

Exploratory Study on Seaweed as Novel Filler in Polypropylene Composite

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ABSTRACT: Seaweed (SW) is employed as filler to prepare composites on the basis of a polypropylene (PP) matrix in the ratio of 10 : 90, 20 : 80, 30 : 70, 40 : 60, and 50 : 50 (wt % SW : wt % PP) by compounding and injection molding. The tensile, bending and impact properties of the composites were investigated. The 30% SW : 70% PP composite showed the best over-all mechanical performance of the composites prepared. Further improvement of this optimal composite was achieved by incorporating 2% thermoplastic elastomer (TPE) as

additive. Interfacial adhesion and bonding between the fibers and PP matrix were investigated by scanning electron microscopy (SEM). Water absorption tests of the different composites were also performed, and addition of TPE was found to lead to a substantial reduction of water uptake. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 109: 1242–1247, 2008

Key words: seaweed; injection molding; composite; mechanical properties; water uptake

INTRODUCTION

In recent decades, ecological concern has resulted in a renewed interest in natural materials and issues such as recyclability and environmental safety are becoming increasingly important for the introduction of new materials and products. Fillers are incorporated into rubber and plastics matrices mainly to achieve improvement of service properties (reinforcing fillers) or to reduce the material cost. In some applications, special types of filler are introduced to polymer matrices to achieve specific properties such as degradability, chemical resistance, and flame retardant performance.^{1–3} However, the performance of a filler is governed by its characteristics, such as particle size, surface activity, degree of interaction with the polymer matrix, chemical composition, and degree of irregularity of the shape of the filler.^{4,5} Therefore, depending on chemical treatment and processing conditions, different grades of fillers with different properties can be obtained from the same basic filler material source. There are a number of widely used commercial fillers, which have different grades with their own distinct characteristics. Carbon black and silica are the classical examples.

Apart from these types of conventional fillers, a considerable interest concerning the utilization of

fillers derived from agricultural and industrial waste has been generated recently. Conversion of such materials into fillers introduces low cost fillers and a solution to disposing waste from various industries. Fillers of both rubber and plastics derived from oil palm wood, wood flour, rice husk, etc., have been investigated by many researchers.^{6–9} The studies have revealed that these fillers enhance some mechanical properties of the composites at the expense of the deterioration of some other properties.

Lignocellulosic fibers have been used as fillers with nondegradable polymers, such as polypropylene,^{10–14} polyethylene,^{15–18} polyvinyl chloride, and polystyrene^{19–21} to reduce the cost of these commodity thermoplastics. Most of the prior works on lignocellulosic fibers in thermoplastics deal with the use of wood flour and native cellulose fibers, such as kenaf, flax, jute, coir, and sisal with additives that can improve dispersion and interaction with the polymeric matrix.^{22–25} The use of the lignocellulosic materials in these thermoplastics results in composites with higher elastic modulus. Their strength is usually lower due to the poor adhesion between the hydrophilic filler and the hydrophobic matrix. As a matter of fact, fibers can work as reinforcement or as filler, depending on the fiber/matrix interaction, on the aspect ratio and on the critical fiber length. A strong interface is necessary to obtain stress transfer from the matrix to the fibers and to avoid that the dispersed phase acts as weak spots in the material. Reinforcing effects can be improved, therefore, if the adhesion is promoted by the use of additives or by chemical modifications of the components.

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The main disadvantages of using lignocellulosic natural fibers in thermoplastics are the high moisture absorption of the fiber and the resulting composite, as well as the reduced processing temperature permissible. The moisture absorbed by the composite and the corresponding dimensional change can be reduced dramatically if the fibers are thoroughly encapsulated in the plastic and there is good adhesion between the fiber and the matrix.

As any other organic materials, polymers and natural fiber are very sensitive to fire/flame and hence improvement of fire retardancy of the composite materials has become more and more important to comply with safety requirements of the natural fiber/composite products. Seaweed is a pure natural material that offers numerous advantages like high heat insulation, heat capacity and, therefore, is an effective heat protection during summer. It has a good ability of humidity regulation, good processing properties, and excellent elasticity characteristics. Other qualities are its full recyclability and strong carbon dioxide fixation. It offers energy absorbing material properties because of fibrils that lead to outstanding insulation and sound-absorption characteristics. In addition, it works as a brilliant flame retardant. Therefore, polymer composite panels may be developed that can be used in automotive industry as interior trim, in construction industry as tiles for balcony balustrades or in furniture industry as desk slabs.

