

Interfacial Modification of High Impact Polystyrene Magnesium Hydroxide Composites Effects on Flame Retardancy Properties

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ABSTRACT: High impact polystyrene (HIPS)/magnesium hydroxide (MH) composites were prepared by melt-blending. Two kinds of interfacial modifiers were used in this research, maleinated poly[styrene-*b*-(ethylene-*co*-butylene)-*b*-styrene] (SEBS-*g*-MA) triblock copolymer and PS. The effects of the use levels of SEBS-*g*-MA on the flame retardancy of HIPS/elastomer/MH based on unmodified and PS-modified surface were investigated by TEM, FTIR, and combustion tests (horizontal burning test and cone calorimetry). The combustion results showed that comparing composites containing unmodified MH, the flame retarding properties of composites containing PS-modified MH were obviously improved. The increased performance can be explained that the PS covered on the surface of MH could further improve dispersion of the filler in

matrix. Furthermore, there existed a critical thickness of interfacial boundary for optimum flame-retarding properties in both ternary composites based MH and PS-modified MH. When the interfacial boundary relative thickness is less than 0.53, the introduction of SEBS-*g*-MA can improve the dispersion degree, leading the improvement of flame retardancy properties. However, with the increase of interfacial boundary thickness, the SEBS-*g*-MA coating around MH acted as a heat and mass transfer barrier, leading to the reduction of flame retardancy. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 578–583, 2008

Key words: high impact polystyrene; magnesium hydroxide; cone calorimetry; flame retardancy; interfacial boundary

INTRODUCTION

In recent years, a great deal of attention has been paid to the use of flame retardant (FR) polymeric composites with halogen-free (HF) FRs, for example conventional MH or alumina trihydrate (ATH), because these materials do not cause environmental pollution, such as toxicity, heavy smoke, and corrosion.^{1–3} MH, by virtue of its high decomposition temperature, can be used in a wider range of thermoplastics than ATH.⁴

The disadvantage of both these fillers, however, is the high level contents (more than 60%w/w) required to achieve the desired FR effect.⁴ Additions of fillers in such high amounts adversely affect mechanical properties of the resulting materials, including reducing elongation at break, sacrificing impact strength, increasing melt viscosity.^{5–8} This problem

can be solved through improving the dispersion and compatibility of the inorganic filler in polymer matrix. While a variety of surface modifiers can be effective dispersants for fillers, only coupling agents and polymer compatibilizers are found to be able to form a chemical connection between the two phases. The coupling agents are designed to be a molecular bridge. Such low-molecular agents with very short chains, e.g., stearic acid, are hard to anchor to the polymer matrix through physical entanglements and van der Waals interactions which has limited adhesion effect.^{9–11} The compatibilizers consist of two parts, the hydrophobic polyolefin copolymer or grafting with polar hydrophilic monomers, such as acrylic acid (AAC), methyl methacrylate (MMA), and maleic anhydride (MAH). These reactive groups interact readily with functional groups on the inorganic filler, and the polyolefin phase is miscible or compatible with the polymer matrix. A durable connection between the filler and the matrix is thereby established. So compatibilizers are also termed “polymer coupling agents.”^{11,12} Many studies about polyolefin/ATH composites were focused on the effects of coupling agents or compatibilizers. Liauw

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et al. systematically studied the effects of various coupling agents on the mechanical properties of polymer/MH composites.^{5,13–30} A recent work reported by Hippel et al. showed that the presence of compatibilizers could improve both stiffness and toughness of PE/ATH composites while FR properties of the composites remained unchanged.³ However, there is little information in the literature about the effects of compatibilizers on the combustion characteristics of magnesium hydroxide (MH) filled high impact polystyrene.

In our laboratory, a new method of polystyrene-encapsulated inorganic FR was explored and the effects of polystyrene-encapsulated magnesium hydroxide (MH) on the rheological and FR properties of high impact polystyrene (HIPS) composites were investigated in our previous work.^{31–34} The present investigation was mainly concerned with flame retardancy of HIPS/poly[styrene-*b*-(ethylene-*co*-butylene)-*b*-styrene] (SEBS-*g*-MA) triblock copolymer/MH based unmodified and modified surface. The modified MH used in this work was encapsulated with desired amounts of PS by *in situ* encapsulation method developed in our laboratory.³⁴ It was expected that the PS covered on the surface of MH could further improve dispersion of the filler in the matrix and enhance the interfacial adhesion between filler-elastomer and/or filler-matrix, leading to improve the properties of the resulting composites.

