Sürdürülebilir Mühendislik Uygulamaları ve Teknolojik Gelişmeler Dergisi
Journal of Sustainable Engineering Applications and Technological Developments
2025, 8(2): 212-225

Pirinç Kabuğu Külü ile Doldurulmuş Sürdürülebilir Polipropilen Kompozitler: Geleneksel Mineral Dolgu Maddeleri ve Silan Modifikasyonu ile Karşılaştırmalı Bir Çalışma

Sustainable Polypropylene Composites Filled with Rice Husk Ash: A Comparative Study with Conventional Mineral Fillers and Silane Modification

Serkan KURT[1,2], Beyza ALTAYLI[2]

[1*]Department of Chemical Engineering, Ankara University, Çankaya-ANKARA
[2] Tisan Engineering Plastic Company ISTANBUL-TURKEY

ÖZET

Sürdürülebilir polimer sistemlerine geçiş, geleneksel mineral dolgu maddelerine biyo-bazlı alternatiflerin kullanımını gerektirmektedir. Bu çalışma, silika bakımından zengin bir tarımsal atık ürün olan pirinç kabuğu külünün polipropilen (PP) için sürdürülebilir bir dolgu maddesi olarak etkisini araştırmaktadır. Kompozitler; kalsit, talk, pirinç kabuğu külü, silan bağlayıcı madde ile birleştirilmiş pirinç kabuğu külü ve silan modifikasyonlu pirinç kabuğu külü kullanılarak çeşitli beslemelerde (%10-40 ağırlık) oluşturulmuş ve mekanik, fiziksel, termal ve morfolojik özellikleri açısından değerlendirilmiştir. Sonuçlar, mineral dolgu maddelerinin (kalsit, talk) sertliği ve sünekliği artırırken, pirinç kabuğu külünün daha düşük yoğunluk gibi ek bir avantajla performans arasında olumlu bir denge sağladığını ortaya koymuştur. Buna ek olarak, çalışmada kullandığımız silan bağlayıcı madde arayüzey yapışmasını ve dispersiyonu önemli ölçüde iyileştirerek, daha iyi yapı ve en yüksek HDT ve Vikat değerleri de dahil olmak üzere üstün termal kararlılık sağlamaktadır. Genel olarak pirinç kabuğu külü, döngüsel ekonomi prensipleriyle uyumlu, hafif, yenilenebilir ve teknik olarak rekabetçi bir dolgu maddesi olarak ortaya çıkmıştır. Bu bulgular pirinç kabuğu külünün, hafif ve sürdürülebilir malzemelere olan talebin arttığı otomotiv, elektronik ve tüketim ürünleri gibi alanlardaki uygulama potansiyelini ve önemini ortaya koymaktadır.

Anahtar Kelimeler: Pirinç kabuğu külü, Polipropilen kompozitler, Sürdürülebilir dolgu maddeleri, Yüzey modifikasyonu.

ABSTRACT

The transition towards sustainable polymer systems requires the use of bio-based alternatives to conventional mineral fillers. This study investigates the effect of rice husk ash (RHA), an agricultural waste product which is abundant in silica, as a sustainable filler for polypropylene (PP). Composites were created using calcite, talc, RHA, RHA combined with a silane coupling agent, and silane modified RHA at various loadings (10–40 wt.%) and were assessed for their mechanical, physical, thermal, and morphological characteristics. Results revealed that while mineral fillers (calcite, talc) enhanced stiffness and ductility, RHA provided a favorable balance of performance with the additional benefit of lower density. In addition to this, the silane coupling agent significantly improves interfacial adhesion and dispersion, provides better structure and superior thermal stability, including the highest HDT and Vicat values. Overall, RHA emerged as a lightweight, renewable, and technically competitive filler, aligning with circular economy principles. These findings reveal the application potential and importance of rice husk ash in areas such as automotive, electronics and consumer products, where the demand for lightweight and sustainable materials is increasing.

Keywords: Rice husk ash (RHA), Polypropylene composites, Sustainable fillers, Surface modification.

Başvuru: 17.10.2025 Revizyon:23.10.2025 Kabul: 06.11.2025
Doi: 10.51764/smutgd.1805493

[1*]Department of Chemical Engineering, Ankara University, Çankaya-ANKARA- skurt@tisan.com.tr; ORCID: 0009-0009-0761-9192
[2] Tisan Engineering Plastic Company ISTANBUL-TURKEY; beyza.altayli15@gmail.com ORCID: 0009-0000-5666-2079

1. INTRODUCTION

In recent years, global research in materials science has prioritized the development of environmentally friendly polymeric systems and the modification of existing production technologies to achieve sustainability goals. The increasing importance of environmental challenges and the decreasing access to raw materials are encouraging researchers to incorporate renewable and biodegradable materials into polymer matrices (Mistry et al., 2015). Over the past two decades, organic fillers have emerged as a viable alternative to traditional inorganic fillers due to their lower density, lower cost, and non-corrosive processing characteristics compared to non-renewable fillers, higher filler loading, recyclability, and biodegradability (Shen et al., 2024). Overall, these organic materials (like wood flour (Chakir et al., 2015), bamboo (Behera et al., 2015), cotton (Wankhede et al., 2023), sisal (Glowinska et al., 2017), wheat (Majewski et al., 2018), flax (Islam et al., 2025), jute (Ashraf et al., 2019), corn (Mohammed et al., 2022), rice husk (Duarte et al., 2013), coconut shell (Radhakrishnan et al., 2025), banana (Kumar et al., 2024) and pineapple (Kumar et al., 2022)) provide superior performance to final products, directly impacting environmental outcomes, particularly in terms of carbon emissions, and are crucial for sustainable materials development (Cutright et al., 2013).

One of the organic materials, rice husk, is an important byproduct of the rice milling process and stands out for its advantages such as non-abrasiveness, low cost, and light weight (Rahman et al., 2010). Thanks to these properties, composites made from rice husk and thermoplastics are becoming increasingly important for environmentally friendly and cost-effective products. Furthermore, the utilization of rice husk adds value to agricultural waste, supporting both the rural economy and providing a solution to waste management problems. Rice husk consists of approximately 35% cellulose by dry weight, 35% hemicellulose, 20% lignin, and 10% ash (94% silica) by weight. This natural structure makes rice husk ash (RHA), obtained by incineration of rice husk and comprising approximately 10% of its dry weight, a valuable resource with great potential for industrial applications (Salih et al., 2014; Nurhayati & Susanto, 2020).

Rice husk ash (RHA) is a useful and sustainable material thanks to its rich silica content and structure. It improves the mechanical properties of polymer composites while offering environmental solutions and waste management alternatives. Rice husk ash is a renewable resource that supports productions from bio based or waste materials research. Polymer science research has explored the potential for combining RHA with other materials such as polypropylene (PP) and polyethylene (PE) (Kenechi et al., 2016).

Polypropylene (PP) is a high-performance thermoplastic with great versatility in industry. The applications of PP range from food packaging to automotive components, wrapping materials, and textiles, making it an essential material for product designers because of its properties such as lightweight, durability, flexibility, and resistance to corrosive materials (Raghu et al., 2018; Hossain et al., 2024). The main disadvantage of polypropylene is its non-polar nature and its inability to bond easily with some fillers such as rice husk, talc, calcite, etc. Typically, this incompatibility will result on polymer composites reinforced with fillers a weak or unstable interface that negatively impacts mechanical stability. (Duy Tran et al., 2013).

