J. Myanmar Acad. Arts Sci. 2019 **Vol. XVII. No.2A**

STRUCTURAL ANALYSIS, MICROSTRUCTURAL INVESTIGATION AND ENERGY BAND GAP DETERMINATION OF ZINC OXIDE AND MAGNESIUM TITANATE MIXED COMPOSITE

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Abstract

Mixed composite materials of Zinc Oxide and Magnesium Titanate ($ZnO:MgTiO_3$) with the compositions of (20%:80%, 40%:60%, 60%:40% and 80%:20%) were prepared by using solid state reaction method. The samples were characterized by powder X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM) and Ultra-Violet-Visible-Near Infrared (UV-VIS-NIR) spectroscopy to analyze the structural, microstructural and optical properties of the samples. The crystalline phase formation and the structural properties were analyzed from the observed XRD patterns. Microstructural properties of grain shape and sizes were investigated by SEM. The energy band gaps E_g were determined from the Tauc plot of $(\alpha h\upsilon)^2$ vs. $h\upsilon$ graphs. It was found that the energy band gap decreased with increase in ZnO composition.

Keywords**:** Mixed composite, $ZnO:MgTiO_3$, XRD, SEM, UV-VIS-NIR, energy band gaps

Introduction

In recent years, TiO_2 has been well known as a semiconductor with photocatalytic activities and has a great potential for applications such as environmental purification. TiO_2 is mainly applied as pigments, adsorbents, catalyst supports, filters, coatings, photoconductors, and dielectric materials. The performance of the material (TiO_2) affected by the size of the particles. Thus particle size plays a great role [Ruhela, (2013)]. Zinc oxide (ZnO) has a relatively large energy band gap (~ 3.3 eV) at room temperature. The band gap of zinc oxide may be increased to nearly 3 – 4 eV by alloying it with

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magnesium oxide. Zinc oxide is commonly used in laser diodes and light emitting diodes (LED). Some optoelectronic applications of ZnO overlap with that of GaN, which has a similar band gap (~ 3.4 eV at room temperature). Now different compositions of materials ($MgTiO_3$ with ZnO) are analyzed by X-ray diffraction (XRD) and comparative study will be observed with the obtained XRD pattern [Azurdia, (2006); Ferri, (2009)]. Magnesium titanate ($MgTiO_3$) has potential applications such as high frequency capacitors, chip capacitors, and temperature compensating capacitors, resonators, filters, antennas for communication, radar and direct broadcasting satellite [Tao, (2011); Wang, (2010)]. In this work, mixed composite materials of $ZnO:MgTiO_3$ were prepared by solid state reaction method and their structural, microstructural and optical properties were reported by XRD, SEM and UV-VIS-NIR spectroscopic measurements.

Materials and Method

Sample Preparation

Mixed composite materials of zinc oxide and magnesium titanate ($ZnO:MgTiO_3$) with the compositions of 20%:80%, 40%:60%, 60%:40% and 80%:20% were prepared from the starting materials of Zinc Oxide (ZnO), Magnesium Oxide (MgO) and Titanium Dioxide (TiO_2). Starting materials of Analytical Reagent (AR) grade those oxide materials were weighed with desired compositions and the weighed powders were mixed each other. The mixed materials were heated at 800°C for 5 h in JLabTech electric oven. The heated composite samples were ground by an agate mortar for 1 h to obtain fine powder. Flow diagram of the sample preparation procedure is shown in Figure 1. Photographs of the sample preparation processes are shown in Figures 2(a – d) respectively.