EXPERIMENTAL

Materials

HIPS (PH-88HT, melt flow index = 4.5 g/10 min at 200°C and 5 kg) with a density 1.04 g/cm³ was provided by Zhenjiang Chimei Corp. Magnesium hydroxide, employed as filler, was a high purity untreated grade (Magnifin H5) supplied by Martinswerk GmbH. The average volume particle size was 1.5 μm and the specific surface area was 5.0 m²/g. The maleinated poly[styrene-*b*-(ethylene-*co*-butylene)-*b*-styrene] (SEBS-*g*-MA) triblock copolymer, KratonTM FG1901X was supplied by Shell Chemicals (29% styrene, nominal weight of grafted maleic anhydride = 1.8 ± 0.4%, quoted at 1.84% as measured by elemental analysis).³⁵ Styrene (St), azobisisobutyronitrile (AIBN) and hydrochloric acid were purchased from Shanghai Chemical Reagents Company and used without further treatment, except St which was distilled before use.

Preparation of MH-T

Magnesium hydroxide (MH) was dried at 120°C for 6 h, and then was put into high speed mixer by

TABLE I
Composition and Flammability of the Flame Retarded HIPS and its Composites

Sample code	Composition (wt %)				HBR (mm/min)
	HIPS	MH ^a	MH-T ^b	SEBS- <i>g</i> -MA	
1	100	0	0	0	35.8
2	60	40	0	0	21.0
3	56	40	0	4	13.9
4	52	40	0	8	14.6
5	48	40	0	12	FH-2
6	44	40	0	16	FH-2
7	40	40	0	20	15.1
8	57.6	0	42.4	0	14.6
9	53.6	0	42.4	4	16.5
10	49.6	0	42.4	8	15.0
11	45.6	0	42.4	12	13.3
12	41.6	0	42.4	16	13.7
13	37.6	0	42.4	20	23.3

^a MH represents untreated MH.

^b MH-T represents MH encapsulated with 6.0 wt % polystyrene by *in-situ* polymerization (i.e. charge rate of PS/MH:6/100 by weight).

heated to 70°C with a rotation speed of 1200 rpm. Then acetone solutions of 3-(methacryloxy)propyltrimethoxy silane (MPS) with water and acetic acid was added under stirring for 20 min. The concentration of MPS in acetone was 37.5 wt %. The use level of this solution was 8 mL/100 g MH. The modified powder was dried at 80°C for 12 h. Finally, modified powder, monomer (St), additional initiator (AIBN) were placed into the mixer with agitation fixed at 1200 rpm and heated in an oil bath at 80°C for 45 min.^{31–34} The crude product at the end of polymerization was dried. And the charge rate of PS/MH was 6.0 wt %. Note that the charge rate of PS/MH was selected based on the earlier experimental results on flame retardancy of HIPS/PS-encapsulated MH composites.³⁴ The PS/MH was designated MH-T in the following discussion.

Material preparation

These raw materials were first mixed in a high speed mixer for about 20 min. Then the mixture was extruded and pelletized in a corotating twin-screw extruder (Nanjing Keya Machinery Co., Ltd) with an *L/D* ratio of 42 and a screw diameter of 35 mm. The temperature profiles of the barrel were 180, 190, 200, 210, 210, and 215°C from the hopper to the die, respectively. The screw speed was set at 400 rpm. A series of samples with different composition were obtained as shown in Table I. The specimens for combustion were molded with an injection molding machine (CJ80M2V, Zhende Plastics Machinery Co., Ltd) and the injection temperature was 210°C.

