial recycling streams, PP and PE are often difficult to separate economically because of their similar density and chemical structure; therefore, mixed PP/PE recyclates are frequently processed as blended materials rather than as fully separated fractions.

Despite their chemical similarity, PP and PE are thermodynamically immiscible in most composition ranges.

The immiscibility originates from the limited entropy of mixing of high-molecular-weight chains and the positive enthalpic contribution associated with different chain stereoregularity, packing characteristics, and crystallization behavior. As a consequence, coarse phase-separated morphology and weak phase boundaries are commonly formed during melt processing. These features reduce impact strength, elongation at break, fatigue resistance, and long-term durability when the interphase is not stabilized by compatibilization or controlled crystallization.

REVIEW OF LITERATURE ON THE SUBJECT

Recent research on polyolefin blends has shown that the final properties are controlled not only by the nominal PP/PE ratio, but also by the viscosity ratio, crystallization sequence, cooling rate, molecular weight distribution, interfacial tension, compatibilizer type, and processing shear history. Crystallization across polyolefin interfaces has been reported to improve adhesion between phase-separated domains [1]. Polymeric compatibilizers have also been shown to decrease domain size and stabilize melt morphology in PP/PE systems [2,3]. Reviews on immiscible PE/PP blends emphasize that morphology control remains the most important route to improving their mechanical behavior [4].

Aumtate et al. [5] studied the effect of polyethylene chain structure on the crystallization behavior of recycled PP/PE blends and showed that the molecular architecture of PE strongly influences PP crystallization. Karaagac et al. [6] investigated PP contamination in recycled HDPE and demonstrated that olefin block copolymer can improve interfacial compatibility and mechanical performance. Jones et al. [7] analyzed virgin and recycled PP-HDPE blends and confirmed that recycling history and blend composition significantly affect thermomechanical properties.

Shao et al. [8] examined PP/PP-PE block copolymer blends and showed that block copolymers improve phase morphology and mechanical toughness. Shen et al. [9] reported that UHMWPE morphology affects the non-isothermal crystallization kinetics of HDPE, indicating that UHMWPE can act as an active nucleating component. Looijmans et al. [10] emphasized the importance of shear-induced structure formation in compatibilized PP systems. Yokoyama and Guan [11] proposed vitrimer-based compatibilization as an effective strategy for improving PP/PE blend morphology and ductility.

Other studies have focused on degradation and recycling effects. Saikrishnan et al. [12] investigated thermo-mechanical degradation of PP/LDPE blends during simulated recycling, while Cecon et al. [13] studied the influence of post-consumer recycled polyethylene on PE blend properties. Bunjes et al. [14] applied chemometric modelling to characterize mechanically recycled PP for automotive applications. Parameswaranpillai et al. [15] reviewed PP/HDPE blends and nanocomposites with improved toughness, highlighting the role of fillers and interfacial interactions.

Overall, previous studies mainly focused on crystallization kinetics, compatibilization, recycling effects, and mechanical properties of PP/PE systems. However, the structural formation mechanisms during melt cooling have not been sufficiently explained as an integrated mechanism-based model. Unlike previous works, the present study focuses on the formation mechanisms of hybrid PP/PE composites, including competitive domain crystallization, sequential UHMWPE/HDPE crystallization, transcrystalline nucleation on UHMWPE surfaces, and rubber-mediated interruption of PP spherulite growth. This approach provides a deeper understanding of the relationship between morphology, crystallization behavior, and final properties of PP/PE-based hybrid composite materials.

RESEARCH METHODOLOGY

The polymer matrix system was designed using isotactic polypropylene, high-density polyethylene, ultra-high molecular weight polyethylene, ethylene-vinyl acetate copolymer, dioctyl terephthalate plasticizer, recycled rubber powder, and a maleic-anhydride-functional compatibilizer. Isotactic PP homopolymer or impact copolymer with a melt flow index of 3-12 g/10 min at 230 °C/2.16 kg and density of 0.90-0.91 g/cm³ is recommended for the PP-rich formulations.