However, to the author's knowledge no detailed work has been carried out on the utilization of seaweed as filler for polymer composites which could be extensively used in many applications. In this work, composites were prepared by using seaweed as filler. The effect of processing and filler composition on mechanical performance and water uptake was investigated.

EXPERIMENTAL

Material

Polypropylene (PP) (Borealis RB 707 CF, MFI = 1,8 g/10 min) was used as the matrix and Urethane based thermoplastic elastomer (TPE) (Elastollane S 50A from Elastogran, Lemfoerle, Germany) was used as additive. The green seaweed (*Ulva* species) was collected from the Baltic sea and was dried at 80°C in a vacuum oven for 24 h before the preparation of the composites. The chemical composition of seaweed is presented in Table I.

Composite fabrication

Composites were produced by a two step process: compounding by extrusion followed by injection molding. The PP matrix polymer and the fillers were dry-mixed in different weight fractions (Table II).

TABLE I
Chemical Composition of the *Ulva*
Species Green Seaweed^{26,2,7}

Composition	Quantity grams/100 g
Ash	13–22
Total carbohydrates	42–46
Alginic acid	0
Xylans	0
Laminaran	0
Mannitol	0
Fucoidan	0
Floridoside	0
Protein	15–25
Fat	0.6–0.7
Potassium	0.7
Sodium	3.3

Compounds were prepared by passing the mixtures through a twin-screw extruder (Leistritz, Nuremberg, Germany). The processing temperature of extrusion was maintained at 180°C, and extrusion pressure was kept constant. The extruded strands of the compounds were cut into small pieces of 15–20 cm length, and were then crushed into small granules using a grinding mill (Herbold, Meckesheim, Germany). The granules were dried in an oven at 105°C for 24 h and were shaped into ASTM (ISO 527) standard specimen by use of an injection molding machine (Boy 30A from Dr. Boy, Neustadt-Fernthal, Germany) at a temperature of 175°C. A mold with a single cavity was used to produce the test specimen.

Determination of mechanical properties of the composites

To investigate the mechanical properties of the composites produced, mechanical test properties like tensile, bending, and impact tests were performed.

Tensile testing was done using dumbbell shaped injection molded specimen on a Zwick Z010 universal testing machine according to ASTM DIN EN ISO 527-1. Bending testing was also done on a Zwick 2201 machine following ASTM DIN EN ISO 178

TABLE II
Relative Amount (wt %) of Reinforcing Materials and
Matrix Polymer in Composites

Fillers as reinforcing materials (%)	Polymer matrix (%)	Composites
None	PP: 100	PP
Seaweed: 10	PP: 90	SW10
Seaweed: 20	PP: 80	SW20
Seaweed: 30	PP: 70	SW30
Sea Weed: 50	PP: 50	SW50
Seaweed: 28; Additive: 2	PP: 70	SWAdd

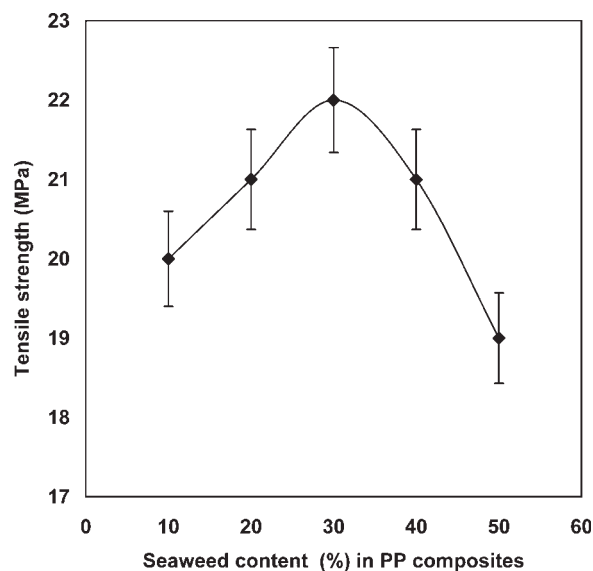


Figure 1 Tensile strength (MPa) of the composites.

methods. Impact strength of samples was measured on a Zwick 5102 machine according to ASTM DIN EN ISO 179. All tests were done at room temperature. At least five samples were tested for each composition and the results were averaged.