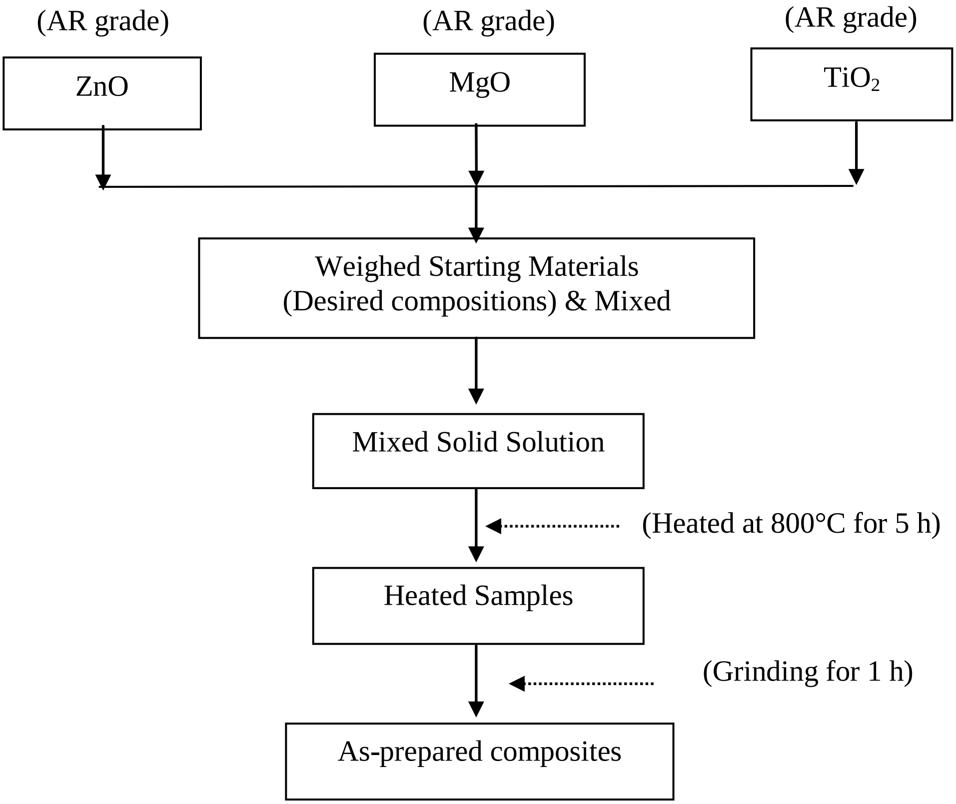


Figure 1: Flow diagram of the sample preparation procedure

Figure 2: Photographs of the weighed starting materials of (AR) grade (a) ZnO, (b) TiO_2 and (c) MgO powders for the preparation of 80%:20%, 60%:40%, 40%:60% and 20%:80% ZnO:$MgTiO_3$ composite materials

Figure 2: (d) Photograph of the as-prepared (80%:20%, 60%:40%, 40%:60% and 20%:80%) ZnO:$MgTiO_3$ composite materials

XRD, SEM and UV-VIS-NIR Measurements

The XRD spectra of the ZnO:$MgTiO_3$ were observed by Rigaku MiniFlex 600 X-Ray Diffractometer [Department of Physics, University of Yangon] using the CuK_α radiation with wavelength of 1.54056 Å. The microstructural properties were investigated by using JEOL JCM-6000Plus Scanning Electron Microscope (SEM) [Department of Physics, West Yangon University] with the accelerating voltage of 15 kV and 7000 times of photo magnification. UV-VIS-NIR (Ultraviolet-Visible-Near Infrared) transmission spectra were collected on PC-controlled SHIMADZU UV-1800 Spectrophotometer [AMTT, Co. Ltd., Yangon] in the wavelength range of 190 nm – 1100 nm.

Results and Discussion

XRD Analysis

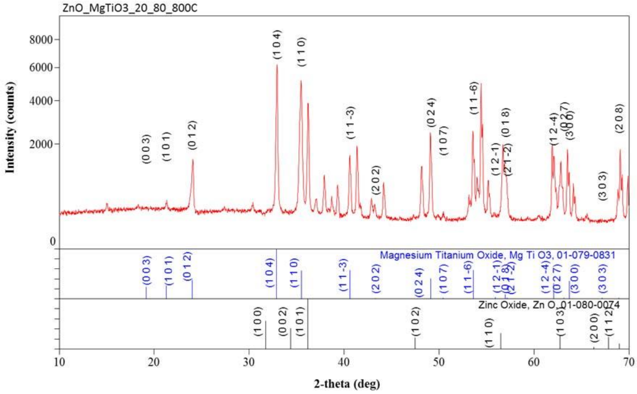
Powder X-ray diffraction patterns of the samples are shown in Figure 3(a – d). To assign the observed XRD lines, the lines were identified by using the Joint Committee on Powder Diffraction Standards (JCPDS) data library files of (i) Cat. No. 01-079-0831> Magnesium Titanium Oxide – $MgTiO_3$ and (ii) Cat. No. 01-080-0074 Zinc Oxide – ZnO for the samples. Most of the observed XRD lines were assigned by the standard JCPDS data library files but some of the lines were not assigned due to the dual-phase of composite materials of $ZnO{:}MgTiO_3$.

