

Coating of Jute with Natural Rubber

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ABSTRACT: Jute fabric was coated with natural rubber to develop double-texture rubberized waterproof fabric and fabric-reinforced rubber sheeting for hospitals. The vulcanization of such natural-rubber-coated flexible composites at 120°C for 3 h produced optimum effects. The jute/natural-rubber composite was much superior to a conventional polyester/natural-rubber composite for producing such double-texture rubberized fabric with respect to the fabric-to-natural-rubber adhesion, breaking strength, tear strength, abrasion resistance, puncture resistance, and biodegradability. For fabric-reinforced rubberized sheeting, the jute/natural-rubber composite was superior to a conventionally used cotton/natural-rubber composite with respect to the fabric-to-natural-rubber adhesion, breaking strength, tear strength, and abrasion resistance. However, for both appli-

cations, the jute-based products were commonly found to be less extensible, heavier, and thicker. Unsaturation in the lignin fraction of jute established a chemical linkage with the unsaturation of natural rubber via sulfur at the jute/natural-rubber interface. An examination of the surface morphology of uncoated and coated jute fabrics by scanning electron microscopy revealed a good degree of deposition and filling even in the intercellular regions of jute by a cohesive mass of natural rubber, which remained unseparated from the fiber, when mechanical force was applied. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 98: 484–489, 2005

Key words: adhesion; coatings; composites; electron microscopy

INTRODUCTION

Environmental preservation, pollution control, and emphasis on the use of energy-efficient materials and processing in the industrial sector have renewed interest in agrobased fibers, including jute, for use even in nontraditional areas. Polyester and cotton textiles coated with suitable elastomers and polymers are already in use in different areas of recently defined technical textiles.^{1,2} Related studies on a lignocellulosic fiber, such as jute, are few, however. Therefore, we thought it would be interesting to investigate the effect of natural rubber as a coating polymer on jute fabric. This article deals with the development of two types of jute-based coated products: double-texture (DT) rubberized fabric,³ which can be used in luggage and related items, school bags, and so forth, and jute-fabric-supported rubberized sheeting for use in hospitals (HS).

EXPERIMENTAL

Materials

Fabric

Plain-weave jute fabric with 46 ends/dm, 54 picks/dm, 207-tex warp, 207-tex weft, and an average area density of 260 g/m² and plain-weave polyester fabric with 128 ends/dm, 160 picks/dm, 40-tex warp, 42-tex weft, and an average area density of 125 g/m² were used to produce DT rubberized fabric.

Plain-weave jute fabric with 38 ends/dm, 41 picks/dm, 206-tex warp, 206-tex weft, and an average area density of 170 g/m² and plain-weave cotton fabric with 256 ends/dm, 220 picks/dm, 5-tex warp, 9-tex weft, and an average area density of 43 g/m² were used for rubberized hospital sheeting (HS).

Rubber and other chemicals

Grade ISNR V natural rubber was used. All other auxiliaries, including paraphenylene diamine, zinc oxide, stearic acid, mercaptobenzothiourea, sulfur, spindle oil, paraffin wax, whiting powder, china clay, and precipitated silica, were commercial-grade.

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Methods

Coating of fabric with natural rubber

The coating of one or both sides of different specified fabrics by a natural-rubber compound was done in a three-roll I-type calendar machine at $75 \pm 5^\circ\text{C}$. The coated fabrics were subsequently vulcanized in a steam-heated autoclave at 120°C for 3 h, unless otherwise specified. Natural-rubber compounding was done by the thorough mixing of natural rubber (100 phr), paraphenylene diamine as an antioxidant (1.5 phr), zinc oxide (5 phr), stearic acid (1 phr), mercapto-benzothiazuram (1.5 phr), sulfur (2 phr), paraffin wax (0.5 phr), china clay (30 phr), whiting powder (50 phr), silica (40 phr), and spindle oil (5 phr) in a laboratory-model two-roll mixing mill. The compound was then allowed to cool in water and was matured subsequently for 24 h. The matured compound was then passed through a cracking, refining, warming, and feeding mill before it was fed to the calendar machine for the coating operation. Fabric-reinforced rubberized HS was produced by the coating of both sides of specified fabrics with natural rubber, and DT rubberized fabric was made by the sandwiching of the rubber compound between two sets of base fabrics as specified in the calendar machine.

