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Article

The Facile Synthesis and Application of Mesoporous Silica Nanoparticles with a Vinyl Functional Group for Plastic Recycling

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Abstract: Due to growing concerns about environmental pollution from plastic waste, plastic recycling research is gaining momentum. Traditional methods, such as incorporating inorganic particles, increasing cross-linking density with peroxides, and blending with silicone monomers, often improve mechanical properties but reduce flexibility for specific performance requirements. This study focuses on synthesizing silica nanoparticles with vinyl functional groups and evaluating their mechanical performance when used in recycled plastics. Silica precursors, namely sodium silicate, and vinyltrimethoxysilane (VTMS), combined with a surfactant, were employed to create pores, increasing silica's surface area. Early-stage introduction of vinyl functional groups prevented the typical post-synthesis reduction in surface area. Porous silica was produced in varying quantities of VTMS, and the synthesized porous silica nanomaterials were incorporated into recycled Polyethylene to induce cross-linking. Despite a decrease in surface area with increasing VTMS content, a significant surface area of 883 m²/g was achieved. In conclusion, porous silica with the right amount of vinyl content exhibited improved mechanical performance, including increased tensile strength, compared to conventional porous silica. This study shows that synthesized porous silica with integrated vinyl functional groups effectively enhances the performance of recycled plastics.

Keywords: plastic recycling; mesoporous silica nanoparticles; vinyl functional groups; sodium silicate; vinyltrimethoxysilane

1. Introduction

The widespread use of plastic products worldwide has naturally led to the emergence of environmental pollution issues related to plastic waste [1–3]. Typically, after the use of plastic products, they are disposed of through landfilling or incineration, resulting in associated soil and air pollution, along with high disposal costs [4,5]. To mitigate these challenges, active research is being conducted on plastic waste recycling. However, currently, only about 9% of plastic waste produced globally is recycled, while the remaining 91% goes unrecycled [6]. Polyolefin, a thermoplastic resin, is a commonly used materials as a raw material for plastics and is extensively employed in various facets of daily life [7–10]. Polyolefin-based polymer refers to chain or ring-like hydrocarbon polymers characterized by one or more unsaturated groups. Notable examples of polyolefins include polyethylene (PE), polypropylene (PP), polyisobutene, and polybutene. These versatile resins are widely employed in packaging materials, tableware, films, and numerous other applications [11–14]. Numerous studies have been conducted to facilitate the recycling of polyolefin-based thermoplastic materials. Among these, notable approaches include the introduction of inorganic nanoparticles [15–18], radical-induced cross-linking reactions [19,20], and the utilization of silane monomers for cross-linking [21,22]. Regarding the incorporation of inorganic nanoparticles, extensive research has been devoted to enhancing the mechanical properties of polyolefins by integrating nano-silica particles, with a particular emphasis on mesoporous silica [23–25]. It is generally observed that silica with a

higher surface area leads to improved performance when introduced into waste plastics. However, commercially available silica often lacks a significantly high surface area, and the synthesis of mesoporous silica with a surface area exceeding 1000 m²/g can be challenging due to complex production methods and the associated high costs. Furthermore, there are limitations in achieving performance improvements solely through the physical dispersion of silica nanoparticles, necessitating further research in this domain introduction of peroxides into recycled plastics. Upon exposure to a specific temperature threshold, peroxides generate oxygen radicals that react with unreacted unsaturated groups, thereby increasing cross-link density. While this can enhance physical properties, the presence of residual unreacted peroxides may lead to adverse effects [26], making it a less desirable option. The use of silane monomers in cross-linking reactions involving unsaturated groups introduces silane monomers processing unsaturation groups into recycled plastics. These unsaturated groups of silanes react with the unsaturated group of the plastics, increasing the cross-linking density and receiving the inorganic properties of the silane. However, silane monomers are typically in liquid form, which can impose limitations during the mixing process. Moreover, relying solely on the introduction of silane monomers may have inherent constraints, necessitating further research efforts. In other words, there is a need for research aimed at increasing the concentration of inorganic components to enhance tensile strength while simultaneously addressing the compromise in flexibility performance resulting from the hindered crosslinking density due to the incorporation of inorganic additives.