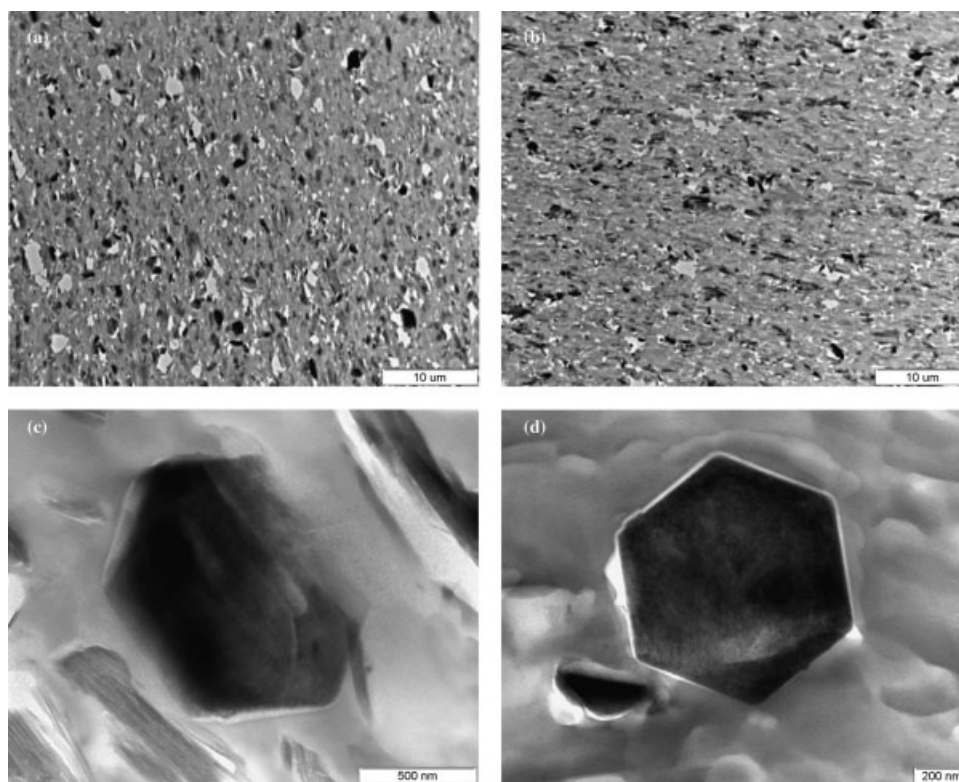


Figure 1 TEM micrographs of ternary composites. (a) sample 4 at low magnification (b) sample 10 at low magnification, (c) sample 4 at high magnification (d) sample 10 at high magnification.

Characterization

To investigate the dispersion of MH and SEBS-*g*-MA particles in HIPS and interfacial adhesion between them, ultra-thin sections (100 nm in thickness) are cut with a diamond knife using a Leica Ultracut S microtome. Thin specimens were collected in a trough filled with water and placed on 200 mesh copper grids. Subsequently, the thin sections were observed using a Hitachi H-800 transmission electron microscope at an accelerating voltage of 60 kV.

The composites were Soxhlet extracted for 72 h with toluene (to remove the matrix HIPS and SEBS-*g*-MA). The residues were then oven dried to constant mass at 150°C. The bound matrix content was then determined by FTIR spectroscopy with a Nicolet 5700 FTIR spectrometer in the range 4500–500 cm^{-1} , with a resolution of 2 cm^{-1} .

Combustion test

The UL-94 horizontal burning test was carried out using a CZF-3 type instrument (made in china) on sheets $127 \times 12.7 \times 3 \text{ mm}^3$ according to ASTM D 635-77.

Fire behavior was also characterized by a cone calorimeter according to ASTM E 1354/ISO 5660 (Fire Testing Technology, East Grinstead, UK). All samples (100 mm in diameter and 4 mm in thickness)

were measured in a horizontal position. The heat flux was maintained at 35 kW/m^2 . The real irradiation sample area was 0.007854 m^2 . Exhaust flow was set at 24 L/s and the spark was continuous until the sample ignited. All samples were run in triplicate and the average value is reported; results from the cone calorimeter are generally considered to be reproducible to $\pm 10\%$.

RESULTS AND DISCUSSION

TEM observation

In ternary composites of polymer/elastomer/inorganic filler, the dispersion state of elastomer and filler plays an important role in determination of properties of composites. Therefore, microstructural characterization of the ternary composites was carried out in detail with the aid of transmission electron microscopy.

