Analysis on Band Gaps of MCM-41 Type of Materials*

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The concept and analysis method of photonic crystals and band gaps are introduced into one-dimensional(1D) ordered mesoporous materials. MCM-41 type of materials are treated theoretically as photonic crystals. The formation of band gaps is exhibited and confirmed by a calculation of transfer matrix technique. PBG was found around 9-42 nm in soft X-ray region. The photonic band-gap was predicted to be dependent on incident direction, pore size and lattice constant. The mesoporous materials with different pore sizes and different lattice constants have different band-gap widths.

Keywords Mesoporous material, Photonic crystal, Photonic band-gap
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Introduction

Since the discovery of M41S family of mesoporous silicates and aluminosilicates in 1992[1,2], numerous mesoporous or nanoporous materials with pore size between 2 and 30 nm have been synthesized because of their potential applications in catalysis, separation of large molecules, medical implants, semiconductor, magnetoelectric devices, *etc*.[3—7]. While many scientific and industrial researches on M41S materials have been focused on the synthesis procedures and chemical behaviour[8—10], there have been few investigations of the optical properties of these materials. To our knowledge these materials have not yet been used as photonic crystals[11,12] and the aim of this paper is to show that one dimensional highly ordered mesoporous materials may be used, in principle, as two-dimensional photonic crystals.

While electron movement in a semiconductive material may be controlled by engineering the electronic band structure of the material, the propagation of an electromagnetic wave within a photonic crystal (PC) may be manipulated by controlling the variation of the refractive index of the crystal. Yablonovitch[13] and John[14] have first shown that PCs may display a range of frequency in which the propagation of an electromagnetic wave in the PCs is completely forbidden. For a PC with a complete photonic band gap (PBG), the electromagnetic wave propagation in the PC is forbidden in all directions, whereas for an incomplete band gap, the PC exhibits the PBG behavior only in a limited range of directions. Depending on the frequency requirement of specific applications, the engineering of the structures which are periodic for waves in all the three dimensions (3D) has been achieved in the infrared, microwave, and millimeter-wave regions. For wavelength shorter than infrared, only several experimental systems have recently been demonstrated to exhibit the signature of complete band gaps, while the study on the PC crystals with PBGs in soft X-ray region is still lacking[15,16].

The pore size and the wall thickness of well ordered MCM-41 type of materials can be controlled in the nanometer range[1—3]. For example, the pore size of MCM-41 can be expanded from about 2.7 nm up to 10 nm by using cosolvent organic molecules, such as 1,3,5-trimethylbenzene. The pore size of SBA-15 can be expanded from about 4.6 nm up to 30 nm by using various surfactants and silica sources. One can expect therefore that the mesoporous materials may be used as PBG crystals with a band-gap located in the

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nanowave regimes. In this work was examined theoretically the PBG for 1D ordered mesoporous materials by means of the transfer matrix method.

Structure

MCM-41 type of materials has a honeycomb structure that is the result of hexagonal pacing of cylindrical pores, but they differ in their pore sizes and lattice constants. A schematic of the structure is shown in Fig. 1. The structure consists of an array of dielectric cylinders, infinitely long in the y-direction and periodic in the xz plane. The cylinders represent pores which are empty (with their refractive index $n = 1$), while the regions between the cylinders are filled with silica. The pore size and its lattice constant of MCM-41 are 3.24 and 5.25 nm according to reference [17]. For SBA-15, its pore size and lattice constant are 20 and 24 nm respectively, according to reference [18]. Here we want to consider what happens when an electromagnetic (EM) wave is incident on the 1D ordered mesoporous structure.

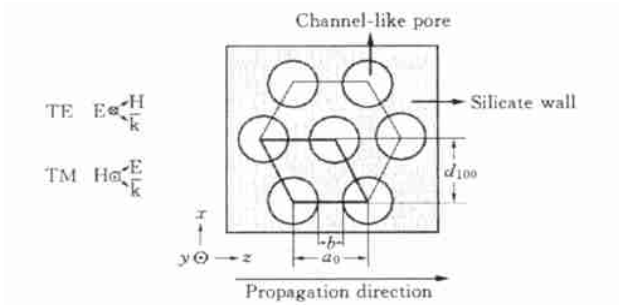

Fig. 1 Schematic diagram showing the set up of the electromagnetic wave propagation and MCM-41 materials.

We define the transverse E-field (TE) or simply E (TM or H) polarized waves as those which have their H (E) field confined to the xy plane, and the corresponding E(H) field parallels to the y axis. The propagation characteristic of an electromagnetic wave inside MCM-41 type of materials was calculated by virtue of "Translight" software package of the University of Glasgow[19,20]. The calculations assumed that MCM-41 type of materials has a finite thickness in the z-direction but is infinite in the y-direction.

Results and Discussion

The transmission response for a MCM-41 material was calculated for various crystal thicknesses. The dielectric constant of the mesoporous silicates was based on Henke′s optical data[21]. The TM and TE transmission results (transmission coefficient *vs*. wavelength) are shown in Figs. 2(A) and (B). Fig. 2 shows that a PBG exists for H-polarization which ranges from 9.38 to 9.42 nm and that for E-polarization ranges from 9.37 to 9.42 nm. An absolute photonic band gap is defined as the overlap region between the H- and E-polarization band gaps. Fig. 2 also shows clearly that the crystal thickness or the number of the layers included in the calculation influences the transmission coefficient. We conclude therefore that a well-defined PBG has been formed when the number of layers involved is greater than 128.