To study the role of only sulfur (in the absence of accelerator and activator systems) for the establishment of a chemical linkage between jute and natural rubber, one side of the jute fabric was coated with the procedure mentioned previously with a natural-rubber compound containing no thiuram, zinc oxide, or stearic acid. In the rubber compound, only 10 phr sulfur was used; the concentrations of the other processing aids were kept the same as specified previously, and the coated product was vulcanized at 150°C for 6 h. During the coating in the calendar machine, the gap between the rollers was adjusted to enable the deposition of equal weights of the natural-rubber compound and jute fabric (with respect to the unit area). This coated jute fabric, with a jute/natural-rubber weight ratio of 1 : 1, was analyzed only by infrared (IR) spectroscopy.

Determination of the weight of rubber

The coating mass per unit of area of different specified fabrics was determined with the method given in IS-7016 Part I.

Determination of the coating adhesion

The coating adhesion of different samples was determined with IS 7016 Part V with a Good Brand constant rate of traverse (CRT) testing machine.

Determination of the tensile properties

The tenacity and elongation at break of specified fabric samples were measured according to the method given in IS-7016 Part II. The results were based on an average of 10 tests for each sample.

Determination of the tear strength

The tear strength was measured with a tongue tear test as described in ASTM D 2261-64T.

Determination of the puncture resistance

The puncture resistance of different specified fabric samples was determined with the method specified in IS 4006-1979.

Determination of the abrasion resistance

The evaluation of the plane abrasion resistance (IS: 127673-1989) of selected fabric samples was performed with a Martindale abrasion tester (Martindale, Coimbatore, India) with standard emery paper (IS 715) as the abrader. The number of abrasion cycles required for the first appearance of a hole in the coated fabric was recorded. The results are the averages of 10 such tests performed for each sample.

IR spectroscopy

IR spectra of the vulcanized jute/natural-rubber composite (having equal weights of jute and natural rubber), unvulcanized natural rubber, and jute were obtained with a KBr pellet technique, as detailed elsewhere,^{4,5} with a Bomem 104 spectrometer (Quebec, Canada). The dried samples were crushed to a size finer than 20 mesh at -120°C , with a cryogenic grinder in the presence of liquid nitrogen, before being pelletized with KBr. Three KBr pellet test specimens (each 0.3 g) were prepared separately for the jute, unvulcanized natural rubber, and vulcanized jute/natural-rubber composite, which contained about 1, 1, and 2% (w/w) powdered samples, respectively.

Scanning electron microscopy

The surface morphology of the uncoated and coated jute, cotton, and polyester fibers (taken from coated fabrics) was studied with a Hitachi model S 340 scanning electron microscope (Tokyo, Japan). For the coated fabrics, fibers from the fabric-rubber interface were considered. Such fibers were obtained when natural rubber was separated from the substrates during the determination of the adhesion of natural rubber to the different substrates specified previously.

TABLE I
Properties of DT Rubberized Polyester and Jute Fabrics

Type of coated fabric	Area density (g/m ²)	Thickness (mm)	Weight of the rubber (g/m ²)	Breaking load (N/5 cm)	Extension at break (%)	Tearing strength (N)	Seam strength (N)	Abrasion resistance (number of cycles)	Puncture resistance (oz. in./tear in.)	Retention of TS after 21 days of soil incubation (%)
DT rubberized jute fabric	844	1.2	324	<i>W_p</i> 1070, <i>W_t</i> 1345	<i>W_p</i> 4.4, <i>W_t</i> 4.8	<i>W_p</i> 101, <i>W_t</i> 92	528	960	1120	95
Conventionally used DT polyester fabric	570	0.55	320	<i>W_p</i> 635, <i>W_t</i> 585	<i>W_p</i> 21.12, <i>W_t</i> 22	<i>W_p</i> 35.8, <i>W_t</i> 45	300	120	400	58

W_p and *W_t* indicate the property parameters shown in the warp and weft directions, respectively, of the fabrics.