In this research, we aim to enhance the physical properties of recycled plastics by simultaneously introducing vinyl functional groups during the synthesis of silica nanoparticles using our room-temperature synthesis method [27], thereby increasing the crosslinking density. This is because the process of introducing vinyl functional groups after synthesizing silica nanoparticles can reduce the specific surface area and complicate the procedure. To synthesize silica nanoparticles with exceptionally high surface area, we propose the synthesis of mesoporous silica nanoparticles using sodium silicate as a precursor while simultaneously introducing vinyl functional groups. This approach not only reduces material costs but also enhances process convenience compared to post-introducing vinyl functional groups into the nanoparticles. Ultimately, it allows for the cost-effective synthesis of mesoporous silica nanoparticles that exhibit excellent performance when incorporated into recycled plastics.

2. Materials and Methods

2.1. Vinyl Functionalized Mesoporous Silica Nanoparticles (V-MS)

In the first beaker, a silica precursor solution was prepared by mixing 600 mL of distilled water with sodium silicate solution. In the second beaker, a solution consisting of equal volumes of distilled water and ethanol (100 mL total) was prepared, and its pH was adjusted to 3 using hydrochloric acid (HCl). Subsequently, Vinyltrimethoxysilane (VTMS) was added dropwise to the second beaker, and the hydrolysis reaction was induced over a period of 3 hours. Upon completion of the hydrolysis reaction, the hydrolyzed VTMS solution in the second beaker, which had a pH above 12 due to the presence of sodium silicate, was added dropwise to the first beaker where the sodium silicate solution had been diluted. The pH was adjusted to 10 using additional hydrochloric acid, and then 20 mL of Cetyltrimethylammonium chloride (CTACl) was added. The mixture was stirred at room temperature for 24 hours. After the stirring was completed, the solution was centrifuged to obtain a residue as a white slurry. To remove CTACl, this slurry was dispersed in a 1.0 M solution of HCl/Ethanol (EtOH) (1 L) and stirred for 24 hours. After 24 hours, another centrifugation was performed to collect a white slurry. This slurry was washed twice by dispersing it in a solution consisting of H₂O/EtOH in a 1:1 volume ratio. Finally, the obtained slurry was dried in a convection oven at 80° for 10 hours to yield white powder.

2.2. Characterization of Vinyl Functionalized Mesoporous Silica Nanoparticles

The morphology and particle shape of the synthesized mesoporous silica were examined using a Field Emission Transmission Electron Microscope (FE-TEM; HF-3300) under 300 kV conditions. Additionally, the specific surface area and porosity characteristics of the porous silica were analyzed using Nitrogen Adsorption-Desorption Isotherm (N_2 -sorption; QUANTACHROME, Qudrasorb SI). The measurements were conducted at a temperature of 77 K, maintained using liquid nitrogen. The adsorbed nitrogen was normalized to standard temperature and pressure. Prior to analysis, a heat treatment was performed at 200 °C for 6 hours to remove moisture and impurities adsorbed on the sample's surface. The Brunauer–Emmett–Teller (BET) specific surface area was calculated from the linear portion ($P/P_0 = 0.05\text{--}0.30$) of the BET equation. The volume and size of pores were calculated using the Barrett–Joyner–Halenda (BJH) equation. For properties confirmation of the mesopores, X-ray diffractometer (XRD; PANalytical X'pert PRO MRD) was employed. Measurements were conducted in 2θ scan mode with Cu-K α rays ($\lambda = 0.0154$ nm). Finally, to confirm the introduction of vinyl functional groups, Fourier Transform Infrared Spectrometry (FT-IR; Thermo, Nicolet iS50) was used for analysis.

2.3. Manufacturing Method of Polyethylene Compound

Polyethylene compounds were manufactured using a Twin Extruder (TEK20-L/D48(9B)-2SF-1V-AC2.2, SM Platek Co., LTD., Korea). Recycled polyethylene (Kwangduk Industry Co., KD-rePE_030A) was fed into the main feeder, along with silica with incorporated vinyl functional groups and carbon black N330 (OCI, Korea). TINUVIN 770 (BASF, USA) was utilized as an amine light stabilizer, UV-326 (Rianlon Corporation, China) as a UV stabilizer, LOXOL 8314 (Emery Oleochemicals, USA) as both a lubricant and antistatic agent, and Zn-st (Duksan General Science, Korea) as a lubricant. All of these were introduced through the side feeder as additives. During this process, the internal temperature of the extruder was maintained at 150–160°C. Subsequently, the material was cooled and formed into pellet shapes. Afterward, the cross-linking reaction was carried out at 90°C for 30 minutes using dicumyl peroxide ($[C_6H_5C(CH_3)_2]_2O_2$, KWANGDUK Industry Co.), resulting in the final product of recycled PE with incorporated silica.