TEM micrographs of the ternary composites based MH and MH-T are shown in Figure 1(a–d). The dark holes represented MH particles and the white holes were the spaces of the MH or their aggregations which were cut from the thin section. TEM micrographs of the ternary composites based untreated MH are shown in Figure 1(a,c). The dark holes and the white voids are big. The broad size distribution of dispersion phase indicated that there existed seriously particle agglomeration. However, for the MH-T based

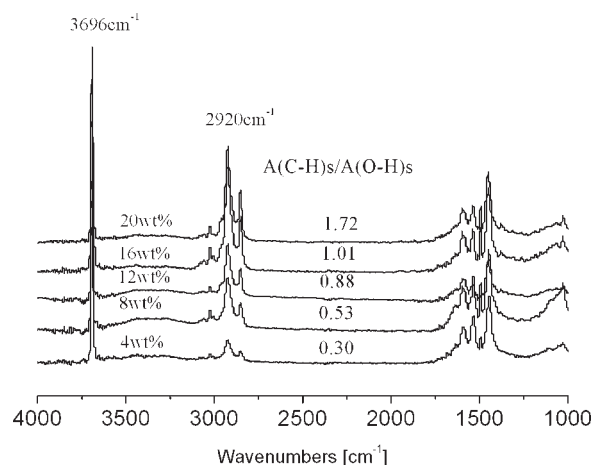


Figure 2 FTIR spectra (normalized to $A(O-H)_s$ of $Mg(OH)_2$) of ternary HIPS/MH-T/SEBS-g-MA composite residues containing different levels of SEBS-g-MA after extraction with toluene for 72 h.

composites, the holes appeared to be evenly distributed in the matrix without obvious particle agglomeration, as shown in Figure 1(b). In addition, the interfacial morphology of the ternary composites based MH and MH-T was also observed, as shown in Figure 1(c,d). It can be seen that SEBS-g-MA could tightly encapsulate MH particles having no obviously phase separation due to the strong interaction between maleic anhydride groups of SEBS-g-MA and MH particles. However, for the MH-T based ternary composites, phase separation gap was observed (about 10–20 nm). This result demonstrated that the interfacial adhesion between MH and SEBS-g-MA was affected resulting from the introduction of PS on the surface of MH, leading to the generation of light phase separation. These results showed that the introduction of PS on the surface of MH could improve the dispersion degree of MH and reduce interfacial adhesion between filler-matrix or filler-elastomer in HIPS/MH/SEBS-g-MA composites. The reason may be attributed to adsorption of SEBS-g-MA onto a pre-adsorbed layer of PS on MH resulted in a very significant reduction in polymer adsorption activity due to blockage of adsorption sites.¹⁴

Assessment of interfacial adhesion qualities of composites

Properties of polymer composites depend on many factors, such as compositions, interphase interactions and the size and morphology of the dispersed phase. However, for highly filled composites, filler-matrix interfacial properties may be the major determinant of composite mechanical properties. Liau et al. reported that the use of FTIR spectra of composites to calculate relative interphase thickness is a successful way to elucidate the enhancement of the interfacial adhesion between polymer matrix and filler particles.²⁸

The FTIR spectra of ternary HIPS/MH-T/SEBS-g-MA composite residues after matrix extraction is shown in Figure 2. The integrated area ratios of the C—H stretching (centered at 2920 and 2850 cm^{-1}) absorptions ($A(C-H)_s$) of the bound matrix to the O—H stretching (centered at 3696 cm^{-1}) absorptions ($A(O-H)_s$) of the filler (i.e., $A(C-H)_s/A(O-H)_s$) provide a measure of the level of bound matrix. It can be clearly seen that the $A(C-H)_s/A(O-H)_s$ value of the composite residues increased with the increase of the content of SEBS-g-MA in the composites, indicating that the interfacial boundary relative thickness increase with increase of the content of SEBS-g-MA. At the same time, FTIR spectra of the ternary composites based MH were also obtained. The $A(C-H)_s/A(O-H)_s$ values were similar as the above-mentioned. The results showed that the relative thickness of interfacial boundary increase with increasing content of SEBS-g-MA in both ternary composites.

Horizontal burning rates (HBR)

Horizontal burning tests are widely used to evaluate FR properties of materials. Table I lists the horizontal burning data obtained from all the ternary composites. It clearly showed that horizontal burning rate substantially decreased with increasing SEBS-g-MA content for both series of composites when the content of SEBS-g-MA was no more than 16 wt %. These results indicated that the introduction of SEBS-g-MA could improve the dispersion of MH. However, the higher burning rate observed when the content of SEBS-g-MA was over 16 wt %.