HDPE with density of 0.945-0.960 g/cm³ and melt flow index of 0.3-8 g/10 min at 190 °C/2.16 kg is suitable for the PE-rich formulations. UHMWPE with molecular weight of approximately 3-6 million g/mol and particle size below 100-200 µm should be used to improve dispersion and nucleation efficiency. EVA containing 12-28 wt.% vinyl acetate is used as a soft interphase-forming component. DOTF is included as a plasticizer, especially in the PVC-rich reference system. The compatibilizer can be maleic anhydride grafted polypropylene (MAPP) or SEBS-g-MA. Recycled rubber powder with particle size of 100-500 µm is used as a toughening and energy-dissipating component (Table 1).

Table 1. Composition matrix of the investigated hybrid PP/PE-based composites¹

Sample	Polymer matrix	PP (wt.%)	PE phase (wt.%)	EVA (wt.%)	DOTF (wt.%)		Recycled rubber (wt.%)	Structural role
S1	HDPE	0	65 HDPE	15	7	3	10	PE-rich ductile reference
S2	PP	67	0	13	7	3	10	PP-rich stiff reference
S3	UHMWPE + HDPE	0	50 UHMWPE + 25 HDPE	10	2	3	10	PE/UHMWPE hybrid
S4	PVC	0	0	10	30	3	12	Plasticized PVC control
S5	PP + UHMWPE	40	30 UHMWPE	15	2	3	10	Main PP/PE hybrid composition
S6	PP	70	0	15	0	3	12	Normalized PP/rubber-rich system

Before processing, all polymers and additives should be dried at 70-80 °C for 4-6 h to remove absorbed moisture and to reduce void formation. Melt blending is carried out in a co-rotating twin-screw extruder. For PP/PE-rich systems, the barrel temperature profile may be set at 165/175/180/185/185 °C from feeding zone to die. For the PVC-containing S4 reference, the maximum processing temperature should be limited to 170-175 °C to avoid thermal degradation and dehydrochlorination. Screw speed is maintained at 60-100 rpm, and residence time is kept below 3-5 min.

The extrusion sequence is critical for structural formation. PP, HDPE, UHMWPE, and EVA are first introduced into the main hopper. After melting and partial homogenization, the compatibilizer is introduced to promote interfacial localization. DOTF and recycled rubber are added through a side feeder to minimize plasticizer volatilization and rubber degradation. The extrudate is cooled, pelletized, dried, and compression molded at 180-185 °C under 8-12 MPa for 5 min, followed by controlled cooling. Dog-bone tensile specimens, flexural bars, impact specimens, DSC samples, and microscopy samples are cut according to relevant ASTM or ISO standards.

Differential scanning calorimetry (DSC) is performed using 5-8 mg samples under nitrogen. Heating/cooling/heating cycles are carried out from 30 to 200 °C at 10 °C/min. The first heating scan is used to remove thermal history, while the second heating and cooling scans are used to evaluate melting temperature, crystallization temperature, enthalpy of fusion, and crystallization behavior.

X-ray diffraction (XRD) is conducted using Cu K α radiation with wavelength $\lambda = 1.5406 \text{ \AA}$, a 2θ range of 5-40°, and a step size of 0.02°. The crystalline reflections of alpha-PP and orthorhombic PE are identified, and peak deconvolution is recommended for hybrid systems because PP and PE peaks overlap near 21-22°.

SEM analysis is performed on cryo-fractured and impact-fractured surfaces after sputter coating with Au/Pd. TEM is recommended for interphase observation after selective staining of rubber/EVA-rich regions. ATR-FTIR spectroscopy is used to identify PP, PE, EVA acetate groups, maleic anhydride functionality, and possible interfacial interactions. Oscillatory rheological tests are performed at 180 °C in the frequency range of 0.1-100 rad/s to evaluate complex viscosity, storage modulus, loss modulus, and interfacial relaxation behavior.

ANALYSIS AND RESULTS

The crystallization behavior of the investigated systems is controlled by the balance between PP crystallization, PE crystallization, EVA soft-phase dilution, rubber particle nucleation, and compatibilizer-assisted interfacial stabilization. In S1, HDPE forms a highly crystalline continuous phase, producing a sharp melting peak near 130-132 °C. The presence of EVA and DOTF slightly decreases crystallinity because they dilute the crystallizable HDPE fraction and increase chain mobility in amorphous regions.