2.4. Characterization of Polyethylene Compounds

To evaluate the physical performance of recycled PE, analyses were conducted using the Melting Index test (QM280, Qmesys, Korea) and Universal Testing Machine (UTM, QM100s, Qmesys, Korea). The Melting Index was carried out in accordance with ASTM D123 standards. Using the UTM, various tests were conducted, including tensile strength, elongation, elastic modulus, flexural strength, and flexural modulus. Tensile strength, elongation, and elastic modulus were assessed by preparing specimens according to ASTM D638 standards. The tests were performed with a testing speed of 50 mm/min, a grip-to-grip distance of 115 mm, a gauge length of 50 mm, and a load cell capacity of 30,000 N. Flexural strength and flexural modulus were evaluated based on ASTM D790 standards. Specimens were prepared, and the tests were conducted with a testing speed of 2.8 mm/min, a grip-to-grip distance of 104 mm, and an elastic range of 0.05% to 0.25%.

3. Results and Discussion

Vinyl functional groups were introduced into porous silica by adjusting the volume ratio of sodium silicate to VTMS from 40:0 to 25:15, as shown in Table 1, and TEM images of the synthesized mesoporous silica are presented in Figure 1. When the vinyl group content exceeded to 20 mL, gelation occurred and the reaction did not proceed. The TEM images confirm the successful formation of well-defined meso-sized pores in the vinyl functionalized MS nanoparticles (V-MS). Based on the TEM images, it can be observed that the meso-sized pores within the particles are partially arranged in a hexagonal configuration. However, it becomes increasingly difficult to identify a regular pore arrangement as the vinyl silane content increases.

Table 1. Volume ratio of sodium silicate to VTMS in the synthesis of vinyl functionalized mesoporous silica (V-MS).

	Sodium Silicate (mL)	VTMS (mL)
V00-MS	40	0
V05-MS	35	5
V10-MS	30	10
V15-MS	25	15

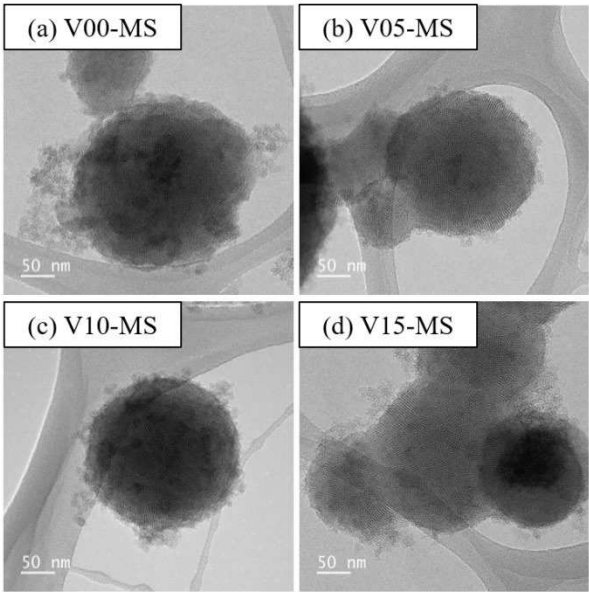


Figure 1. TEM images of V-MS nanoparticles.

The low angle XRD analysis data of the V- MS nanoparticles are shown in Figure 2. In the XRD result of V00-MS, which does not contain a vinyl functional group, a main peak corresponding to (100) is observed at around 2.06 in 2-theta. The small and broad peak approximately 3.91 in 2-theta, corresponding to (110), is also observed, confirming the hexagonal arrangement of the pore [28]. However, the incorporation of vinyl silane during sample preparation renders the (110) peak undetectable in the XRD data, clearly indicating a significant reduction in the hexagonal pore arrangement of the MS nanoparticles. Additionally, it was observed that the main peak associated with (100) undergoes a noticeable rightward shift as the content of vinyl silane increases. It can be inferred a reduction in the pore size of mesoporous silica nanoparticles. The V15-MS sample, with the highest vinyl group content, shows broader peaks of XRD analysis compared to other samples, indicating an inconsistency in pore size. This can be attributed to the fact that the vinyl silane only possesses three alkoxysilane groups, leading to uneven silane polymerization when compared to other samples.