XRD patterns show that the samples analogous to the hexagonal structure. The lattice parameters of the samples are evaluated by the equation: $\frac{1}{d^2} = \frac{4}{3}\left(\frac{h^2 + hk + k^2}{a^2}\right) + \frac{l^2}{c^2}$, where *d* is the interplanar spacing (Å), *a* and *c* are the lattice parameters (Å) of the unit cell, (*hkl*) are the Miller indices. The obtained lattice parameters of the samples are tabulated in Table 1. The diffraction lines appear in the diffraction angle range of about 23° – 32° are not assigned with the JCPDS data library files because these lines may be attributed to the heat treatment effect of the ZnO and $MgTiO_3$ samples. They are new compound formation from the ZnO and $MgTiO_3$ samples due to thermal agitation.

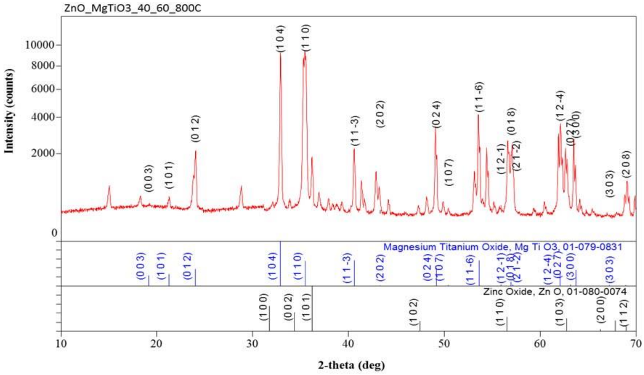
The Scherrer formula for the calculation of crystallite size (grain diameter), D, of the sample is $D = k\lambda / \beta \cos\theta$, where, D is the crystallite size (nm) , k is a constant varies with crystallite shape but usually nearly equal to 0.94, λ is the wavelength of source radiation (Å)and β is full-width at half maximum (FWHM) of the peak (radian) and θ is Bragg's angle (Å). The crystallite sizes are listed in Table 1.

SEM Analysis

Microstructural properties of the solid samples can be analyzed by using Scanning Electron Microscope. SEM micrographs of the samples are shown in Figures 4(a – d). As shown in micrographs, the image reveals that the grain shape of the samples was spherical with the poor boundary. Some of the grains are agglomerated particles. The obtained grain sizes of the samples are tabulated in Table 2.



(a)



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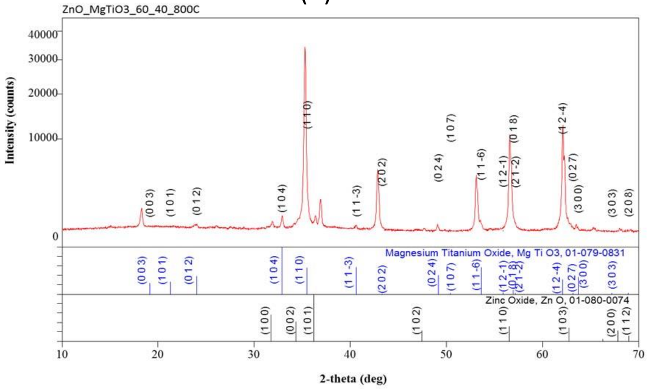