Scanning electron microscopy

To study the fracture surface of tensile specimens, scanning electron microscopy (SEM) micrographs were obtained on gold-coated fractured composite samples using scanning electron microscope. The coated samples were examined under a CAM SCAN 4 DV SEM at an operation voltage of up to 20 kV.

Water absorption test

Dumbbell shaped samples were used for measurements of water absorption. After being oven dried at 105°C for 24 h, the specimens were kept in the desiccators at room temperature. Then the specimens were weighed before being immersed in distilled water. The mass was recorded as the mass of samples before immersion. The specimens were periodically taken out of the water, surface dried with absorbent paper, reweighed, and immediately put back into the water. Water absorption was calculated according to ASTM D 5229/D 5229M-92.

RESULTS AND DISCUSSION

Mechanical Properties

Tensile strength and elongation at break

The tensile properties like tensile strength (TS) and elongation at break (Eb) of the composite containing 10, 20, 30, 40, 50% fillers were studied and the

results are presented in Figures 1 and 2, respectively. It is observed that with an increase of filler content from 10 to 30% the tensile strength gradually increases, but the tensile strength of the composites are found to decrease with increasing filler loading for weight fractions from 40 to 50%. This may be due to the lack of stress transfer from the PP matrix to the filler. The elongation at break of the composites shows a similar trend as shown by the tensile strength performance, and the maximum elongation at break is obtained for the 30% SW sample. An increase of the elongation at break of the composites increases the toughness and ductility of the composite.²⁸ The composites might show moderate adhesion between two polymeric surfaces via interdiffusion of the macromolecules of both polymers,²⁹ or physical interlocking between filler and matrix.

Bending strength

The bending strength of the different composites is shown in Figure 3. A similar enhancement of the bending strength of the composites as in the case of the tensile strength can be observed. The bending strength increases up to 30% seaweed content, and thereafter remains constant.

Impact strength

Considering the impact strength of the composites as presented in Figure 4, an increasing trend with increasing seaweed content from 10% to 30% is observed, followed by a decreasing trend. It is envisaged that as the size of filler would become smaller, greater interaction between filler and matrix could result in better and more efficient stress transfer

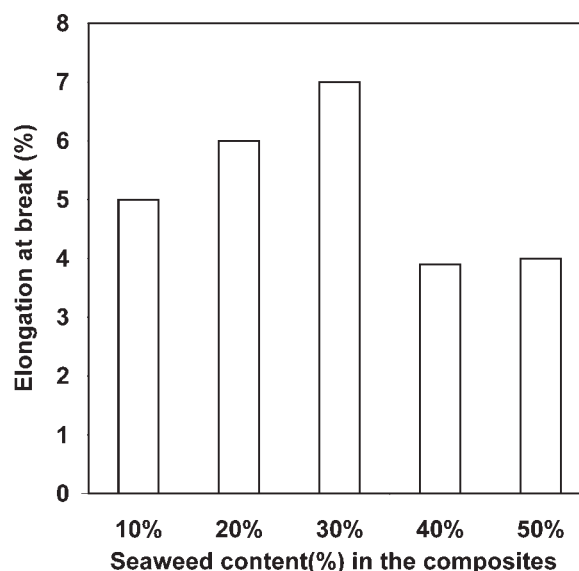


Figure 2 Elongation at break (%) of the composites.

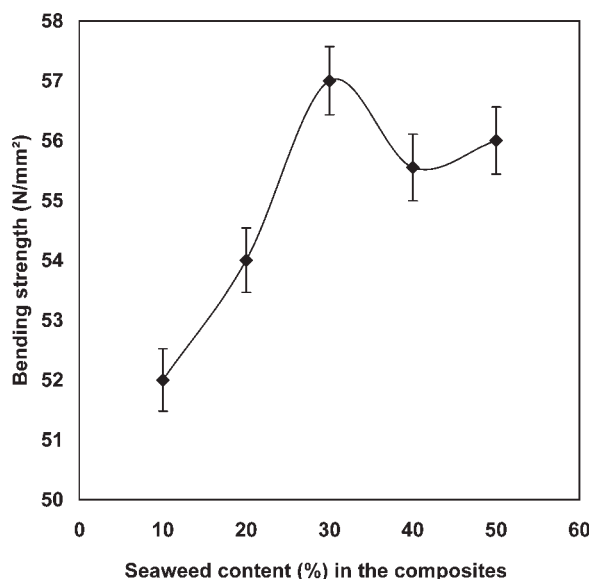


Figure 3 Bending strength of the composites.