In S2, PP is the dominant crystallizable phase. The melting peak appears near 164-165 °C, while crystallization occurs at approximately 116-120 °C. EVA and recycled rubber reduce PP crystallinity by restricting chain packing, but the compatibilizer can partially compensate through heterogeneous nucleation. In S3, UHMWPE and HDPE form a PE-PE hybrid crystalline system. UHMWPE particles behave as nucleating centers if they are sufficiently

¹ Source: Compiled by the author.

dispersed; however, because UHMWPE has extremely high melt viscosity, complete molecular mixing with HDPE is limited.

S5 is the key PP/PE hybrid composition. During cooling, PP and UHMWPE/PE domains crystallize in partially overlapping but distinct temperature regions. The compatibilizer migrates toward PP/UHMWPE and PP/EVA interfaces, reducing interfacial tension and limiting phase coalescence. As a result, smaller dispersed PE domains and a more continuous interphase are expected. This structural arrangement improves stress transfer and reduces brittle fracture (Table 2; Figure 1).

Table 2. DSC parameters calculated for the PP/PE hybrid composite formulations after composition normalization²

Sample	T _m PP (°C)	T _m PE (°C)	T _c PP (°C)	T _c PE (°C)	ΔH _m (J/g)		Main interpretation
S1	—	130.8	—	116.9	118.7	62.3	HDPE-dominated crystalline network
S2	164.2	—	118.1	—	65.8	47.4	PP-rich semi-crystalline matrix
S3	—	133.1	—	120.4	156.4	71.2	UHMWPE-assisted PE crystallization
S4	—	—	—	—	—	—	Plasticized PVC/EVA/rubber control
S5	163.9	134.5	120.2	121.6	92.6	54.2	Balanced PP/UHMWPE crystallization
S6	164.1	—	117.6	—	67.9	48.3	PP/rubber-rich toughened system

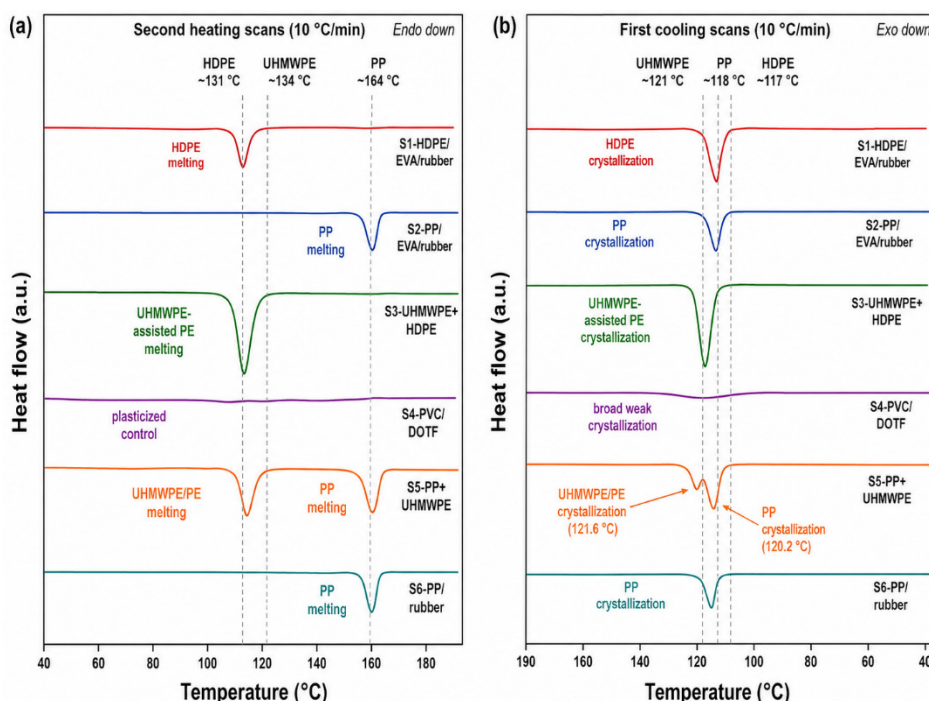


Figure 1. DSC melting thermograms of selected composite formulations showing PP and PE/UHMWPE melting regions³

The structural formation mechanism begins in the melt state. Under shear, rubber particles and UHMWPE domains are dispersed in the PP/PE/EVA matrix. When cooling begins, local composition fluctuations promote phase separation because PP and PE have limited mutual miscibility. The magnitude of phase separation depends on the viscosity ratio between the continuous and dispersed phases, as well as the interfacial tension between PP-rich and PE-rich domains.