To mitigate the shortcomings of polypropylene's non-polar nature, surface modification techniques are crucial in enhancing the bond strength between the filler and polymer matrix. In this regard, there are many different types of surface modification techniques that will truly enhance interfacial adhesion by adding functional groups on the surface of the filler, which can then chemically or physically bond to polypropylene (Kumar & Kumar, 2022). Compatibilizers such as maleic anhydride-grafted polypropylene (MAPP) (Petchwattana et al., 2012; Morales et al., 2025) and silane (Zhang et al., 2025; Guo et al., 2025) can perform this task, as well. Among these chemical treatments, alkaline treatment (mercerization), benzylation, acetylation, treatment with stearic acid, peroxide treatment, permanganate treatment, isocyanate treatment, and plasma treatment are the other important surface modification technics (Halip et al., 2021). On the other hand silane, titanate and maleic anhydride compatibilizers are reactive agents and provide the formation of chemical bonds between the polymer and the inorganic filler (Lee et al.,2017).

While MAPP-based compatibilizers are added to the polymer composite structure containing fillers at certain rates simultaneously during the processing steps, silane compatibilizers are used to modify the filler surface. Metin et al. (2003) reported that by surface treatment of natural zeolite with three different types of silane coupling agents; 3-aminopropyltriethoxysilane (AMPTES), methyltriethoxysilane (MTES) and 3-mercaptopropyltrimethoxysilane (MPTMS), mechanical properties of the zeolite reinforced PP composite significantly improved. Ansari&İsmail (2009) produced 3-(aminopropyl) triethoxy silane (3-APE) treated feldspar reinforced PP by using melt mixer and the tensile strength, elongation at break, Young's modulus, and impact strength were found to be increased in silane treated composites.

Diversity of compatibilizers and surface modification methods reported in the literature to improve interfacial bonding and mechanical performance of PP composites reinforced with lignocellulosic or mineral fillers have been explored. Kabir et al. (2014) reported that in rice husk-filled liquid-based resin composites, the filler content and particle size significantly affect tensile strength, Young's modulus, elongation at break, flexural strength, and impact strength. Yang et al. (2004) produced rice husk flour-filled polypropylene (PP) composites and found that increasing the filler content led to a slight decrease in tensile strength. The mechanical properties of rice husk powder-filled PP composites were compared with those of talc-filled PP composites by Premalal et al. (2002), who reported that in both composites, an increase in filler content resulted in higher Young's modulus and flexural modulus, but lower yield strength and elongation at break. Tanış et al. (2021) observed an increase in flexural and tensile modulus, but a decrease in tensile strength, with increasing RHA content in polypropylene/RHA composites. Fuad et al. (1995) reported that in polypropylene–RHA composites, the flexural modulus increased with filler content, while tensile strength and elongation at break decreased. In this research, since silane coupling agents were also employed, particular attention was given to their role in improving interfacial adhesion and enhancing the mechanical performance of the composites.

Unlike previous research, this study aimed at the development of high-strength polypropylene compounds reinforced with rice husk ash (RHA) as a sustainable alternative to conventional fillers, with the goal of improving the mechanical and physical properties of the material. Furthermore, the performance of RHA-reinforced compounds, including those with RHA surface-modified with silane and those produced with the addition of a silane coupling agent, was comparatively evaluated against polypropylene compounds reinforced with conventional fillers such as calcite and talc. In all formulations, 3 wt. % MAPP, antioxidant, and internal and external lubricants were used at constant ratios.

2. EXPERIMENTAL

2.1 Materials

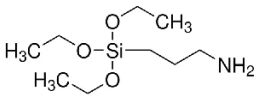
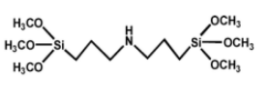
In this study, polypropylene (PP) homopolymer with a melt flow index of 25 g/10 min (230°C/2.16 kg) and a density of 0.905 g/cm^3 was used. The content shown in Table 1, gray colored rice husk ash (RHA) with a density of 0.18–0.35 g/cm^3, calcite with particle sizes of D_{50}: 5.4 µm and D_{97}: 21.5 µm, and talc with D_{50}: 5.3 µm and D_{97}: 22 µm were selected as fillers. MAPP, with a melt flow index of 115 g/10 min (190°C/2.16 kg) and a density of 0.9 g/cm^3, was used as the binding agent. Phenolic and phosphite-based antioxidants were used for stabilization; an external and an internal lubricant were used for ease of processing. A silane-based coupling agent was also added to PP matrix and used for surface modification of RHA to enhance the filler-matrix interaction. Ethanol, acetic acid, and distilled water were used as the solvent in the silica surface treatment process.

Table 1. Compositional analysis of the rice husk ash

Content, wt.%			
SiO_2	88-97%	**CaO**	1.5%
$N_2O + Na_2O$	2-4%	**Fe_2O_3**	1.0%
MgO	1.5%	**C**	1.5-3%
Al_2O_3	1.0%	**Remainder**	<2.0%

A combination silane coupling agent containing 80% 3-Aminopropyltriethoxysilane (APTES) and 20% Bis(triethoxysilylpropyl)amine (Bis-aminosilane) was used in the study. Table 2, summarizes the properties of the silane coupling agent.

Table 2. Characteristics of the Silane Coupling Agent

Silane coupling agent name	Chemical structure	Density (g/cm^3)	Flash point (°C)	Viscosity (mPa·s)
3-Aminopropyltriethoxysilane (APTES) %80		0,95	98	2
Bis(triethoxysilylpropyl)amine (Bisaminosilane) %20				

2.2. RHA Surface Treatment and Polymer Composites Preparation

The experimental work was carried out in three stages. In the first stage, composites containing calcite, talc and RHA were prepared separately at filler loadings of 10, 20, 30, and 40 wt%. In the second stage, RHA-filled polypropylene composites were produced with a fixed amount of silane (1.5 wt%) coupling agent, again at 10, 20, 30, 40 wt% filler loadings. For performing the surface treatment of silica samples, wet method was employed in in the third stage and rice husk ash (RHA) was surface-modified with silane to enhance filler–matrix interfacial adhesion, improve dispersion, and reduce agglomeration within the polypropylene matrix. In this stage, polypropylene composites were prepared with 10 wt% RHA surface-modified with silane prior to incorporation.

The silane coupling agent was hydrolyzed in a 90:10 ethanol–water solution (v/v) at 70°C for 1 h using magnetic stirrer and pH is adjusted to 5 using acetic acid. Hydrolysis mechanism of silane summarized in Figure 1.

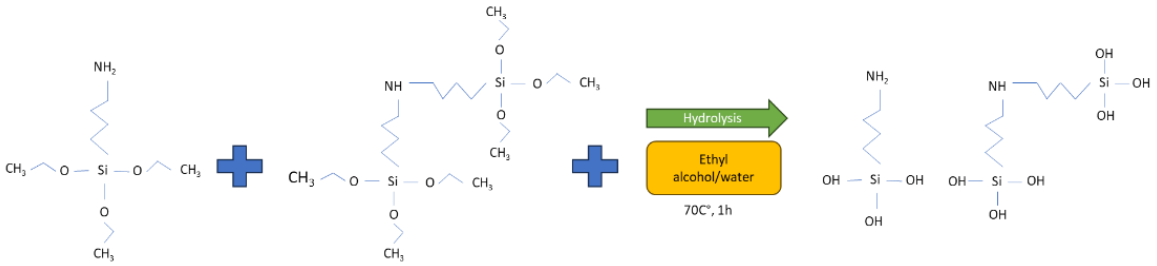


Figure 1. Schematic mechanism of silane hydrolysis

RHA was then added to the hydrolyzed solution and stirred at 70°C for 1 h to allow silane molecules to react with the surface hydroxyl groups, resulting in silane coating. After coating, the treated RHA is dehidrated/cured at 105°C for 3 h to complete condensation and remove volatiles, followed by final drying. The surface-modified RHA was subsequently incorporated into polypropylene at a loading of 10 wt%.