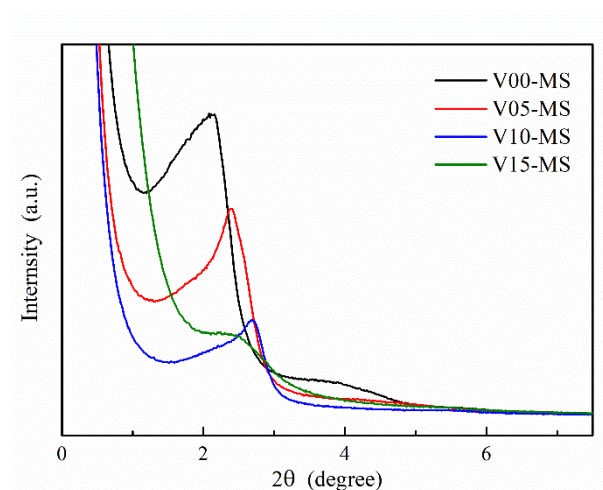


Figure 2. Low angle XRD analysis of vinyl functionalized mesoporous silica nanoparticles.

The N₂ adsorption-desorption isotherm graphs for silica samples with incorporated vinyl functional groups are presented in Figure 3. It is evident that all four types of mesoporous silica nanoparticles exhibit a Type IV hysteresis loop pattern [29]. Furthermore, the presence of separated adsorption and desorption curves suggests the existence of pores within the silica. The detailed pore properties of the MS are summarized in Table 2.

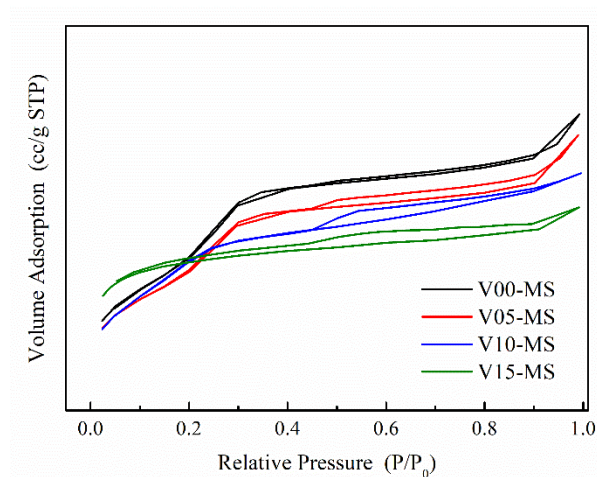


Figure 3. N₂-adsorption desorption isotherm graphs of V-MS nanoparticles.

Table 2. The textural properties of V-MS nanoparticles.

	BET (m ² /g)	Pore Size (nm)	Pore volume (cc/g)
V00-MS	1263	3.826	0.387
V05-MS	1139	3.486	0.463
V10-MS	1012	3.168	0.513
V15-MS	883	3.817	0.626

As the amount of VTMS in the composite increases, it can be observed that the specific surface area decreases while the pore volume increases. Since VTMS possesses a three-functional alkoxy group in contrast to the sodium silicate, the cross-linking density between the silanes is lower than that of sodium silicate. It leads to the distribution of relatively large pores within the mesoporous silica nanoparticle. As a result, the increase in pore volume can be attributed to the decrease in specific surface area. While there doesn't seem to be a substantial difference in pore size, it can also be inferred

that the presence of significant internal pores is suggested by the separation phenomenon between the adsorption and desorption curves, even at higher relative pressures (P/P_0).