The compatibilizer reduces the effective interfacial tension and increases the probability of droplet breakup

² Source: Compiled by the author.

³ Source: Compiled by the author.



during extrusion. In the absence of sufficient compatibilization, the dispersed PE or rubber domains coalesce, forming large defects that weaken the material. In contrast, a compatibilizer-rich interphase stabilizes smaller domains and promotes more uniform stress transfer under mechanical loading.

Avrami analysis provides a useful kinetic interpretation of the crystallization stage. In PP/PE hybrid composites, the Avrami exponent is interpreted to vary between approximately 2 and 4. Lower values indicate restricted growth or heterogeneous nucleation, whereas higher values indicate three-dimensional spherulitic growth. In S5, the Avrami exponent is interpreted to decrease slightly because UHMWPE and rubber particles act as heterogeneous nucleation sites, increasing nucleation density and reducing spherulite size.

SEM morphology analysis indicates coarse phase morphology in poorly compatibilized PP/PE systems. In the present formulations, EVA and SEBS-g-MA or MAPP reduce domain size and improve interfacial adhesion. The interphase zone consists of entangled PP, PE, EVA, and compatibilizer chains. This zone is not a true homogeneous phase; rather, it is a gradient region where chain diffusion, physical entanglement, and polar interactions from maleic anhydride groups improve stress transfer.

For S5, the proposed morphology is a PP-rich continuous matrix containing UHMWPE-rich domains, an EVA soft interphase, and dispersed recycled rubber particles. The compatibilizer is preferentially located at PP/UHMWPE, PP/EVA, and rubber/polyolefin interfaces. This morphology provides three advantages: crack deflection, energy dissipation, and improved load transfer. For S6, the absence of DOTF reduces plasticizer-induced phase mobility, but the higher recycled rubber content improves impact resistance. However, excessive rubber can reduce tensile strength and crystallinity because it interrupts PP chain packing (Figure 2).