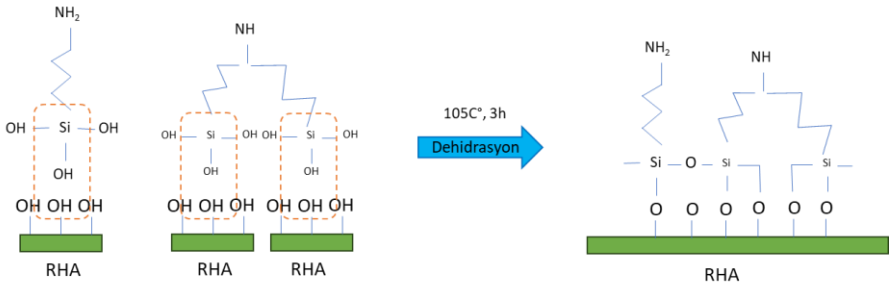


Figure 2. Schematic mechanism of silane dehydration and curing

As shown in Figure 2, hydroxyl groups of silanes interact with the hydroxyl groups on the silica-containing RHA surface, forming H-bonding. In the next step, curing occurs by dehydration, hydroxyl groups are removed and Si-O-Si covalent bonding is achieved.

As seen in Table 3 with sample codes and rates, in all formulations polypropylene was melt-compounded with the selected fillers (rice husk ash, calcite or talc) in varying loadings (10–40 wt.%), together with 3 wt.% MAPP coupling agents, antioxidant, and internal and external lubricants at constant ratios. Figure 3 schematically illustrates the preparation of RHA-based PP composite. The compounding process was carried out in a prototype co-rotating twin-screw extruder (L/D ratio of 46 and a screw diameter of 30 mm) at a temperature profile of 210–240°C and a screw speed of 350–400 rpm, with a throughput capacity of 30-35 kg/h. The extrudates were pelletized to obtain PP compound granules, which were subsequently injection-molded into test specimens. Potential application areas of the developed compounds include under-the-hood automotive parts, instrument panels, interior trims, and exterior claddings, in which these materials are included.

Table 3. Composite formulations showing filler type and filler content.

Sample Code	Polymer (PP)	Calcite	Talc	RHA	MRHA*	Silane	MAPP	Antioxidant and Lubricant
C1	86.1	10	-	-	-	-	3	0.9
C2	76.1	20	-	-	-	-	3	0.9
C3	66.1	30	-	-	-	-	3	0.9
C4	56.1	40	-	-	-	-	3	0.9
T1	86.1	-	10	-	-	-	3	0.9
T2	76.1	-	20	-	-	-	3	0.9
T3	66.1	-	30	-	-	-	3	0.9
T4	56.1	-	40	-	-	-	3	0.9
R1	86.1	-	-	10	-	-	3	0.9
R2	76.1	-	-	20	-	-	3	0.9
R3	66.1	-	-	30	-	-	3	0.9
R4	56.1	-	-	40	-	-	3	0.9
RS1	84.6	-	-	10	-	1.5	3	0.9
RS2	74.6	-	-	20	-	1.5	3	0.9
RS3	64.6	-	-	30	-	1.5	3	0.9
RS4	54.6	-	-	40	-	1.5	3	0.9
RSS	86.1	-	-	-	10	-	3	0.9

All values are expressed as weight percent (wt.%).
***MRHA represents RHA modified with silane.**

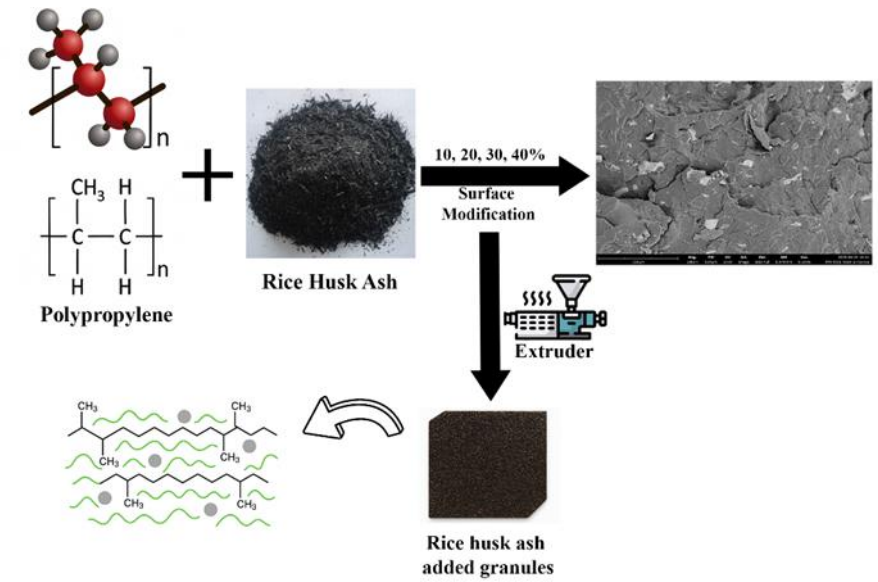


Figure 3. Schematic representation of the preparation of RHA-based PP composites.

2.3. Determination of Physical Properties

Density was measured in accordance with ISO 1183 using the immersion method. Gloss was evaluated in accordance with ISO 2813 using a CS-380 Mini Gloss Meter. Specimens were cleaned, dried, and smoothed before measurement. The device was operated at geometries of 20°, 60°, and 85°, depending on surface gloss level; in this study, 60° measurements were taken as the reference.

2.4. Determination of Mechanical Properties

2.4.1. Tensile Tests

Tensile tests were conducted in accordance with ISO 527-1 and ISO 527-2 using a universal testing machine equipped with a ZwickRoell Longstroke extensometer. Specimens were prepared according to the standard dimensions (170 mm × 10 mm × 4 mm) and fixed in the machine grips.

2.4.2. Impact Strength

Impact strength was measured in accordance with ISO 180 using a ZwickRoell HIT5.5P pendulum impact tester. Specimens with dimensions of 80 mm × 10 mm × 4 mm were conditioned at room temperature prior to testing. For the notched test, a V-notch (45° angle, 2 mm depth, 0.25 mm base radius) was machined. Specimens were clamped vertically in an Izod-type fixture, and a pendulum with a nominal energy of 5 J was released from a specified height.

2.4.3. Shore D hardness

Shore D hardness was determined in accordance with ISO 868 using a ZwickRoell Shore-D analog hardness tester. Measurements were taken at 23 ± 2°C on flat, smooth specimen surfaces. The indenter was applied perpendicularly with uniform pressure until the presser foot fully contacted the surface, and the hardness value was read after 15 s, as specified in the standard.

2.5. Determination of Thermal Properties

2.5.1. Heat deflection temperature (HDT) and Vicat

Heat deflection temperature (HDT) and Vicat softening temperature measurements were carried out in accordance with ISO 75 and ISO 306, respectively, using a ZwickRoell HDT/Vicat tester. In HDT testing, standard-sized specimens were heated in an oil bath under a bending stress of both 1.80 MPa and 0.45 MPa, and the temperature at 0.34 mm deflection was recorded. In Vicat testing, loads of 10 N and 50 N were applied, the heating rate was 50 ± 5°C/h, and the temperature corresponding to 1 mm indentation was determined. Three replicates were performed for each measurement, and the mean values were reported.

2.6. Morphology

Morphological analyses were carried out using a Thermo Scientific Phenom XL G2 desktop scanning electron microscope. Prior to imaging, specimens were sputter-coated with a thin layer of gold to improve surface conductivity and minimize charging effects.

3. RESULTS AND DISCUSSION

3.1. Mechanical Properties of PP Composites with Different Fillers

In this study, the mechanical properties of polypropylene (PP) composites reinforced with different fillers, including conventional mineral-based fillers (calcite and talc) and sustainable alternatives (rice husk ash (RHA)), were comparatively evaluated. Furthermore, the effect of silane coupling agent addition (RS series) and silane surface modification of RHA (RSS series) on the mechanical performance of PP composites was investigated.

Test results that reflecting the mechanical properties of the calcite and talc reinforced PP composite are presented in Table 4, RHA reinforced PP composite results are presented in Table 5 with standard deviation (std).