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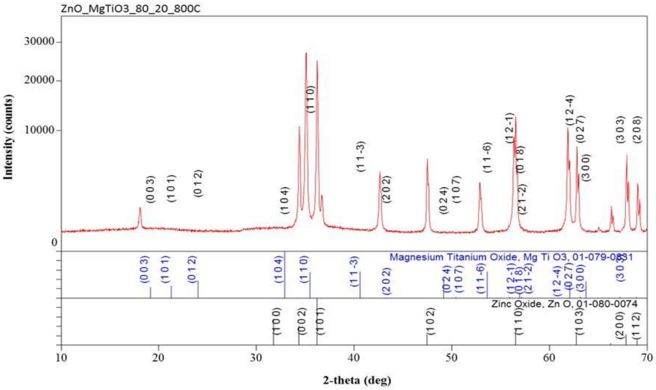
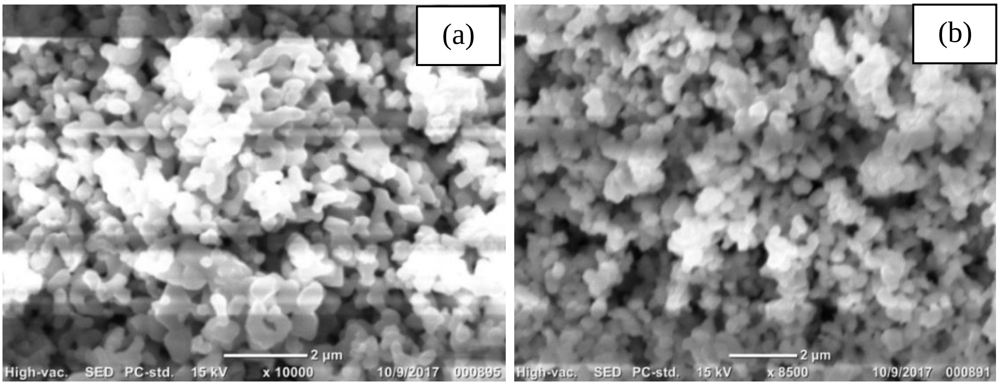


(d)

Figure 3: XRD patterns of the mixed composite materials of ZnO:MgTiO$_3$ with the compositions of (a) (20%:80%), (b) (40%:60%), (c) (60%:40%) and (d) (80%:20%)

Table 1: The lattice parameters and crystallite sizes of the samples

Sample (ZnO:MgTiO$_3$)	$a = b$ (Å)	c (Å)	D (nm)
(20%:80%)	3.2503	5.2158	20.71
(40%:60%)	3.2211	5.1617	72.51
(60%:40%)	3.2463	5.1491	48.27
(80%:20%)	3.2504	5.2069	59.59



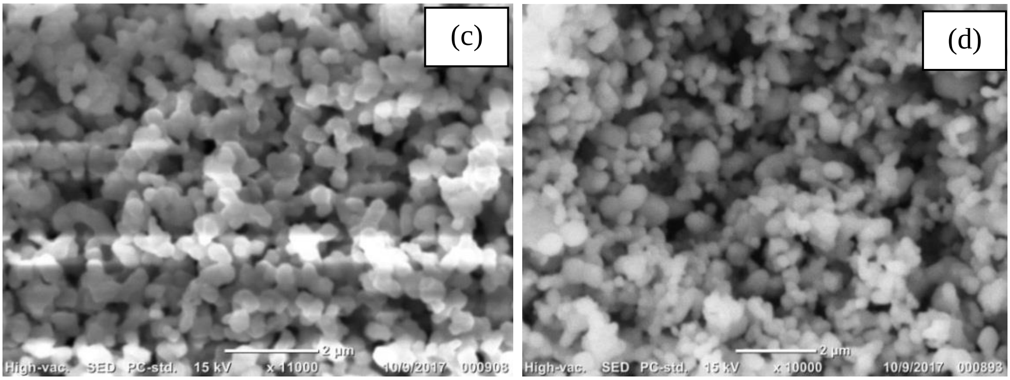


Figure 4: SEM micrographs of the mixed composite materials of ZnO:MgTiO$_3$ with the compositions of (a) (20%:80%), (b) (40%:60%), (c) (60%:40%) and (d) (80%:20%)