which in turn could lead to an increase in impact strength of the composites.³⁰

From the studies on the effect of filler content on the mechanical performance of the composites, we find that the mechanical properties increase with increasing filler content up to an optimal value, and thereafter with further increase in filler content, the properties either remain constant or show a decreasing trend. The optimal filler content varies with the nature of filler and matrix, filler aspect ratio, filler/matrix interfacial adhesion, etc. The low value at high filler content may be due to the presence of so many filler ends in the composites, which could cause crack initiation and hence potential composite failure.^{31,32}

Tensile and bending modulus

Both tensile and bending modulus are increasing with increasing filler loading as compared to 100%

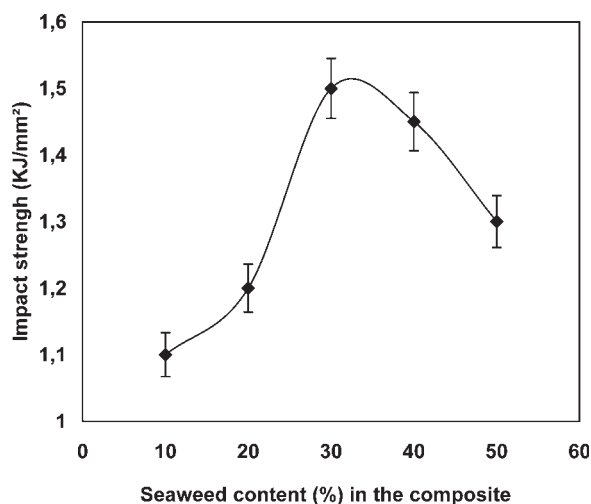


Figure 4 Impact strength of the composites.

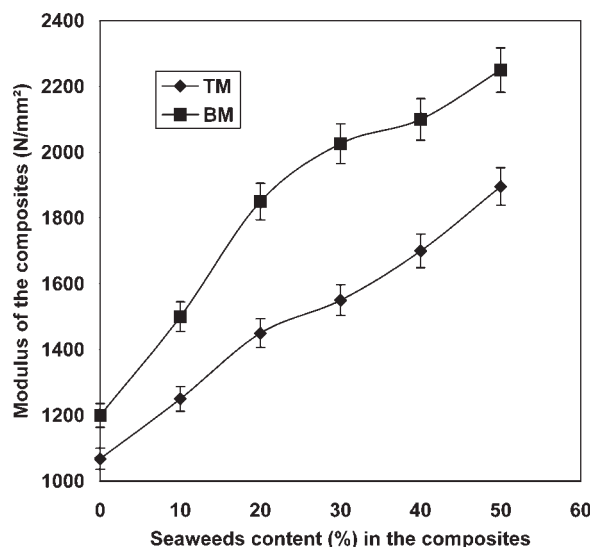


Figure 5 Tensile modulus (TM) and bending modulus (BM) of the composites.

PP (Fig. 5). While filler loading more than 30% SW has adverse effect on the tensile strength, at the same time, it has direct proportional effects on the tensile modulus and the bending modulus. Both effects are due to the high stiffness of the seaweed fibrils.