Table 4. Calsite and talc reinforced PP composite test results

Test Parametresi	Tensile Stress (Mpa)+std	Elongation at Yield (%)+std	Elastic Modulus (Mpa) +std	Notch IZOD Impact Strenght (kj/m²) +std	Tensile Stress at Break (Mpa) +std	Strain at Break (%)+std	SHORE D Hardness +std	Density (g/cm³) +std
C-1 (C-%10)	29,79±0,35	7,43±0,38	1125±58	3,4±0,3	16,8±0,51	40,34±1,91	63±1	0,9745±0,001
C-2 (C-%20)	25,31±0,18	5,4±0,27	1150±42	4±0,34	17,84±0,81	30,62±1,57	65±0,5	1,04±0,008
C-3 (C-%30)	23,43±0,34	4,58±0,21	1414±62	4,23±0,41	17,6±0,78	14,25±1,14	65±1	1,145±0,003
C-4 (C-%40)	20,4±0,44	4,5±0,2	1568±42	3,1±0,35	18±0,70	50±1,82	69±0,5	1,24±0,009
T-1 (T-%10)	37,7±0,4	5,5±0,13	1.996±72	3,2±0,51	34±0,98	11,7±1,5	59±0,5	0,976±0,01
T-2 (T-%20)	35,5±0,44	6,1±0,14	1.990±42	3,75±0,61	25±0,77	6,5±1,61	65±1,5	1,05±0,007
T-3 (T-%30)	36,8±0,38	3,5±0,18	2.612±92	3±0,21	34,5±0,48	3,9±1,11	60±0,5	1,146±0,005
T-4 (T-%40)	36,1±0,41	3,6±0,2	2.565±121	2,78±0,27	35,5±0,54	4,1±0,93	61±0,5	1,225±0,003

Table 5. RHA and Modified RHA reinforced PP composite test results

Test Parametresi	Tensile Stress (Mpa) +std	Elongation at Yield (%)+std	Elastic Modulus (Mpa) +std	Notch IZOD Impact Strenght (kj/m²) +std	Tensile Stress at Break (Mpa) +std	Strain at Break (%)+std	SHORE D Hardness +std	Density (g/cm³) +std
R-1 (R-%10)	30,7±0,68	5,7±0,78	1.380±78	2,5±0,15	28,3±0,81	11,5±0,74	60±1	0,96±0,008
R-2 (R-%20)	29,5±0,71	4,5±0,83	1.550±51	1,9±0,21	27,1±0,79	14,2±0,81	62±1	1,02±0,007
R-3 (R-%30)	27,8±0,61	2,6±0,21	2.010±91	1,6±0,08	25,5±0,7	3,6±0,51	60±0,5	1,08±0,008
R-4 (R-%40)	27,6±0,41	1,7±0,08	2.725±142	1,5±0,07	27,6±0,73	1,7±0,1	63±1	1,147±0,01
RS-1 (R-%10)	33,1±0,48	3,9±0,21	1.995±131	2,4±0,1	32,5±0,65	4,5±0,15	59±1	0,967±0,002
RS-2 (R-%20)	35,5±0,81	4,1±0,33	1.850±110	2,1±0,2	29,5±0,68	6,5±0,44	61±0,5	1,019±0,005
RS-3 (R-%30)	36,1±0,77	3,5±0,44	2.550±128	1,7±0,03	36,1±0,71	3,5±0,14	61±0,5	1,084±0,003
RS-4 (R-%40)	35±0,48	3,3±0,41	2.450±123	1,7±0,05	35±0,7	3,3±0,11	57±1	1,14±0,004
RSS-1 (R-%10)	32,1±0,53	7,8±0,61	1.303±51	2,5±0,2	31,5±0,64	8,7±0,65	57±1	0,945±0,004

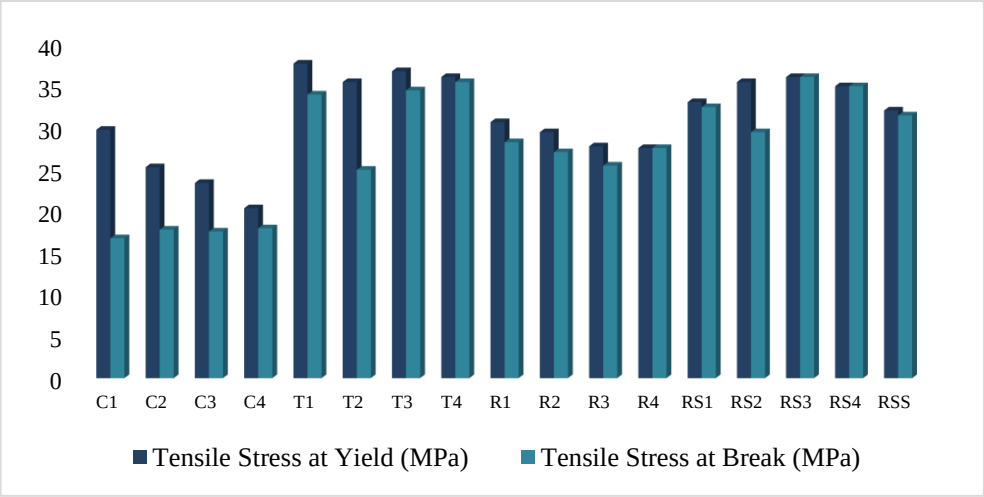


Figure 4. Comparison of Tensile Stress at Yield and Break of PP Composites with Different Fillers.

The tensile performance of PP composites, in terms of tensile stress at yield and tensile stress at break, is presented in Figure 4. The C series showed decreasing yield values with increasing filler content, while tensile stress at break remained relatively low. The T series exhibited the highest values for both yield and break, indicating strong reinforcement. The R series demonstrated moderate performance, outperforming the C series but remaining below the T series. The RS series improved both yield and break values, approaching those of the T series, while the RSS sample achieved comparable results, highlighting the effectiveness of surface modification even at low filler loading.

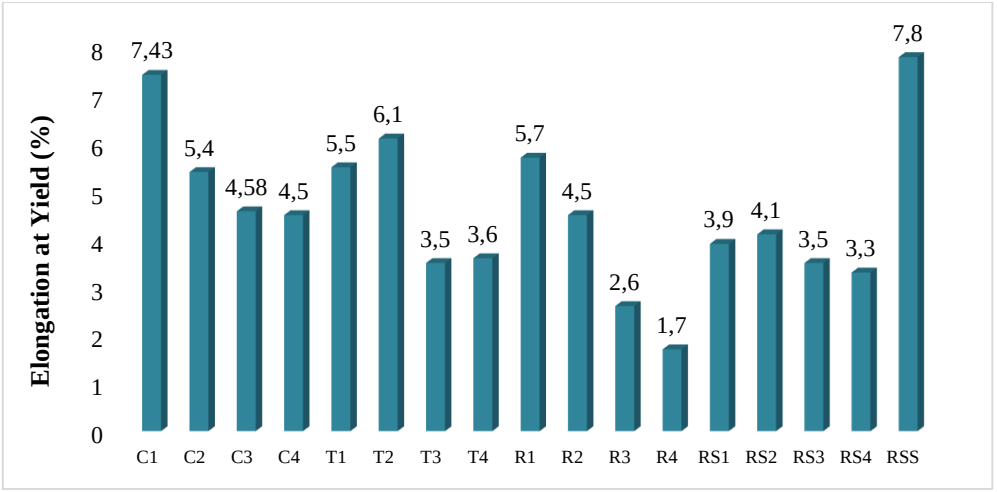


Figure 5. Comparison of Elongation at Yield of PP Composites with Different Fillers.