Combustion of HIPS/SEBS-g-MA/MH composites

The changes of HRR and THR as a function of burning time for HIPS/MH composites containing different content of SEBS-g-MA are shown in Figure 3. The various parameters that may be evaluated using cone

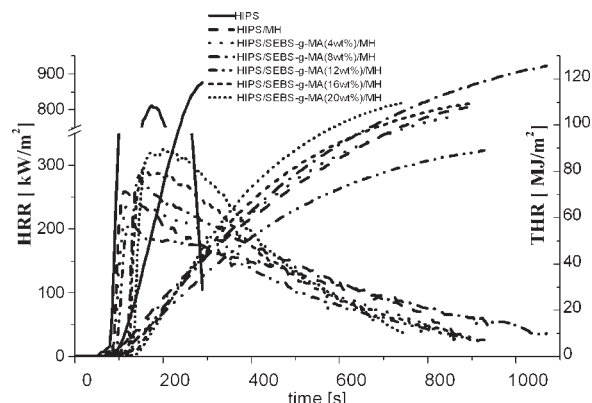


Figure 3 Heat release rate and total heat release for composites containing MH.

TABLE II
Data Obtained from the Cone Calorimeter Tests of Composites Containing MH

Samples	1	2	3	4	5	6	7
Elastomer content (wt %)	–	0	4	8	12	16	20
Time to ignition (s)	65	74	73	106	79	107	118
PHRR (kW/m ²)	806	260	244	285	208	295	325
Time to PHRR (s)	170	115	125	145	130	155	200
FPI (m ² s/kW)	0.080	0.285	0.299	0.372	0.380	0.363	0.363
MHRR ₁₈₀ (kW/m ²)	320	115	107	84	83	79	69
THR (MJ/m ²)	119	108	103	125	89	109	110
Average EHC (MJ/kg)	28	31	34	38	35	34	34
Mass loss rate (g/m ² s)	14.0	3.5	3.6	3.1	3.4	3.6	4.2
SEA (m ² /kg)	907	732	764	722	721	603	660

calorimetry, including the time to ignition, t_{ign} , the heat release rate curve, especially its peak value, the peak heat release rate, PHRR and the time to PHRR, t_{PHRR} , Mean heat release rate from ignition to 180 s, $MHRR_{180}$, the mass loss rate, MLR, and the specific extinction area, SEA, total heat evolved, THR, effective heat of combustion, EHC, are tabulated in Table II. It is striking that there is no change in PHRR with less than 12 wt % SEBS-*g*-MA; it must be remembered that the usual spread of values in a cone experiment is $\pm 10\%$, so any change that is less than 10% is considered to be no change. When 12 wt % SEBS-*g*-MA is present, there is a measurable reduction in PHRR which exceeds the 10% value and this must be considered significant. This result showed that the incorporation of a suitable amount of SEBS-*g*-MA can improve the dispersion of MH in the matrix, leading to a decrease in PHRR. However, with the continuous increase of SEBS-*g*-MA, the values in PHRR are higher than that the composites without SEBS-*g*-MA. This result confirmed that extent of filler dispersion reached limiting values at the optimum SEBS-*g*-MA level in HIPS/MH/SEBS-*g*-MA.

Combustion of HIPS/SEBS-*g*-MA/MH-T composites

The changes of HRR and THR as a function of burning time for HIPS/MH-T composites containing different content of SEBS-*g*-MA are shown in Figure 4. The various parameters that may be evaluated using cone calorimetry are shown in Table III. It showed that when 4–12 wt % SEBS-*g*-MA is present, there is an evidently reduction in PHRR than that of the composites without SEBS-*g*-MA. However with an increase of SEBS-*g*-MA level, the flame-retarding properties of the composites declined somewhat, compared with the composites without SEBS-*g*-MA. This result showed that there appeared to be a critical SEBS-*g*-MA level, namely 8.0 wt %, for optimum flame-retarding properties,