RESULTS AND DISCUSSION

DT rubberized fabric

Table I shows property profiles of DT rubberized jute and polyester fabrics. Because of differences in the nature, surface characteristics, weight, and linear density of the yarns used and their spacing, the two DT fabrics show significant differences in their physical and mechanical properties. The DT jute fabric is much superior to the DT polyester fabric in terms of the breaking strength, puncture resistance, and bursting strength. The extensibility and fineness, however, of the DT jute fabric are less than those of the DT polyester fabric.

The rot resistance or resistance to microbial attack and degradation of the DT polyester and DT jute fabric samples, as studied by a standard soil burial test and expressed in terms of the tensile strength (TS) retained after a specified period of soil burial, are also given in Table I. The DT jute fabric is susceptible to microbial attack and degradation, unlike the DT rubberized polyester fabric, because natural rubber even in its vulcanized state supports the growth of microorganisms on account of its small amount of protein.¹

Fabric-reinforced rubberized HS

The properties of jute-reinforced HS and conventionally used cotton-reinforced HS are shown in Table II.

Because of the use of coarser jute and jute fabric of higher area density, the jute-reinforced rubberized HS is heavier and thicker than the cotton-reinforced HS. The breaking strength, tear strength, and abrasion resistance of the jute-reinforced HS are much higher than those of the conventional cotton-reinforced HS for close or comparable extensibility. The flexibility, as revealed by the bending length of jute-based HS, is less than that of cotton-based HS; however, the flexibility of the jute-based product is sufficient for its practical use. Improvements in the mechanical properties and a reduction in the flexibility for the jute-reinforced rubberized HS may be viewed as results of the incorporation of strong and stiff jute in the product.

Role of sulfur in the vulcanization of coated jute fabric

It would be useful to consider, at the outset, the mechanism of the modification of jute with natural rubber under the influence of sulfur as the vulcanizing agent in the absence of any accelerator system. Figure 1 shows the sequence of reactions expected to take place, ultimately giving rise to the strong adhesion of natural rubber to jute. It has already been reported⁶ that sulfur dissolves in rubber and exists in rubber as a stable S₈ ring. At an elevated temperature under the

TABLE II
Properties of Jute- and Cotton-Reinforced Rubberized Sheeting for Hospitals

Type of coated fabric	Area density (g/m ²)	Thickness (mm)	Weight of rubber (g/m ²)	Breaking load (N/5 cm)	Extension at break (%)	Tear strength (N)	Abrasion resistance (number of cycles)	Bending length (cm)
Jute-reinforced HS	1018	1.1	848	<i>W_p</i> 436, <i>W_t</i> 327	<i>W_p</i> 4.1, <i>W_t</i> 4.5	<i>W_p</i> 41.72, <i>W_t</i> *-46.2	880	3.5
Conventionally used cotton-reinforced HS	728	0.6	685	<i>W_p</i> 300, <i>W_t</i> 215	<i>W_p</i> 4.57, <i>W_t</i> 4.7	<i>W_p</i> 27.66, <i>W_t</i> 29	215	2.9

W_p and *W_t* indicate the property parameters shown in the warp and weft directions, respectively, of coated fabrics.