The FT-IR analysis results of mesoporous silica nanoparticles incorporating vinyl groups (V-MS) are shown in Figure 4. Symmetric and asymmetric stretching vibrations of Si-O-Si were observed at 1057 cm^{-1} and 1215 cm^{-1} , respectively [27]. For the signal of vinyl group, a peak corresponding to the C=C double bond was detected at 1596 cm^{-1} (dash line in Figure 4), and C-H peaks associated with the C=C double bond were observed at 3097 cm^{-1} and 3181 cm^{-1} (dotted line in Figure 4) [30]. In V00-MS, there is no observable peak associated with vinyl groups. It can be noted that as the VTMS content increases, the peaks related to vinyl groups also increase. Nevertheless, when comparing V10-MS and V15-MS, the peaks associated with vinyl groups are nearly identical, suggesting that there may be a limit to the extent of vinyl group incorporation. These results indicate that vinyl groups have been successfully introduced into mesoporous silica.

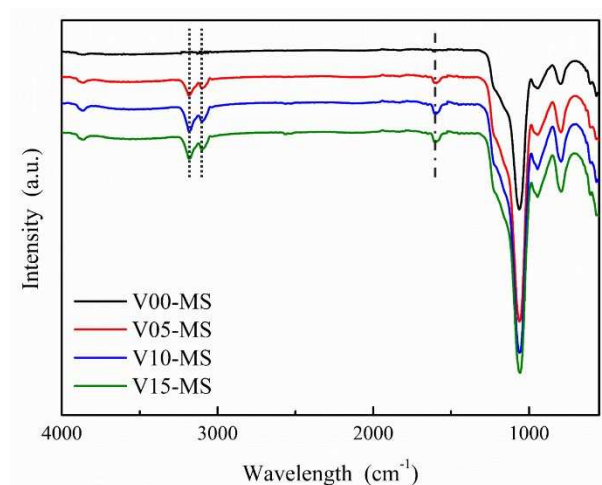


Figure 4. FT-IR graphs of V-MS.

Based on the analysis results of the MS samples with vinyl groups, we proceeded to manufacture pellets by incorporating them into recycled PE, followed by characterizing their properties. To determine the optimal MS nanoparticles with vinyl group content, we conducted a series of tests using V05-MS samples at various concentrations, and the results are presented in Table 3.

Table 3. Characterization of recycled polyethylene compounds with various V05-MS NPs contents.

Sample	Melt Index (g/10min)	Tensile Strength (kg/cm ²)	Elongation (%)	Elastic Modulus (kg/mm ²)	Flexural Strength (kg/cm ²)	Flexural Modulus (kg/mm ²)
STD PE_A						
rePE_A 100phr	2.6	97.1	113.6	15.3	48.5	14.0
V05-MS 0 phr	±0.5	±9.7	±11.3	±1.5	±4.8	±1.4
V05-MS_PE_A #1						
rePE_A 100phr	2.3	100.5	101.8	15.7	50.1	13.9
V05-MS 1 phr	±0.5	±10.0	±10.1	±1.5	±5.0	±1.3
V05-MS_PE_A #2						
rePE_A 100 phr	1.3	113.1	84.1	9.7	65.8	7.2
V05-MS 2 phr	±0.5	±11.3	±8.4	±0.9	±6.5	±0.7
V05-MS_PE_A #3						
rePE_A 100 phr	0.6	124.9	70.2	5.4	77.4	3.3
V05-MS 3 phr	±0.5	±12.4	±7.0	±0.5	±7.7	±0.3

** "phr" means Part per Hundred Rubber.

The results show that with increasing V05-MS NPs content, both tensile and flexural strengths improve compared to virgin recycled polyethylene (rePE). However, a decrease in elongation, elastic modulus, and flexural modulus was observed. The most suitable composition was found to be 1 part of V05-MS NPs per 100 parts of recycled PE, demonstrating the most superior characteristics. This can be explained by the fact that as the content of mesoporous silica with introduced vinyl functional groups increases, there are challenges with dispersion during the production of recycled PE, leading to a decrease in performance. This is because that as the specific surface area of mesoporous silica increases, particularly due to its high surface energy, it induces aggregation. [31] Therefore, while keeping the content of V-MS fixed at 1 part, the experiments were conducted with the varying content of the vinyl functional group, and the results are presented in Table 4.

Table 4. Characterization of recycled polyethylene compounds according to the content of the vinyl functional group introduced into mesoporous silica.