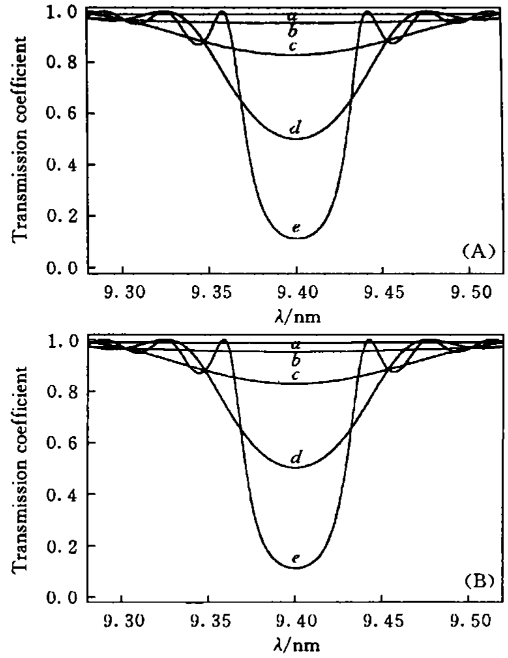

Fig. 2 Theoretical transmission coefficient *vs*. wavelength for H-polarization (A) and E-polarization (B) of MCM-41.

a. 8 Layers; *b*. 16 layers; *c*. 32 layers; *d*. 64 layers; *e*. 128 layers.

Fig. 3 shows the transmission coefficients for vector k (an incident wave vector) lying in the xz plane (off-plane propagation, meaning that the propagation is no longer along the z direction). In particular, a zero angle of incidence corresponds to a normal incidence where vector k is parallel to the z axis. As the angle increases, vector k tilts toward the x axis and the angle of incidence is measured between axis z and vector k. The transmission spectra for H-polarization is shown in Fig. 3(A). With an increased angle of incidence, both the lower and upper edges of the original band gap move toward higher frequencies, and both the width and depth of the gap are affected by the angle of incidence. This is caused by the increasingly compromised periodicity of the structure seen from the incident wave for the tilted incidence. When the angle of incidence increases up to about 35°, the original PBG disappears.

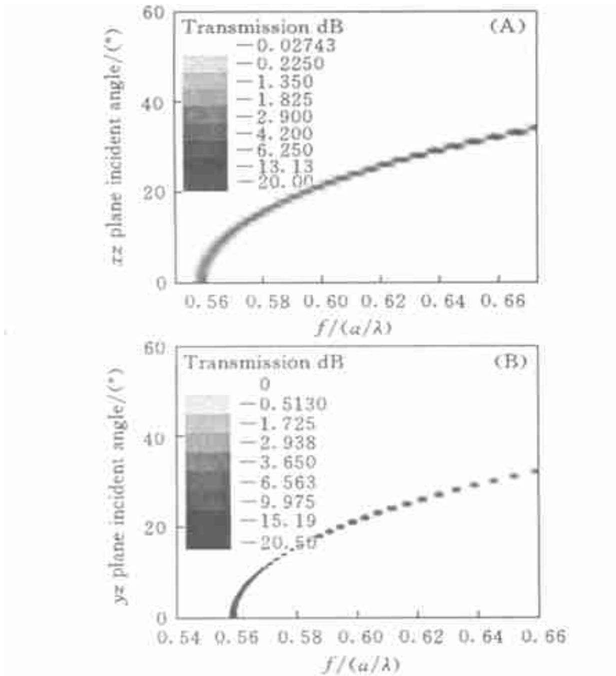

Fig. 3 Calculated in and out of plane angular response.
The TM transmission response for an angular scan within the periodic xz plane(A) and the yz plane(B).

For k in the yz plane[Fig. 3(B)], both the lower band-gap edge and the upper band-gap edge move to higher frequencies like a parabola with an angle of incidence. When the angle of incidence increases up to about 29°, the original PBG disappears.

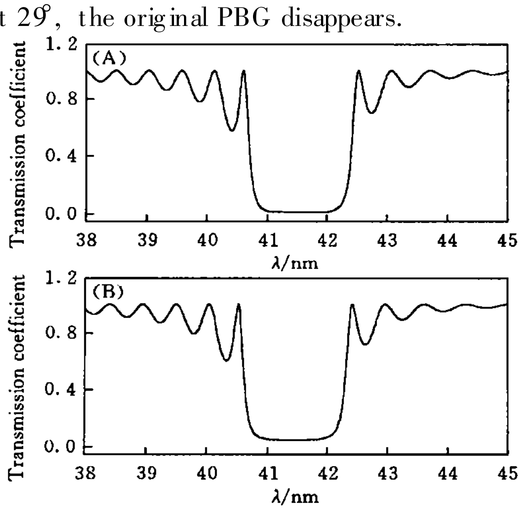

Fig. 4 Calculated transmission coefficients of SBA-15 for H polarization(A) and for E polarization(B).

We also calculated the TM and TE response of SBA-15, another well-ordered hexagonal mesoporous material, for normal incidence(k along z-direction). The results of SBA-15(Fig. 4) are obviously similar to those of MCM-41. The only two differences between them are the width and the position of the PBG. For SBA-15, the band-gap for 32 layers may be changed from 40.71 to 42.37 nm for H-polarization and from 40.62 to 42.31 nm for E-polarization. The pore size and the lattice constant are the two major reasons that result in the different PBG. Because the pore size and the lattice constant of these mesoporous silica can be controlled, the band-gap width can be also controlled when they are used as photonic crystals.

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