Effect of TPE as additive

The composite SW30PP prepared by 30% SW and 70% P showed the best mechanical performance, and this optimal system was used to test the effect of TPE as additive. To study the effect of TPE addition on the tensile properties of the composite, a small amount of 2% TPE was incorporated into the optimal formulation. The effect of the TPE additive on

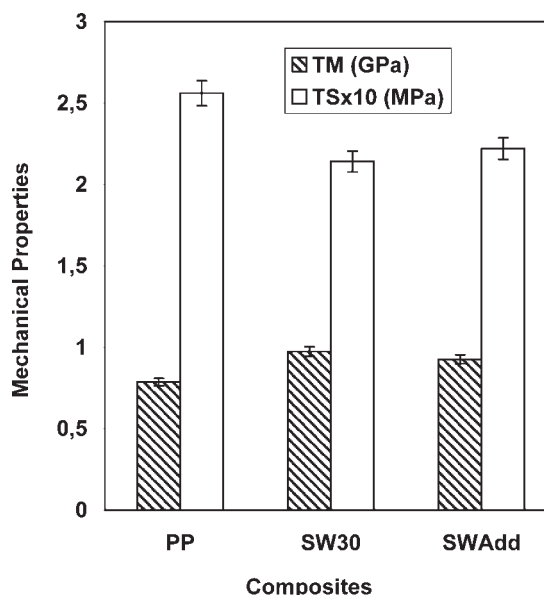


Figure 6 Effect of additive on tensile strength (TS) and tensile modulus (TM) of composites.

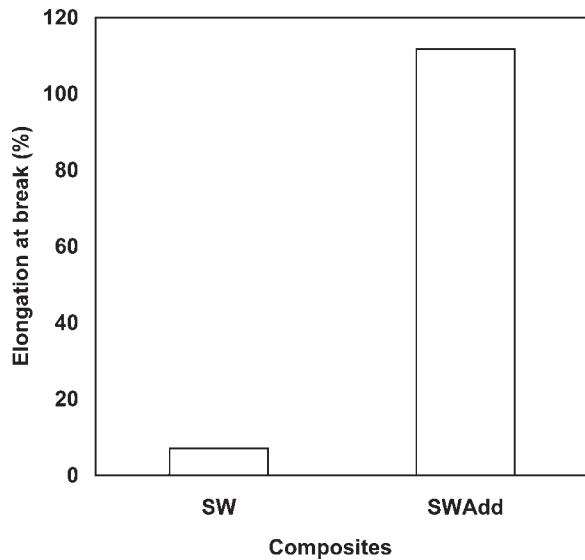


Figure 7 Effect of additive on elongation at break (%) of the composites.

tensile properties like tensile strength, tensile modulus and elongation at break are shown in Figures 6 and 7 respectively. A significant improvement in the mechanical performance was obtained for the TPE treated composite with an enhancement especially of the elongation at break.

TPE is a polymer which exhibits properties of thermoplastics at high temperature and can be molded like normal thermoplastics. Hence TPE can be molded with general processing technologies for thermoplastics such as extrusion as well as injection molding.^{34–36} If added to PP, it acts as lubricant in extrusion thereby reducing extrusion pressures. In our present work, TPE acts as lubricant as well as compatibilizer in the blend of seaweed and PP dur-

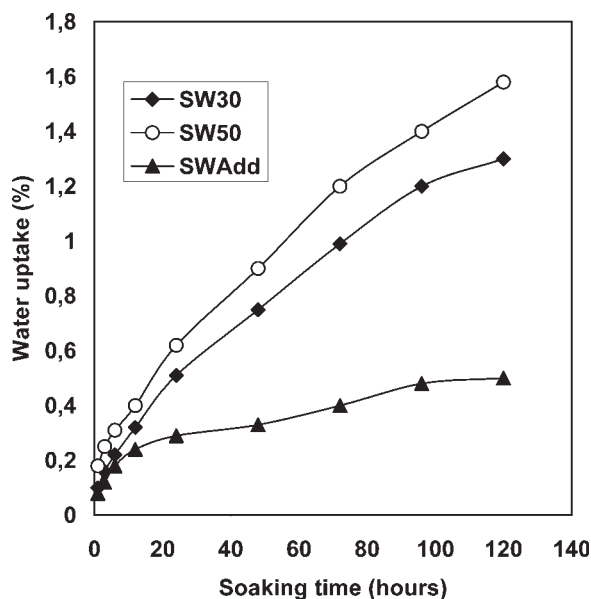


Figure 8 Water uptake of the composites.