Sample	Melt Index (g/10min)	Tensile Strength (kg/cm ²)	Elongation (%)	Elastic Modulus (kg/mm ²)	Flexural Strength (kg/cm ²)	Flexural Modulus (kg/mm ²)
STD PE_A						
rePE_A 100phr	2.6	97.1	113.6	15.3	48.5	14.0
V05-MS0 phr	±0.5	±9.7	±11.3	±1.5	±4.8	±1.4
V00-MS_PE_A						
rePE_A 100phr	2.5	101.1	90.6	14.4	41.8	10.6
V00-MS1 phr	±0.5	±8.6	±8.0	±0.6	±4.1	±0.8
V05-MS_PE_A						
rePE_A 100phr	2.3	100.5	101.8	15.7	50.1	13.9
V05-MS1 phr	±0.5	±10.0	±10.1	±1.5	±5.0	±1.3
V10-MS_PE_A						
rePE_A 100phr	2.4	109.5	121.4	20.5	65.6	16.0
V10-MS1 phr	±0.5	±10.2	±12.1	±2.0	±6.5	±1.6
V15-MS_PE_A						
rePE_A 100phr	2.3	102.1	103.3	17.7	60.2	11.0
V15-MS1 phr	±0.5	±10.2	±10.3	±1.7	±6.0	±1.1

** "phr" means Part per Hundred Rubber.

When various amounts of vinyl functional groups were introduced into mesoporous silica and mixed into recycled PE, V10-MS showed the best properties. As the vinyl content increases, there is a presumed enhancement in the physical properties of recycled PE, attributed to the escalating cross-linking density among vinyl functional groups. However, in the case of V15-MS, which introduced a similar amount of vinyl functional groups based on FT-IR measurements, the physical properties improved compared to the pure recycled PE sample. In contrast, when compared to V10-MS, the physical properties actually decreased. This is attributed to the fact that, even with a similar amount content of the vinyl functional groups, the specific surface area of silica mesoporous particles, due to their inherent porosity, is reduced, resulting in a slight reduction in the physical properties of recycled PE. The introduction of inorganic silica nanoparticles typically results in an increase in tensile strength due to the inherent performance of these inorganic particles, even in the absence of direct bonding with recycled PE. This often leads to a reduction in the flexibility or properties requiring flexibility, as is commonly observed. [32] This can be observed in the performance indicators of recycled PE produced with VT00-MS, as evidenced in Table 4. However, in the case of introducing silica with incorporated vinyl functional groups into recycled PE, it can be observed that performance indicators requiring flexibility also increase. This phenomenon is attributed to the silica nanoparticles becoming homogeneously cross-linked through chemical bonding rather than physical dispersion, resulting in an increase in cross-linking density and a more homogeneous composite

structure. [33] In conclusion, it can be stated that the introduction of mesoporous silica nanoparticles into recycled PE results in superior performance, with higher specific surface areas and higher vinyl group contents proving to be advantageous.

4. Conclusions

In this study, we synthesized mesoporous silica nanoparticles with incorporated vinyl functional groups using a hydrolysis process with sodium silicate solution and VTMS, without the need for additional polymerization or sintering processes. Previous research typically involved introducing vinyl functional groups after the synthesis of porous silica nanoparticles, but this approach often resulted in a significant decrease in specific surface area after vinyl group incorporation. However, with our synthesis method, we were able to produce mesoporous silica with high specific surface areas and incorporated vinyl functional groups through a straightforward process. This suggests the potential for practical application beyond the scope of this study.

We incorporated the synthesized mesoporous silica nanoparticles with vinyl functional groups into the production of recycled PE and compared its physical properties. The results showed that as the specific surface area and vinyl group content increased, all physical properties improved. Conventional introduction of silica nanoparticles into recycled PE generally led to an increase in tensile strength while reducing other performance indicators that require flexibility. However, with the incorporation of vinyl functional groups, it became possible to induce chemical bonding in the inorganic silica nanoparticles, leading to an increase in cross-linking density. As a result, not only tensile strength but also other flexibility-related performance indicators improved.

Through this study, our research team has confirmed the potential performance benefits of mesoporous silica nanoparticles with incorporated vinyl functional groups. We aim to further investigate its application in various recycled plastics and rubber materials.

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Conflicts of Interest: The authors declare no conflict of interest.

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