In addition, fire performance index (FPI) is defined as the ratio of TTI to PHRR, which is inde-

pendent of the tested sample thickness and is often used to predict whether a material can easily develop combustion after ignition.³⁶ Therefore, the greater the FPI value, the better is the fire resistance. Tables II and III also lists the FPI values for all samples in this study. It can be seen that the FPI values of HIPS/MH-T/SEBS-*g*-MA are higher than that of HIPS/MH-T composites when the content of SEBS-*g*-MA was no more than 8.0 wt %. And the greatest value of FPI is 0.867 corresponding to the HIPS/MH-T composites with 8.0 wt % SEBS-*g*-MA, which was much higher than that of corresponding HIPS/MH/SEBS-*g*-MA composites, 0.372. However, with the continuous increase of SEBS-*g*-MA, FPI values of the ternary composites decreased obviously.

It can be seen that the cone calorimeter results corresponded well to the results of horizontal burning tests both of which showed that the fire properties of ternary composites containing untreated MH can be enhanced by the incorporation of a suitable amount of SEBS-*g*-MA as an adhesion promoter. Furthermore, comparing to the ternary composites containing untreated MH, the FR properties of ternary composites containing PS-encapsulated MH (MH-T) were found to be significantly improved. This improvement is mostly attributed to a better dispersion of the encapsulated filler in the matrix, as

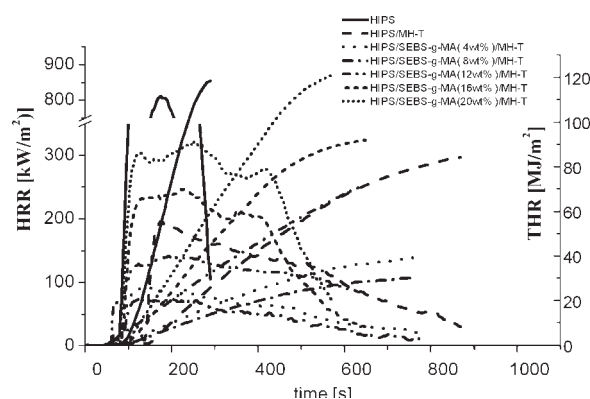


Figure 4 Heat release rate and total heat release for composites containing MH-T.

TABLE III
Data Obtained from the Cone Calorimeter Tests of Composites Containing MH-T

Samples	8	9	10	11	12	13
Elastomer content (wt %)	0	4	8	12	16	20
Time to ignition (s)	129	65	65	65	68	63
PHRR (kW/m ²)	196	92	75	143	247	324
Time to PHRR (s)	175	140	125	205	230	250
FPI (m ² s/kW)	0.658	0.696	0.867	0.455	0.275	0.194
MHRR ₁₈₀ (kW/m ²)	35	44	34	73	106	142
THR (MJ/m ²)	84	40	30	71	92	121
Average EHC (MJ/kg)	35	14	9	24	39	39
Mass loss rate (g/m ² s)	3.8	4.0	4.0	5.2	4.8	6.0
SEA (m ² /kg)	775	740	619	656	654	553

revealed by TEM observations. In addition, with the continuous increase of SEBS-g-MA, the flame retardancy of both ternary composites declined somewhat. This result showed that there existed a critical relative thickness of interfacial boundary for optimum flame-retarding properties in both ternary composites based MH and MH-T, as revealed by FTIR. When the interfacial boundary relative thickness is less than 0.53, the introduction of SEBS-g-MA can improve the dispersion degree, leading the improvement of flame retardancy properties. However, with the increase of interfacial boundary relative thickness, the SEBS-g-MA coating acted as a heat and mass transfer barrier, leading to the reduction of flame retardancy.

CONCLUSIONS

In this work, the effect of the use levels of SEBS-g-MA on the flame retardancy of HIPS/elastomer/MH based on unmodified and modified surface was investigated. The combustion results showed that the fire properties of HIPS/MH composites can be enhanced by the introduction of optimum content of SEBS-g-MA as an adhesion promoter. The modified MH (MH-T) used in this work was encapsulated with desired amounts of PS by *in situ* encapsulation method developed in our laboratory. The results showed that the PS covered on the surface of MH could improve dispersion of the filler in matrix and enhance the interfacial adhesion between filler-elastomer and/or filler-matrix, leading to further improvement in the FR properties of the ternary composites.

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