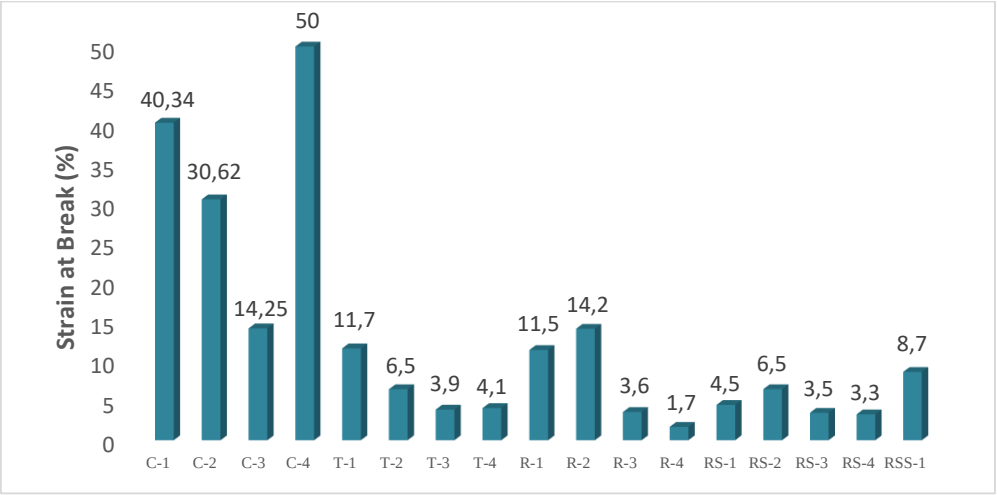


Figure 6. Comparison of Strain at Break of PP Composites with Different Fillers.

The elongation behaviour of PP composites, in terms of elongation at yield and strain at break, is summarized in Figure 5 and 6, respectively. The C series showed decreasing elongation at yield with increasing filler content, while strain at break values remained relatively high compared to other series. The T series exhibited relatively low elongation for both parameters, indicating stiff behaviour. The R series demonstrated moderate values, generally higher than the T series but lower than the C series. The RS series slightly improved strain at break in some samples, though values remained moderate. The RSS sample achieved relatively high elongation at yield and moderate strain at break, suggesting that surface modification helps maintain flexibility while reinforcing the matrix.

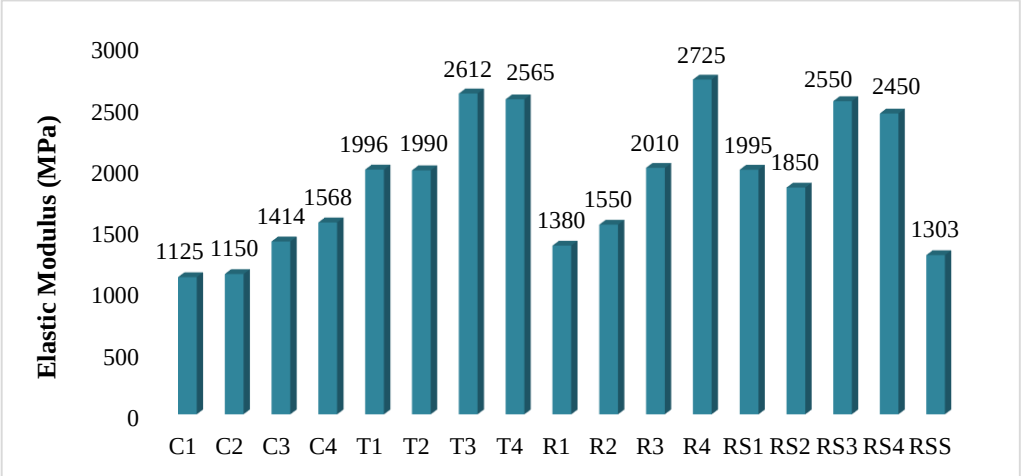


Figure 7. Comparison of Elastic Modulus for PP Composites with Different Fillers.

The elastic modulus values of PP composites reinforced with different fillers are summarized in Figure 7. C series composites exhibited the lowest stiffness, indicating that calcite provides limited stiffness to the matrix. T series and R series exhibited similar or higher stiffness values, demonstrating that these filler types effectively strengthen the matrix. RS series generally provided higher stiffness than R series, indicating that improved interfacial adhesion enhanced mechanical performance. RSS, on the other hand, exhibited lower stiffness values, suggesting that the applied modification has a limited effect on this property.

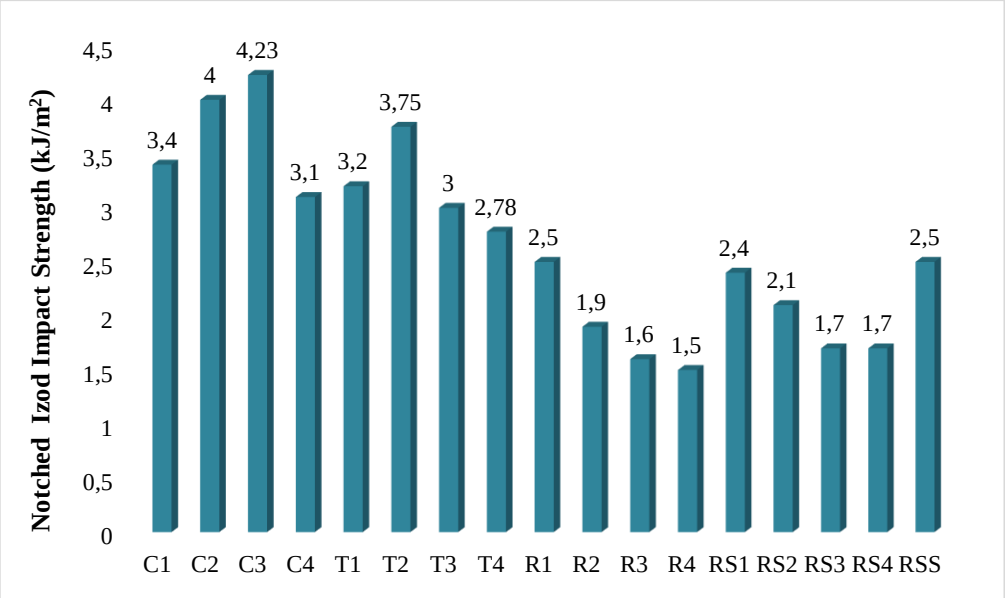


Figure 8. Comparison of Notched Izod Impact Strength for PP Composites with Different Fillers.

The notched Izod impact strength of PP composites reinforced with different fillers is presented in Figure 8. The C series composites exhibited relatively stable and moderate impact resistance across all filler loadings. The T series exhibited moderate impact resistance with slight fluctuations but decreased at higher filler loadings. The R series generally had lower impact resistance, decreasing with increasing filler content. RS series partially increased impact resistance, with a decrease observed in some samples. RSS provided moderate impact resistance and performed similarly to the RS series.

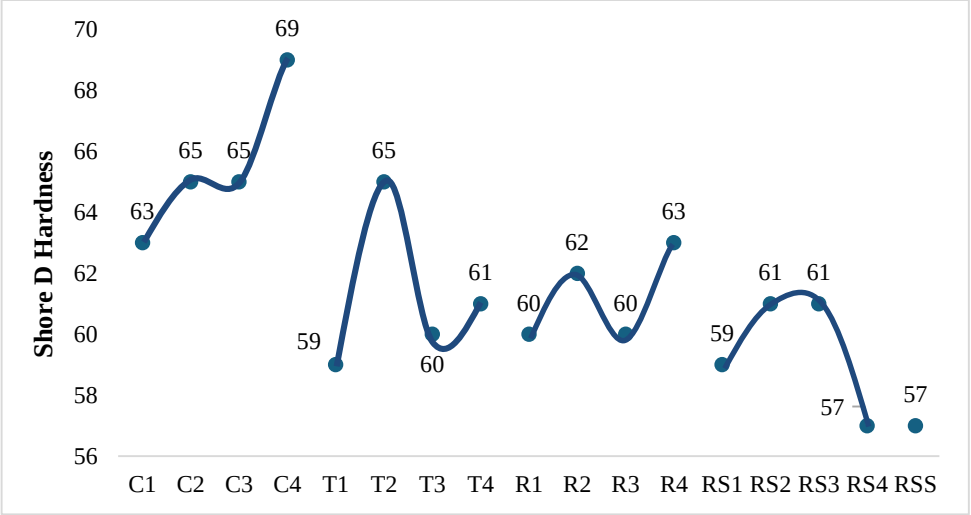


Figure 9. Comparison of Shore D Hardness for PP Composites with Different Fillers.