Table 2: The grain sizes of ZnO:MgTiO$_3$ mixed composites

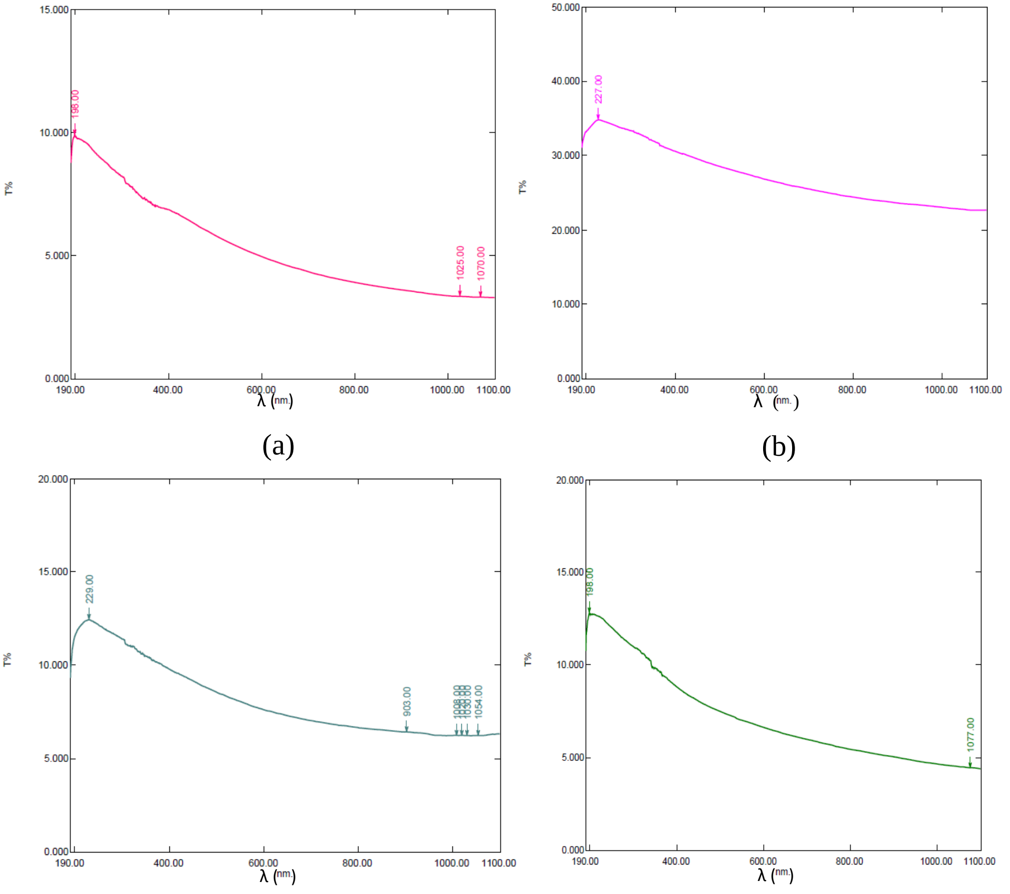
Sample (ZnO:MgTiO$_3$)	**Grain size (μm)**
(20%:80%)	0.18 – 0.50
(40%:60%)	0.20 – 0.50
(60%:40%)	0.22 – 0.45
(80%:20%)	0.18 – 0.72

UV-VIS-NIR Study

The collected UV-VIS-NIR transmission spectra of “T%” vs. “λ" of the ZnO:MgTiO$_3$ with the compositions of 20%:80%, 40%:60%, 60%:40% and 80%:20% samples are shown in Figures 5(a – d). As depicted in figures, the samples demonstrate that less than 100% transmittance of throughout the Ultraviolet-Visible-Near Infrared region.

The energy band gaps E_g of all the samples have been examined with the help of optical absorption and percentage transmission data using the graph of $(\alpha h\nu)^2$ versus hν. The theory of optical transmission gives the relationship between the absorption coefficient “α” and the photon energy “hν’ has a relation; $\alpha = - \ln(1/T)$. From the optical transmission spectrum, the measured transmittance “T” was used to calculate the absorption coefficient “α”. To estimate the energy band gap for all the samples, the graph of $(\alpha h\nu)^2$ versus hν has been plotted. The interception of the line at $\alpha = 0$ in $(\alpha h\nu)^2$ versus hν graph gives the value of energy band gap of the material.