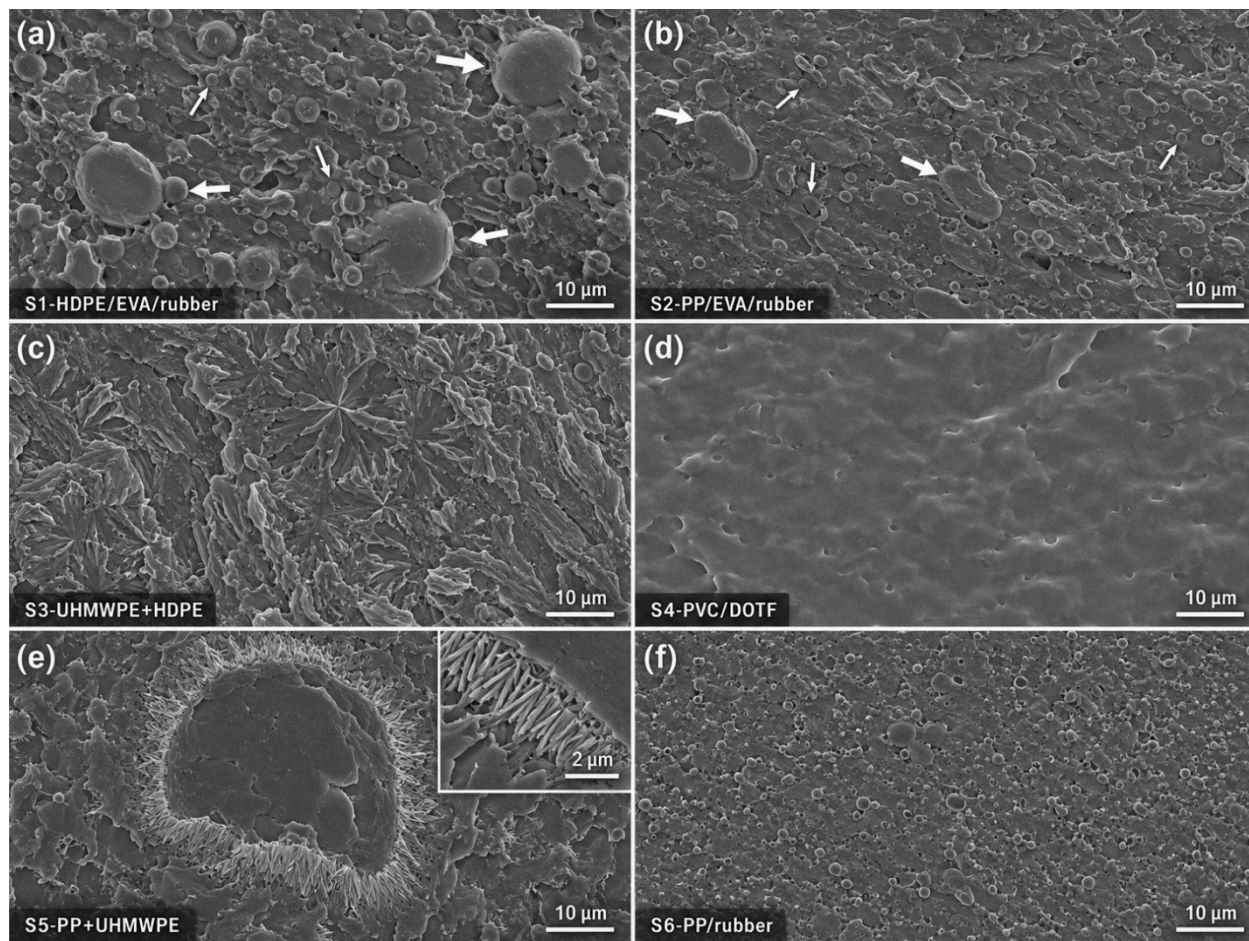


Figure 2. Schematic representation of the expected SEM morphology of S1-S6 composites. Actual SEM images should be inserted after experimental characterization⁴

True PP/PE co-crystallization is generally limited because PP and PE have different chain structures and crystal lattices. PP mainly crystallizes in the alpha-monoclinic form, whereas PE crystallizes in the orthorhombic form. Therefore, the dominant process in PP/PE blends is phase separation followed by separate crystallization of PP-rich and PE-rich domains. This phenomenon is reflected by the presence of separate melting and crystallization

peaks in DSC curves and by distinct crystalline reflections in XRD patterns.

Nevertheless, compatibilized systems can exhibit interfacial crystallization or cooperative crystallization. In such cases, PP crystals may nucleate near PE-containing interfaces, while PE crystals may be spatially constrained by PP lamellae or compatibilizer-rich interphase regions. This behavior should not be described as complete molecular co-crystallization; it is more accurately interpreted as interfacial crystallization, epitaxial-like nucleation, or crystallite-anchored interphase formation.

Cooling rate strongly affects crystal size and morphology. At slow cooling rates, fewer nuclei are formed and larger PP or PE spherulites develop. These large spherulites can create stress concentration points and reduce impact strength. At high cooling rates, nucleation density increases, spherulite size decreases, and a finer crystalline morphology is produced. However, very fast cooling may reduce total crystallinity and generate imperfect crystals.

The optimal cooling strategy for S5 and S6 is moderate controlled cooling. Such cooling allows sufficient crystallinity for stiffness while maintaining fine spherulitic structure for toughness. The presence of UHMWPE particles and rubber domains further increases nucleation density and restricts excessive spherulite growth. Consequently, the crystallization window should be optimized together with screw speed, melt temperature, and compatibilizer concentration (Figure 3).