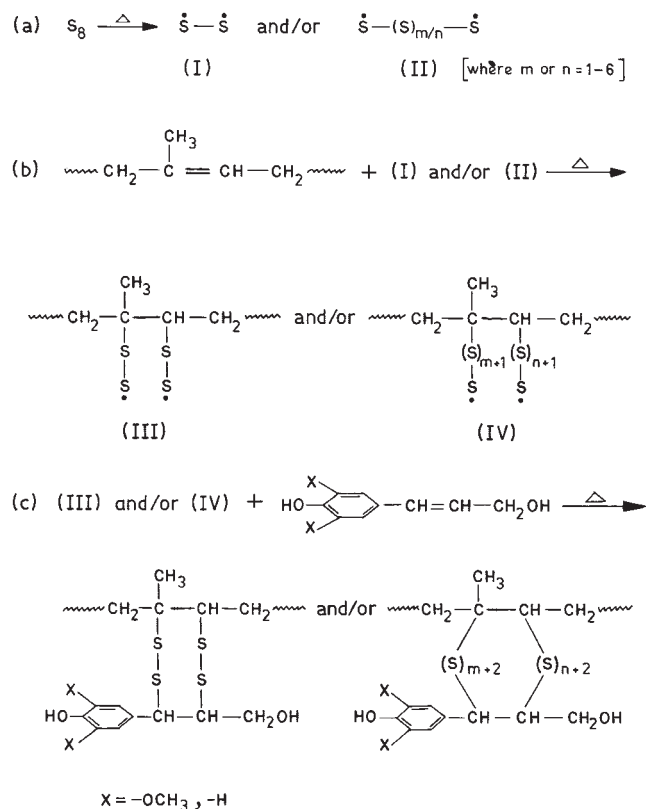


Figure 1 Possible reaction scheme showing the modification of jute with natural rubber in the presence of sulfur as the vulcanizing agent.

condition of vulcanization, this S_8 ring splits, producing sulfur biradicals, as shown in eq. (a). The biradicals thus formed have the distinct possibility of combining with the unsaturated carbon atoms of the polyisoprene chains in the manner shown in eq. (b) on the basis of the already established reaction shown to take place between sulfur and low-molecular-weight olefins.^{7,8} Pendant sulfur radicals duly grafted to the polyisoprene chain may then react with unsaturation present in the α position to the benzene ring of coniferyl, cumaryl, and sinapyl alcohols of the lignin fraction of jute, as shown in eq. (c). Possible products following eq. (c) are analogous to crosslinked structures of 2 mol of octene-1, which have been reported elsewhere to join each other via sulfur; this involves sulfur addition to their respective double bonds, which leads to a closed ring structure.⁹ Moreover, an additional reaction leading to the formation of a chemical linkage via sulfur involving methylene groups (adjacent to double bonds of polyisoprene chains of natural rubber and coniferyl, cumaryl, and sinapyl alcohols of lignin) can take place. The aforementioned crosslinking reactions ultimately lead to the establishment of a chemical linkage between rubber and jute involving multitudes of polyisoprene chains and lignin.

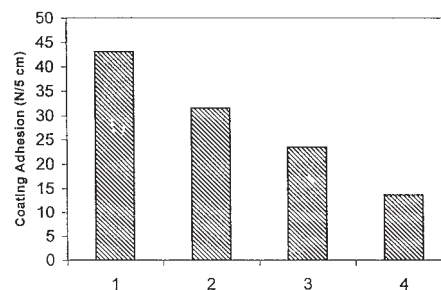


Figure 2 Coating adhesion of (1) jute with natural rubber containing an accelerating system, (2) jute with natural rubber containing no accelerating system, (3) cotton with natural rubber containing an accelerating system, and (4) polyester with natural rubber containing an accelerating system.

However, such formations of chemical linkages between jute and natural rubber are expected to be far more frequent and consequential in the presence of an accelerator and an activator.

Adhesion of natural rubber to the substrate

The adhesion of natural rubber to jute, cotton, and polyester is shown in Figure 2. The results show that the adhesion of natural rubber to the substrates considered in this study follows the order jute > cotton > polyester. The adhesion of polyester to natural rubber has been reported to be poor because it offers an inactive fiber surface.^{2,10} Among the substrates studied, cotton has an intermediate level of adhesion. The adhesion of jute to natural rubber, even in the absence of any accelerator and activator system, appears to be substantially higher than the adhesion of polyester and cotton to natural rubber; however, it is improved further upon the incorporation of an accelerator and activator (Fig. 2). The high level of adhesion of natural rubber to

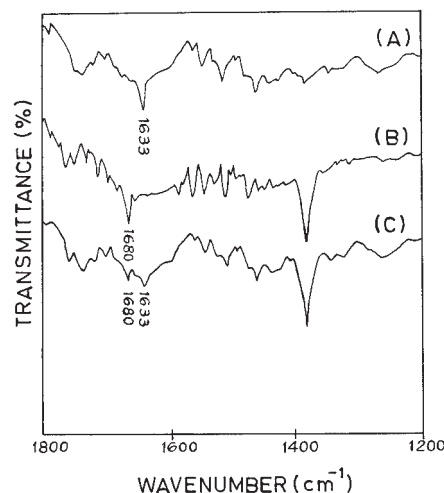


Figure 3 IR spectra of (A) jute, (B) unvulcanized natural rubber, and (C) a vulcanized jute/natural-rubber composite.