Shore D hardness values of PP composites reinforced with different fillers are summarized in Figure 9. Surface hardness increased significantly with increasing filler content in the C series composites. The T series offered moderate hardness, with a slight improvement observed with increasing filler content. The R series provided only a limited increase in hardness. RS series did not significantly differ from R series in hardness, while RSS performed similarly to the RS series with moderate hardness values.

3.2. Physical Properties of PP Composites with Different Fillers

The physical properties of the PP composites, such as density and surface gloss, were investigated to evaluate the effects of different fillers and surface modification.

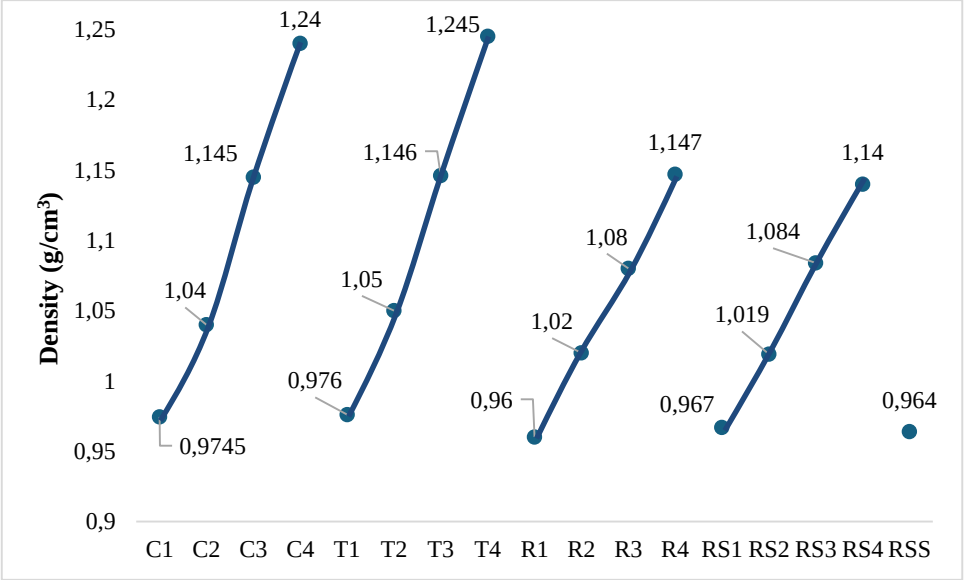


Figure 10. Comparison of Density for PP Composites with Different Fillers.

The effect of different fillers on the density of PP composites is illustrated in Figure 10. The C and T series composites showed a significant increase in density with increasing filler content, reaching the highest density values among all series. Since RHA is used in all of the R, RS and RSS series, no change in density values is observed in these series. RHA has a specific gravity of 2.05 g/cm^3 compared to 2.7 g/cm^3 of talc and calcite, which makes the RHA reinforced polymer composite structure lighter.

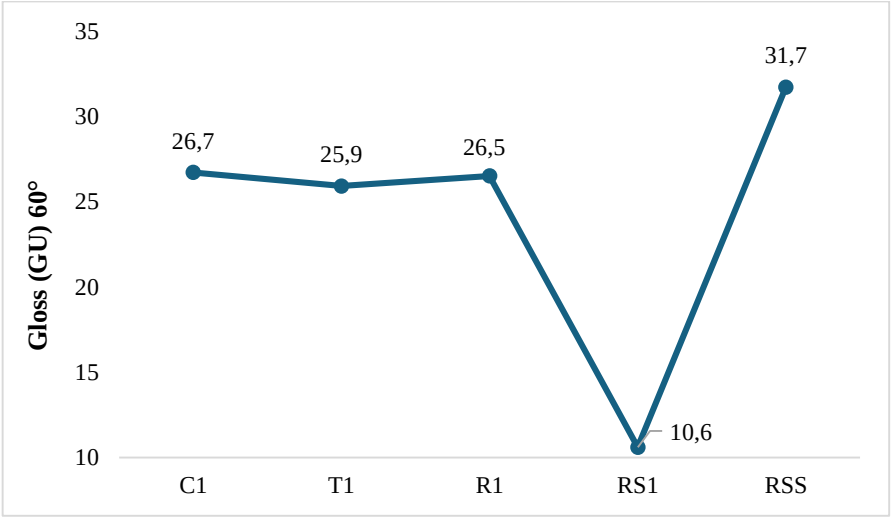


Figure 11. Comparison of Surface Gloss for PP Composites with Different Fillers.

The evaluation of surface gloss for PP composites, conducted at a measurement angle of 60°, is presented in Figure 11. Composites C, T and R series exhibited similar to moderate surface gloss with average 26,4 and revealing that filler type has a limited effect on gloss. Besides this RS series exhibited a significant decrease in gloss, indicating that the silane binder may increase surface roughness or reduce light reflectance. In contrast, RSS achieved the highest gloss, demonstrating that the applied surface modification on RHA significantly improves the visual appearance of the composite.

3.3. Thermal Properties of PP Composites with Different Fillers

Table 6. HDT and Vicat softening temperature of PP composites containing various fillers.

Sample Code	Vicat B (°C)	Vicat A (°C)	HDT A (°C)	HDT B (°C)
PP	101.3	156.0	63.53	72.02
C1	102.8	157.5	70.45	78.38
T1	105.8	158.5	66.73	83.09
R1	104.5	157.5	66.93	76.37
RS1	107.8	157.9	75.79	89.98
RSS	101.7	157.8	65.41	77.41

In addition to mechanical and physical characterizations, the thermal performance of PP composites containing different fillers was evaluated through Heat Deflection Temperature (HDT) and Vicat softening temperature tests. As seen in Table 6, the results revealed that filler type significantly influenced thermal behaviour. Compared to unfilled PP, RS1 exhibited the highest values, which can be explained by the stronger interphase interaction between the polymer matrix and the filler due to the binding agent. In contrast, C1, T1, and R1 showed limited improvement, while RSS performed below expectations.