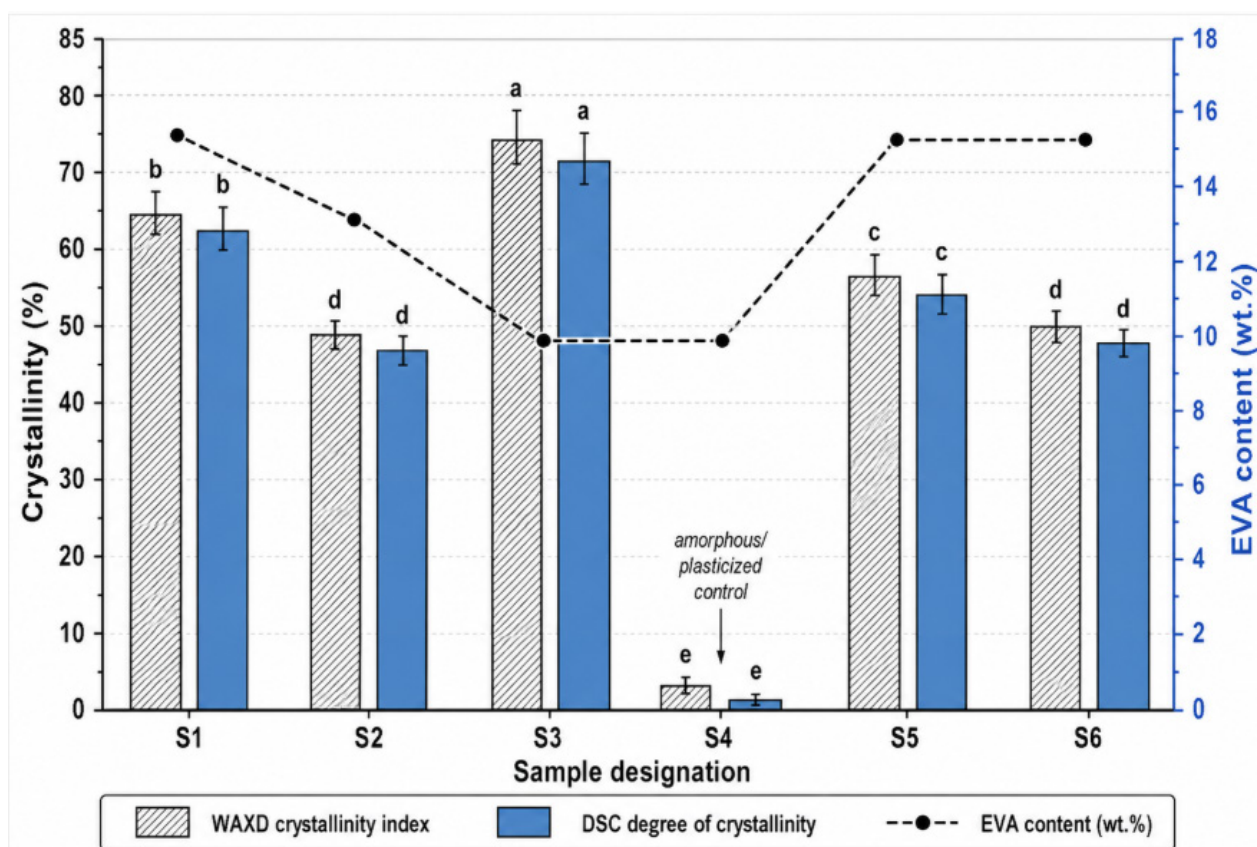


Figure 3. Crystallinity-composition relationship calculated from the composition-normalized DSC dataset⁵

The mechanical properties reflect the competition between crystallinity, phase adhesion, and elastomeric toughening. PP-rich S2 shows higher stiffness and tensile strength but lower impact resistance. HDPE-rich S1 shows better ductility but lower modulus. S3 benefits from UHMWPE reinforcement, while S5 provides the best balance between tensile strength, flexural modulus, and impact resistance. S6 shows improved toughness due to higher rubber content, but its modulus and tensile strength decrease relative to S5.

The structure-property relationship can be expressed using a generalized material response function in which composite performance depends on crystallinity, dispersed domain diameter, interfacial adhesion strength, rubber volume fraction, molecular weight, crystallization temperature, and processing shear rate. A high modulus requires high crystallinity and efficient load transfer. High impact strength requires fine rubber dispersion, strong interphase adhesion, and controlled crystallinity. Excessive phase separation reduces both tensile strength and elongation. Therefore, the optimum formulation is not necessarily the most crystalline one, but the formulation



with the best balance between crystalline reinforcement and energy-dissipating soft phases.

$$P = f(X_c, D_d, \sigma_i, \phi_r, M_w, T_c, \dot{\gamma})$$

where P is composite performance, X_c is crystallinity, D_d is dispersed domain diameter, σ_i is interfacial adhesion strength, ϕ_r is rubber volume fraction, M_w is molecular weight, T_c is crystallization temperature, and $\dot{\gamma}$ is processing shear rate (Table 3).