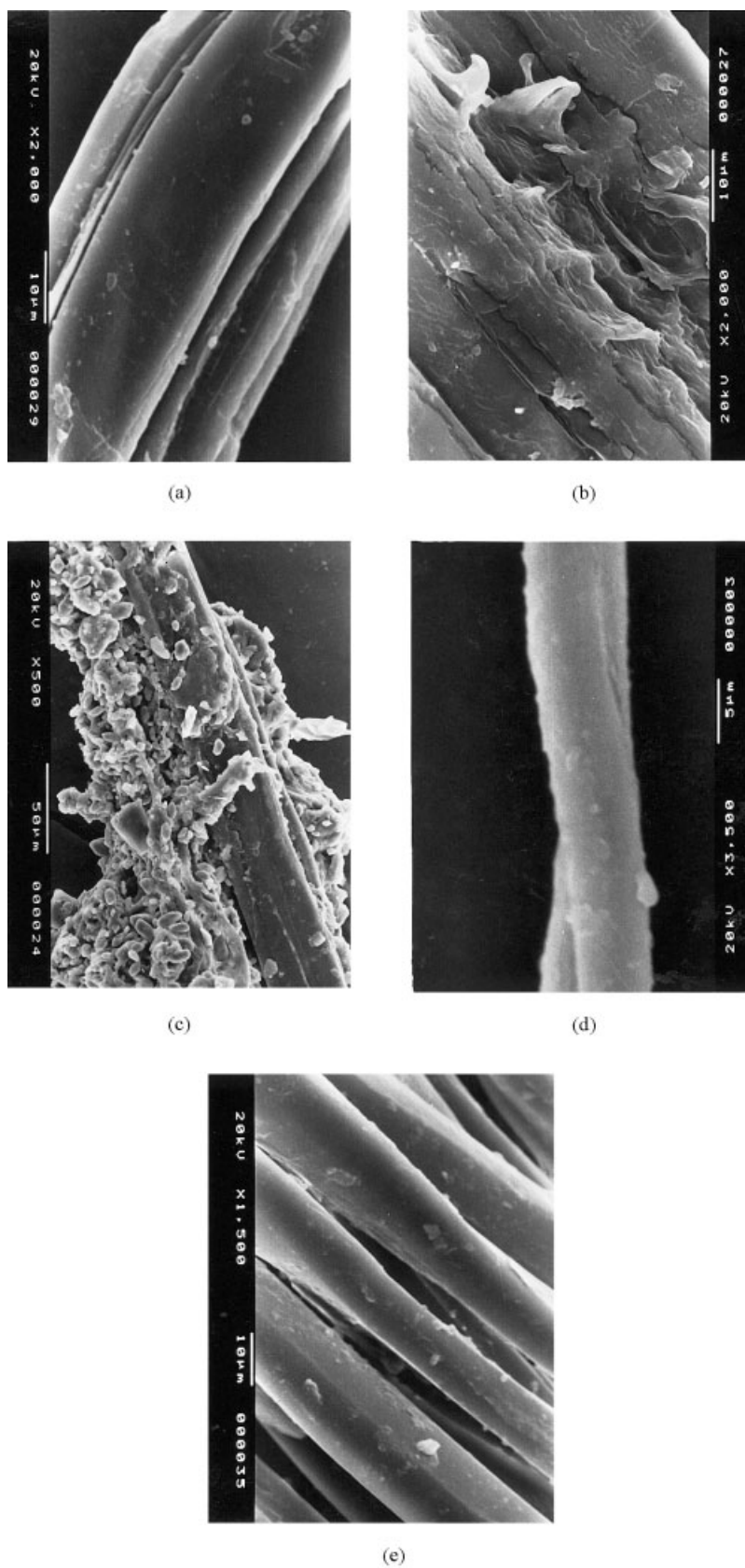


Figure 4 Scanning electron micrographs of (a) uncoated jute (2000 \times), (b) natural-rubber-coated jute (2000 \times), (c) natural-rubber-coated jute (500 \times), (d) natural-rubber-coated cotton (3500 \times), and (e) natural-rubber-coated polyester (1500 \times) fibers taken from the fabric/natural-rubber interface.

jute is presumably due to the good mechanical and chemical adhesion of natural rubber to the fiber. The unique surface features of jute and the unsaturation present in lignin appear to be responsible for such mechanical and chemical adhesion, respectively.