3.4. Morphology of PP Composites

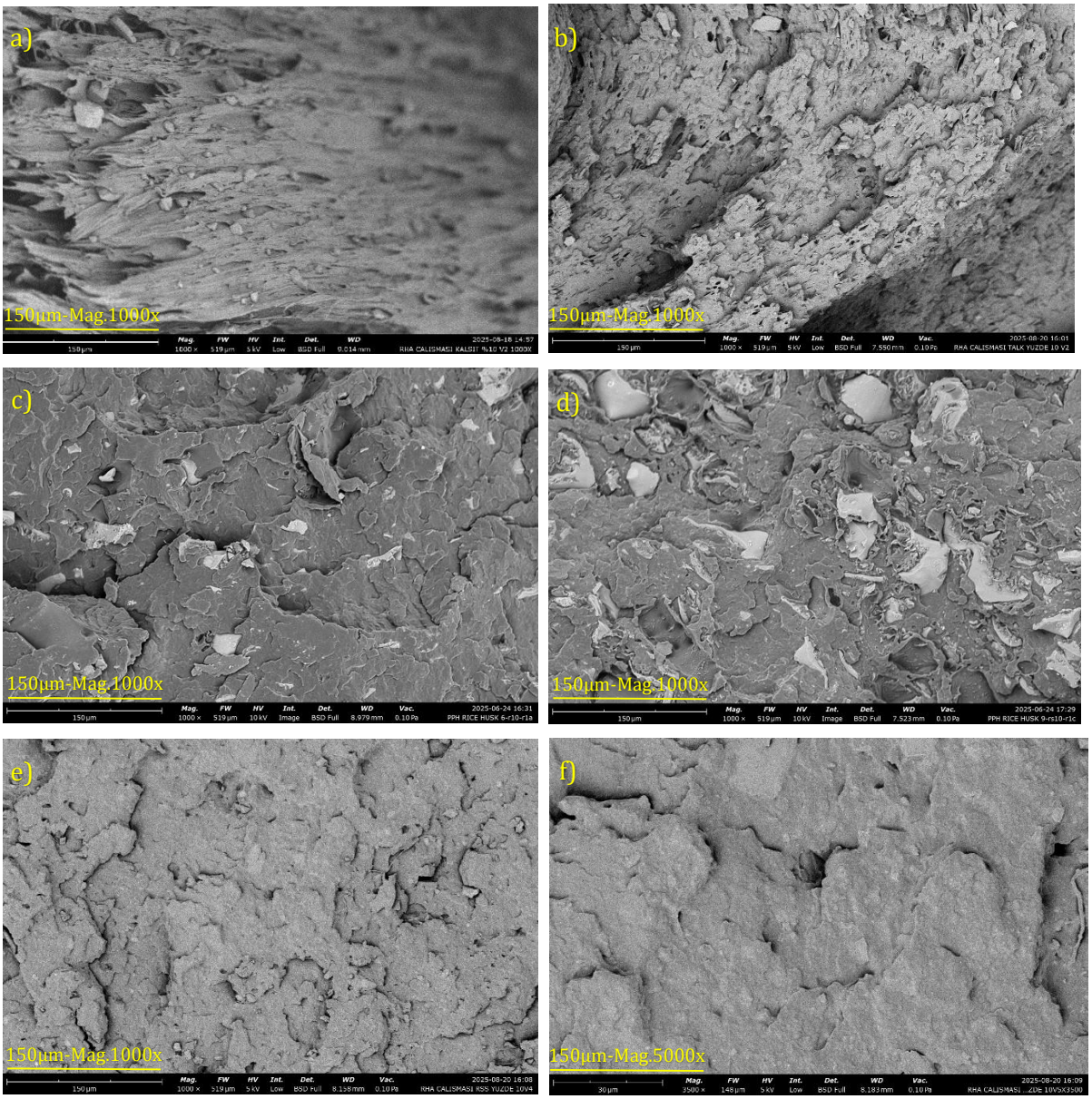


Figure 12. SEM Images of PP Composites **a)** C1 1000x **b)** T1 1000x **c)** R1 1000x **d)** RS1 1000x **e)** RSS 1000x **f)** RSS 5000x magnification.

Figure 12 presents SEM images at 1000x and 5000x magnification, showing the effects of different filler types and surface modification on the PP matrix. PP with 10% calcite (a) exhibits a relatively smooth and compact fracture surface because of regarding at particle size distribution. PP with 10% talc (b) shows a layered and slightly rough fracture surface. PP with 10% RHA (c) contains noticeable voids and separations within the matrix, whereas RHA with silane coupling agent (d) demonstrates a more homogeneous distribution and improved interfacial interaction. Silane-modified RHA (e) exhibits a compact and integrated structure. The high-magnification (5000x) image of the same sample (f) further highlights reduced voids and stronger bonding at the interface. These observations indicate that surface modification can improve interfacial interactions and structural integrity in RHA-containing PP composites.

4. Conclusion

This study presents a comparative evaluation of polypropylene (PP) composites with conventional mineral fillers (calcite and talc) and rice husk ash (RHA), with a particular focus on silane coupling agents and surface modification. The results showed that the type and treatment of the filler play an important role in the overall performance of PP composites. As anticipated, talc showed the best performance for tensile stress at yield, while calcite maintained higher strain at break values. RHA, showed marked improvements, especially with combinations of silane coupling agent (RS series) and with surface modification (RSS). Although RSS exhibited some benefits in surface appearance, overall RS composites had a more consistent mechanical performance, had strengths comparable to talc and stronger than calcite.

While mineral fillers increased the density of polypropylene composites, RHA-based composites exhibited lower density due to the lower specific gravity of RHA compared to talc and calcite. For example, the composite containing 40 wt.% RHA had a density approximately 10% lower than those with talc and calcite, indicating its potential for lightweight applications. It is considered that reducing the weight of polymer composites by 10% will contribute to sustainability-oriented targets by providing less fuel consumption and efficiency in the automotive industry.

Impact resistance was generally reduced at higher RHA contents. Gloss performance varied depending on the filler; silane-treated RHA reduced gloss, while surface-modified RHA significantly increased gloss and demonstrates that it may provide application-specific advantages in visually appealing areas such as vehicle interior parts. Thermal characterizations are HDT and Vicat tests, confirmed that RHA-based composites can meet operating temperature requirements for applications needing heat resistance, such as automotive under-the-hood parts or electronic housing. RS1 demonstrated improved thermal performance compared to all other samples, supporting its applicability under harsh conditions. SEM analyses indicated that surface modification of RHA generally improved interfacial adhesion and contributed to mechanical performance improvements. By surface modification of RHA, higher stress at yield and strain at break values with higher elongation and therefore better toughness was obtained compared to other RHA samples.

Being an agricultural by-product, it contributes to sustainability by reducing reliance on other commercial mineral-based fillers, lowering carbon footprint, and supporting circular economic practices. The use of silane-modified RHA as a filler in polymer matrices can maintain or improve important mechanical properties, especially those that achieve the mechanical properties of talc and surpass those of calcite, and also provide advantages in terms of density reduction, high thermal stability and high surface gloss. These combined benefits suggest potential applications in sectors such as automotive components, consumer electronics and electric vehicles where lightweight; durability, environmental effects and sustainability are important.

In summary, RHA represents a viable alternative to conventional mineral fillers in polypropylene. Further research on optimization of modification strategies and long-term performance evaluation will strengthen the industrial feasibility of RHA-based composites.

References

Ansari, M. N. M. & İsmail, H. (2009). The Effect of Silane Coupling Agent on Mechanical Properties of Feldspar Filled Polypropylene Composites. *Journal of Reinforced Plastics and Composites, Vol. 28, No. 24/2009.*

Ashraf, M. A., Zwawi, M., Taqi Mehran, M., Kanthasamy, R., & Bahadar, A. (2019). Jute Based Bio and Hybrid Composites and Their Applications. *Fibers, 7(9), 77.*

Behera, D., Pattnaik, S. S., Nanda, D., Mishra, P. P., Manna, S., & Behera, A. K. (2025). A Review on Bamboo Fiber Reinforced Composites and Their Potential Applications. *Emergent Materials, 8(1), 157-172.*

Chakir, A., Alami, M., Assouag, M., El Amarty, F., & Noureddine, O. (2024). Utilization of Wood Flour Waste as a Filler

on Polypropylene Random Pipes Industry. *Ecological Engineering & Environmental Technology, 25.*

Cutright, T., Mahoney, M., Franey, K., & Patnaik, A. (2013). Carbon Footprint Assessment of Polypropylene Fiber Reinforced Concrete Floors. *The International Journal of the Constructed Environment, 3(1), 73.*

Duarte, G. W., Fiori Jr, J., Peterson, M., Fiori, M. A., Colle, G. N., Riella, H. G., & Neto, A. B. S. S. (2013). Effect of Mass Flow and Mold Temperature in the Mechanical Properties of Polypropylene-Rice Rusk Ash. *Advances in Materials Science and Applications, 2, 101-109.*

Radhakrishnan, S., Krishna, J. S., Dwivedi, S. P., Gupta, S., Gupta, P., & Chaudhary, V. (2025). Experimental Investigation of Mechanical and Physical Properties of Coconut Shell and Eggshell Filler-Based Bio-Fiber Reinforced Epoxy Hybrid Composites. *Biomass Conversion and Biorefinery, 15(2), 3201-3213.*