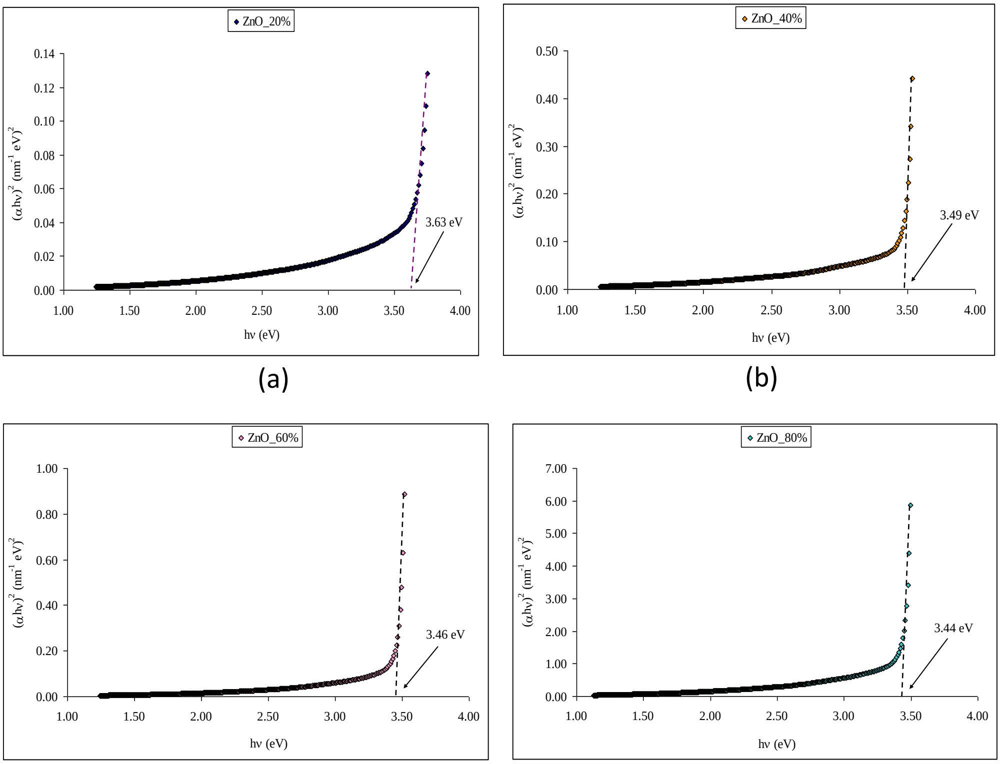
The plots of $(\alpha h\nu)^2$ versus hν of the samples are shown in Figures 6 (a –e). The obtained energy band gaps are listed in Table 3. The variation in band gap may be attributed to the variation of structural parameter (lattice constants) with ZnO semiconducting materials effects on $MgTiO_3$. Ferri, et. al. (2009) was reported that the E_g of $MgTiO_3$ prepared at 700°C was 4.05 eV. In this work the obtained E_g decreased with the increase in concentration of ZnO.



(a) (b)

(c) (d)

Figure 5: UV-VIS-NIR transmission spectra of $ZnO:MgTiO_3$ with the compositions of (a) (20%:80%), (b) (40%:60%), (c) (60%:40%) and (d) (80%:20%)



(a) (b)

(c) (d)

Figure 6: Plots of $(\alpha h\nu)^2$ vs. hν of the ZnO:$MgTiO_3$ samples with the compositions of (a) (20%:80%), (b) (40%:60%), (c) (60%:40%) and (d) (80%:20%)

Table 3: The energy band gaps of ZnO:$MgTiO_3$ mixed composites

Sample (ZnO:$MgTiO_3$)	**E_g (eV)**
(20%:80%)	3.63
(40%:60%)	3.49
(60%:40%)	3.46
(80%:20%)	3.44

Conclusion

Zinc oxide and magnesium titanate (ZnO:$MgTiO_3$) mixed composite materials were prepared by solid state reaction method. XRD patterns show that the samples analogous to the hexagonal structure. SEM micrographs showed that the grain shape of the samples was spherical with the poor boundary. Some of the grains were agglomerated particles. The samples demonstrate that less than 100% transmittance of throughout the Ultraviolet-Visible-Near Infrared region. The energy band gaps E_g decreased with the increase in concentration of ZnO from 20% to 80%.

Acknowledgement

The authors would like to acknowledge Professor Dr Khin Khin Win, Head Department of Physics, University of Yangon, for her valuable discussions and comments on this work.

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