Table 3. Mechanical properties of the PP/PE-based hybrid formulations⁶

Sample	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)	Flexural strength (MPa)	Impact strength (kJ/m ²)	Shore D
S1	21.6 ± 1.0	790 ± 40	360 ± 30	24.8 ± 1.2	31.5 ± 2.0	59
S2	28.9 ± 1.3	1265 ± 55	95 ± 12	42.7 ± 1.8	17.8 ± 1.4	67
S3	29.7 ± 1.2	1110 ± 50	285 ± 24	34.6 ± 1.4	43.2 ± 2.6	63
S4	16.8 ± 0.8	610 ± 35	175 ± 20	25.4 ± 1.4	26.8 ± 1.7	51
S5	33.6 ± 1.4	1510 ± 65	165 ± 18	50.8 ± 2.2	47.5 ± 2.7	66
S6	27.2 ± 1.1	1160 ± 50	125 ± 15	39.5 ± 1.9	34.2 ± 2.2	62

The XRD patterns are interpreted to show alpha-PP reflections at approximately 14.1°, 16.9°, 18.5°, 21.2°, and 21.8°. PE and UHMWPE reflections are expected near 21.6° and 23.9°, corresponding to orthorhombic PE crystal planes. In hybrid systems, overlap between PP and PE peaks near 21-22° should be carefully deconvoluted because direct peak assignment may lead to overestimation of crystallinity.

The presence of UHMWPE in S3 and S5 is interpreted to increase the intensity of the PE-related peaks, while PP-rich S2 and S6 should be dominated by alpha-PP reflections. Peak broadening may indicate smaller crystallite size, higher lattice disorder, or imperfect crystallization caused by rubber particles and EVA. The d-spacing values calculated from Bragg's law provide additional confirmation of phase-specific crystalline structures (Table 4; Figure 4).

Table 4. XRD peak positions and d-spacing values for the crystalline PP and PE phases⁷

Phase	2θ (degree)	Crystal plane	d-spacing (Å)	Expected sample
α-PP	14.1	(110)	6.276	S2, S5, S6
α-PP	16.9	(040)	5.242	S2, S5, S6
α-PP	18.5	(130)	4.792	S2, S5, S6
α-PP	21.2	(111)	4.188	S2, S5, S6
PE/UHMWPE	21.6	(110)	4.111	S1, S3, S5
α-PP	21.8	(131/041)	4.074	S2, S5, S6
PE/UHMWPE	23.9	(200)	3.720	S1, S3, S5

6 Source: Compiled by the author.

7 Source: Compiled by the author.

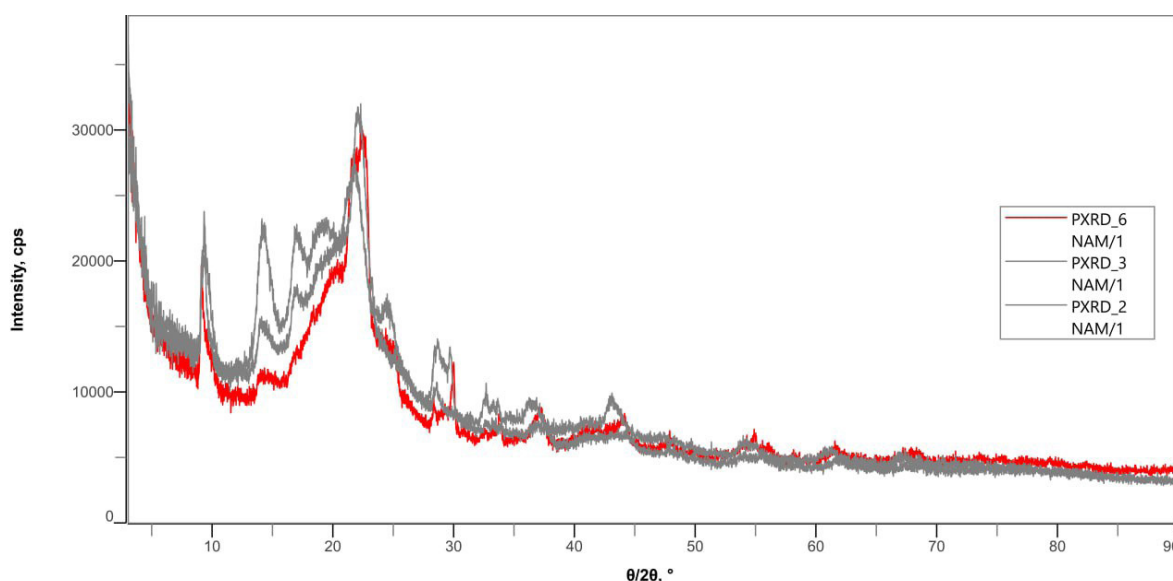


Figure 4. XRD diffractograms showing alpha-PP and orthorhombic PE/UHMWPE crystalline reflections⁸