Głowińska, E., Datta, J., & Parcheta, P. (2017). Effect of sisal fiber filler on thermal properties of bio-based polyurethane composites. *Journal of Thermal Analysis and Calorimetry, 130(1), 113-122.*

Guo, J., Xu, M., Li, H., Huang, X., Lin, H., Jiang, S., ... & Chen, C. (2025, May). Molecular Dynamics Simulation of The Effect of Silane Coupling Agent-Modified SiO_2 on the Diffusion of Water Molecules in Polypropylene. *Journal of Physics: Conference Series (Vol. 3009, No. 1, p. 012063). IOP Publishing.*

Halip J. A., Lee S. H., Tahir P. M., Chuan L. T., Selimin M. A, Saffian H. A., (2021). A Review: Chemical Treatments of Rice Husk for Polymer Composites. *Biointerface Research in Applied Chemistry, Volume 11, Issue 4, 2021, 12425 – 12433.*

Hossain, M. T., Shahid, M. A., Mahmud, N., Habib, A., Rana, M. M., Khan, S. A., & Hossain, M. D. (2024). Research and Application of Polypropylene: A Review. *Discover Nano, 19(1), 2.*

Islam, S., Hasan, M. B., Karim, F. E., Kodrić, M., Islam, M. R., Khatun, M. M., & Motaleb, K. A. (2025). Thermoset and thermoplastic polymer composites reinforced with flax fiber: Properties and application—A review. *SPE polymers, 6(1), e10172.*

Kabir, H., Gafur, M. A., Ahmed, F., Begum, F., & Qadir, M. R. (2014). Investigation of Physical and Mechanical Properties of Bamboo Fiber and PVC Foam Sheet Composites. *Universal Journal of Materials Science, 2(6), 119-124.*

Kenechi, N. O., Linus, C., & Kayode, A. (2016). Utilization of rice husk as reinforcement in plastic composites fabrication-A review. *American Journal of Materials Synthesis and Processing, 1(3), 32-36.*

Kumar, A., Kumar, S., Kumar, D., Mahakur, V. K., & Bhowmik, S. (2024). Utilization of Banana Peel as Bio-Filler to Develop Bio-Composite Materials and Characterize Their Physical, Mechanical, Thermal, and Tribological Characteristics. *Polymer Bulletin, 81(17), 16175-16196.*

Kumar, P., & Kumar, M. (2022). Natural Nano-Fillers Materials for the Bio-Composites: A Review. *Journal of the Indian Chemical Society, 99(10), 100715.*

Kumar, J., Maurya, A. K., Gupta, H. S., Singh, S. P., & Sharma, C. (2022). Development of Eco-Friendly Bio-Composite by Reinforcing Pineapple Fruit Waste Crown Fiber to Ethylene-Propylene Rubber Modified Polyethylene. *Polymer Composites, 43(11), 8259-8273.*

Lee, M., Kim, Y., Ryu, H., Baeck, S-H., & Shim S.E. (2017). Effects of Silane Coupling Agent on the Mechanical and Thermal Properties of Silica/Polypropylene Composites. *The Polymer Society of Korea, Vol. 41, No. 4, pp. 599-609 (2017).*

Majewski, Ł., & Gaspar-Cunha, A. (2018). Evaluation of Suitability of Wheat Bran as a Natural Filler in Polymer Processing. *BioRes. 13(3), 7037-7052.*

Metin, D., Tıhmınlıoğlu, F., Balköse, D., & Ülkü, S. (2003). The Effect of Interfacial Interactions on The Mechanical Properties of Polypropylene/Natural Zeolite Composites. *Composites: Part A 35 (2004) 23–32.*

Mistry, B. (2015). Properties and Industrial Applications of Rice Husk. *SOCET Journal of Engineering and Technology, 4, 7-11.*

Mohammed, A. A., Hasan, Z., Omran, A. A. B., Kumar, V. V., Elfaghi, A. M., Ilyas, R. A., & Sapuan, S. M. (2022). Corn: its structure, polymer, fiber, composite, properties, and applications. *Polymers, 14(20), 4396.*

Morales, M. A., Porras, J., Maranon, A., Casas, J. P., Hernandez, C., Michaud, V., & Porras, A. (2025). Effect of Polypropylene-Grafted-Maleic Anhydride and Rice Husk Particle Size on 3D-Printing Filament Composites from Recycled Polypropylene. *Polymer Composites.*

Nurhayati, C., & Susanto, T. (2020, September). The Effects of Rice Husk Ashes Filled Recycled Polyethylene Composites towards Physical, Mechanical Properties and Its Degradation Characteristics. *In IOP Conference Series: Materials Science and Engineering (Vol. 924, No. 1, p. 012029). IOP Publishing.*

Petchwattana, N., Covavisaruch, S., & Chanakul, S. (2012). Mechanical Properties, Thermal Degradation and Natural Weathering of High-Density Polyethylene/Rice Hull Composites Compatibilized with Maleic Anhydride Grafted Polyethylene. *Journal of Polymer Research, 19(7), 9921.*

Salih, S. E., Hashim, B., & Al-Tamimi, H. M. (2014). The Mechanical and Thermal Properties of Polymer Hybrid Composite Reinforced by Rice Husk Ash at Different Conditions. *Eng. & Tech. Journal, 32(9), 2119-1132.*

Shen, Y. D., Deng, X. L., Feng, H. P., Gong, R. Q., Ren, L., Wang, Y. B., ... & Zhang, M. Y. (2024). Crystallization Behaviors Regulations and Thermostability Enhancement Strategy of Polypropylene Composites Modified by Dispersed Rice Husk Charcoal. *Journal of Polymer Research, 31(7), 190.*

Tanış, N. A., Okur, O., Duran, O., Yılmaz, E., Türkmen, M., Ariff, A. H. M., ... & Çalın, R. (2021). An Investigation of Production and Properties of RHA Reinforced Hybrid Composites by Vacuum Infiltration Method. *International Journal of Engineering Research and Development, 13(2), 704-710.*

Duy Tran, T., Dang Nguyen, M., Thuc, C. H., Thuc, H. H., & Dang Tan, T. (2013). Study of Mechanical Properties of Composite Material Based on Polypropylene and Vietnamese Rice Husk Filler. *Journal of Chemistry, 2013(1), 752924.*

Fuad, M. Y., Ismail, Z., Mansor, M. S., Ishak, Z. A., & Omar, A. K. (1995). Mechanical Properties of Rice Husk Ash/Polypropylene Composites. *Polymer journal, 27(10), 1002-1015.*

Raghu, N., Kale, A., Chauhan, S., & Aggarwal, P. K. (2018). Rice Husk Reinforced Polypropylene Composites: Mechanical, Morphological and Thermal Properties. *Journal of the Indian Academy of Wood Science, 15(1), 96-104.*

Rahman, M. R., Islam, M. N., Huque, M. M., Hamdan, S., & Ahmed, A. S. (2010). Effect of Chemical Treatment on Rice Husk (RH) Reinforced Polyethylene (PE) Composites. *BioResources, 5(2), 854-869.*

Premalal, H. G., Ismail, H., & Baharin, A. J. P. T. (2002). Comparison of the Mechanical Properties Of Rice Husk Powder Filled Polypropylene Composites with Talc Filled Polypropylene Composites. *Polymer Testing, 21(7), 833-839.*

Yang, H. S., Kim, H. J., Son, J., Park, H. J., Lee, B. J., & Hwang, T. S. (2004). Rice-Husk Flour Filled Polypropylene Composites; Mechanical and Morphological Study. *Composite structures, 63(3-4), 305-312.*

Zhang, Z., Sun, W., Huang, W., Zhu, Y., Li, J., Liang, C., & Gao, S. (2025). Interfacial and Mechanical Properties of Silane Coupling Agent Interface-Modified Basalt Fiber Reinforced Thermoplastic Polypropylene Resin Composites. *Polymer Composites, 46(7), 6582-6593.*