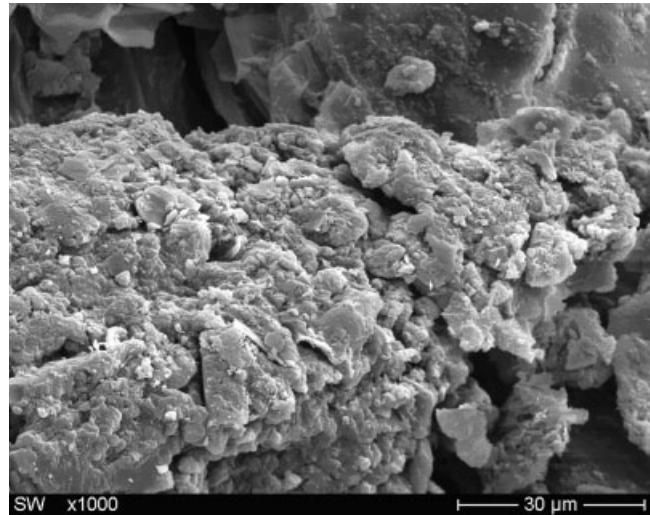


Figure 9 SEM micrograph of fractured specimen of untreated seaweed.

ing composite fabrication. More research is needed to clarify the role of TPE during compounding and injection molding of composite materials.

Water absorption test

One of the important characteristics of composite materials is water absorption capacity which determines the end use application of composites. The results of water uptake tests are presented in Figure 8 as water uptake versus soaking time. As expected, the water uptake increases with the filler loading of the composites. However, the lowest water uptake is shown by the sample treated with the TPE additive (SWAdd) indicating that the TPE additive plays an important role in reducing water uptake. The TPE acts as a compatibilizer between the PP matrix and the seaweed fibers, and during compounding the TPE will coat the sea-

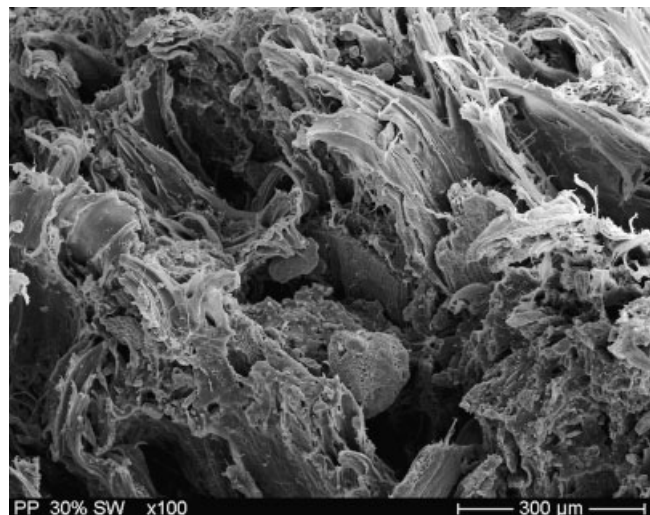


Figure 10 SEM micrograph of fractured specimen of seaweed PP composite SW30 without TPE additive.



Figure 11 SEM micrograph of fractured specimen of seaweed PP composite SWAdd with TPE additive.

weed, thereby sealing the seaweed effectively from water. It is obvious that water absorption increases as the immersion time is increased. This is expected because the longer the samples are exposed to water, the higher the water content absorbed in the cells of the seaweed fibrils, and the seaweed cells are swollen accordingly. Only for the composite with TPE additive, the equilibrium water uptake is observed within the maximum soaking time investigated.

Scanning electron microscopy

Scanning electron micrographs (SEM) are presented in Figures 9–11, showing the micrographs of neat seaweed, of the seaweed PP composite SW30 without TPE additive, and of seaweed PP composite SWAdd with TPE additive, respectively. It is observed that there are some voids around the seaweed platelets and some spaces where platelets have been pulled out, indicating that there exists a kind of interfacial adhesion and/or interlocking contact between seaweed and PP matrix as expected from the enhanced mechanical properties of the composites.

CONCLUSIONS

The results of this exploratory study demonstrate that the PP composite prepared with 30% of seaweed showed the optimal mechanical performance. Incorporation of a small amount of TPE in the composite formulation resulted in a new composite with enhanced tensile properties and lower water uptake. Interfacial adhesion and bonding between seaweed filler and PP matrix as investigated by scanning electron microscopy (SEM) was found to be limited, which suggests that surface treatment of the filler to achieve better compatibility between seaweed filler

and matrix PP might improve mechanical properties further. This will be the subject of future work.

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