Thermal stability is interpreted to follow the order $S5 \approx S2 > S3 > S1 > S6 > S4$. PP and UHMWPE provide relatively high thermal resistance, whereas high DOTF and rubber content may reduce onset degradation temperature. The PVC-rich S4 system requires special stabilization because PVC may undergo dehydrochlorination at elevated temperatures. For PP/PE-based systems, improved thermal stability is associated with higher crystallinity, lower plasticizer content, better dispersion, and reduced defect concentration at phase boundaries.

Thermogravimetric analysis should be used to determine the onset degradation temperature, the temperature at 5% mass loss, the maximum degradation rate temperature, and the residual char content. If the recycled rubber contains carbon black or mineral fillers, higher residue may be observed. However, high residue alone should not be interpreted as improved thermal stability; it must be considered together with degradation onset and derivative mass-loss curves.

The structural formation mechanism of PP/PE hybrid composites during melt processing can be summarized in six stages. The first stage is melting and viscosity-controlled dispersion. PP, HDPE, EVA, and rubber become deformable under shear, whereas UHMWPE softens but remains highly viscous. Therefore, UHMWPE behaves as a partially dispersed reinforcing phase rather than as a fully miscible polymer component.

The second stage is compatibilizer migration. SEBS-g-MA or MAPP migrates to PP/PE, PP/EVA, and rubber/polyolefin interfaces. This reduces interfacial tension and suppresses droplet coalescence. The third stage is phase separation. Because PP and PE are thermodynamically immiscible, phase separation occurs during melt mixing and cooling. The final domain size depends on viscosity ratio, shear rate, compatibilizer concentration, and cooling profile.

The fourth stage is heterogeneous nucleation. UHMWPE particles, rubber surfaces, and compatibilizer-rich interfaces act as nucleation sites. This increases nucleation density and reduces spherulite size. The fifth stage is crystal growth. PP and PE crystallize separately: PE-rich domains form orthorhombic crystals, while PP-rich domains form alpha-monoclinic crystals. In compatibilized systems, interfacial crystallization improves phase anchoring. The final stage is interphase locking after cooling. The composite consists of crystalline PP lamellae, PE/UHMWPE crystals, EVA/rubber soft zones, and compatibilizer-stabilized interphase regions. This multi-phase structure controls stiffness, toughness, creep resistance, and thermal stability.

CONCLUSIONS AND SUGGESTIONS

This manuscript proposed a structural formation model for hybrid PP/PE-based composites containing EVA, DOTF, recycled rubber, UHMWPE, and compatibilizer. The investigated systems form through simultaneous melt dispersion, phase separation, heterogeneous nucleation, crystallization, and interphase development. True molecular co-crystallization between PP and PE is limited; therefore, the dominant mechanism is separate crystallization of PP-rich and PE-rich phases combined with interfacial crystallization in compatibilized systems.

UHMWPE acts as both a reinforcing phase and a heterogeneous nucleation site, particularly in S3 and S5. EVA



and SEBS-g-MA or MAPP improve interfacial adhesion and reduce domain coalescence, thereby increasing impact resistance. Among the proposed compositions, S5 provides the most balanced mechanical behavior within the analyzed composition matrix because it combines PP stiffness, UHMWPE reinforcement, EVA-assisted interphase formation, low DOTF content, and rubber-based energy dissipation.

Avrami analysis is suitable for evaluating crystallization kinetics, but secondary crystallization and multi-phase crystallization must be considered for accurate interpretation. Future research should include experimental DSC kinetic curves at different cooling rates, SEM/TEM image analysis with domain-size distribution, rheological master curves, long-term aging, creep testing, and tribological evaluation under industrial contact conditions.

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2026. № 6

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