IR analysis

IR spectra of uncoated jute [Fig. 3(A)], an unvulcanized natural-rubber compound with no accelerating system [Fig. 3(B)], and a jute/natural-rubber composite [Fig. 3(C)] are shown in Figure 3. In the spectrum of uncoated jute, the absorption at 1633 cm^{-1} , appearing with a strong intensity, is the characteristic of the carbon-carbon double bond conjugated with the aromatic ring;¹¹ this absorption band arises from the three chief precursors of lignin (coniferyl, sinapyl, and cumaryl alcohols). In the spectrum of the unvulcanized natural rubber, the absorption at 1680 cm^{-1} , appearing with a medium intensity, is the characteristic of the carbon-carbon double bond present in the hydrocarbon chain of polyisoprene. Substantial weakening of both absorption bands at 1633 and 1680 cm^{-1} in the spectrum of the jute/natural-rubber composite indicates the establishment of a chemical bond between the lignin fraction of jute and the polyisoprene chain via sulfur by addition to their respective double bonds during vulcanization under the given conditions; as a result, the double bonds are partly saturated in the composite. The results of the IR analysis agree with the proposed mechanism.

Scanning electron microscopy

Scanning electron micrographs of jute without coating and of jute, cotton, and polyester fibers with a natural-rubber coating (taken from the substrate fabrics when the fabrics were separated from the coating polymer during the determination of adhesion) are shown in Figure 4. Figure 4(a) for the simple jute fiber clearly shows its multicellular structure with a distinct presence of an intercellular region between the neighboring unit cells. The coating of jute with natural rubber brings about some change in the surface morphology of the jute fiber [Fig. 4(b)]; a developing masking effect by a cohesive film of rubber and a partially filled intercellular space with randomly flown and tenaciously adhered rubber are distinctly visible. Such a masking effect is, however, much more pronounced because of the overwhelming spread and large-scale deposition of natural-rubber moieties on the jute fiber [Fig. 4(c)], which tenaciously adhered and remained unseparated even when mechanical force was applied

for the determination of the adhesion of the coating polymer to the substrate. Such tenacious adherence of natural rubber to jute fiber is the result of chemical and mechanical adhesion of natural rubber to jute fiber. Natural rubber in its unvulcanized plastic state flows freely during coating and fills even the intercellular regions of jute fiber, which are separated by its unit cells. When vulcanized, such coated natural rubber, with an interfacial morphology conforming to that of jute at the microlevel, is chemically bound with jute at the interfacial region, in addition to its self-crosslinking at the bulk.

The surface morphology of cotton fiber with its typical convolution [Fig. 4(d)] and that of polyester [Fig. 4(e)] show that such deposition and spread of inseparable natural-rubber moieties are discontinuous, limited, and far less than those observed for jute fiber [Fig. 4(b,c)].

CONCLUSIONS

The use of jute fabric in the manufacture of DT rubberized fabric and fabric-reinforced rubber sheeting for hospitals imparts to the products an improved balance of physical, mechanical, and physicochemical properties superior to that obtainable from existing polyester- or cotton-based products of this kind, with respect to the fabric-to-natural-rubber adhesion, TS, tear strength, abrasion resistance, and particularly puncture resistance and biodegradability for DT rubberized fabric. The incorporation of coarse jute fabrics, however, makes the products heavy and thick.

IR spectroscopy and scanning electron microscopy analysis indicate that the high adhesion of natural rubber to jute is the direct result of the appreciable mechanical anchorage of natural rubber to the irregular surface of jute and the notable crosslinking of natural rubber with the lignin fraction of jute through sulfur.

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