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The Compound Refractive Lens for Hard X-ray Focusing

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The compound refractive X-ray lens (CRL) for focusing hard X-rays is investigated to determine the parameters such as the focal length, the focal spot size, and spatial distribution at the focal spot using a simple theoretical calculations and CRLs fabricated by the self-assembly method. The number of individual compound lenses are defined for the given focal length of 1 m. The X-ray energy of 1 to 40 keV is used in the calculations. The CRL for focusing hard X-rays which generated from the X-ray tube is fabricated by nanoparticle-polymer composite in the form of circular concaves. The self-assembly method is applied to form the nanoaluminum-poly (methly methacrylate) composite and carbon-polymer composite CRL lenses. Aluminum nanoparticles of 100 nm and carbon microparticles are diffused in the polymer solution then the high gravity up to 6000G is applied in it to form the concave lens shape. X-ray energy at 8 keV is used for characterization of the composite CRLs. The FWHM of intensity for the fabricated nanoaluminium composite CRL system, $N=10$ is measured as 1.8 mm, which would give about 70 µm in FWHM at 1 m of the focal length.

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I. INTRODUCTION

The compound refractive X-ray lenses (CRL) have been developing for use on hard X-ray microscopy techniques to visualize the interior of opaque and live samples [1]. One of the major advantages of the techniques over other microscopy techniques is that the large penetration depth of hard X-rays allows one to image the interiors without opening the region of investigation. In order to realize an X-ray microscope in the normal laboratory, many important efforts have been attempted in the field of X-ray optical system. A number of different X-ray optical elements have been utilized such as Kirkpatrick mirrors [2], Fresnel or Bragg-Fresnel zone-plate [3], refractive lenses [4] and polycapillary [5] for generation of micro, or submicrometer size beam diameter.

High irradiance X-ray microbeam is requried for many experimental at techniques such as X-ray scanning microscopy [6], X-ray photon and fluorescence correlation spectroscopy [7], and scattering techniques [8] for the study of nanoparticles and bio-samples. Due to the requirement of high flux in X-ray microscopy and other X-ray measurements, development of X-ray optics system has been mainly related to synchrotron radiation sources until now.

In recent years, the compound refractive lenses, which are part of relatively new X-ray optical devices have demonstrated many advantages such as robustness and ease of alignment and operation [9]. One of the simple ways to utilize them is by drilling many holes along the optical axis. A lens with parabolic holes has lower aberration, large transmission and higher resolution have been reported [10]. Many approaches have been tried to fabricate nanocomposite materials especially metal-polymer systems. There methods include precursor techniques [11], intercalation polymerization techniques [12] and self-assembly [13].

Encapsulating polymerization [14] may be the most attractive technique to prepare polymer-nanometal-polymer composite as well as microcomposite. Nanoparticles are important material because their physical and chemical properties differ from those of the bulk phase. In general, the nanophase or nanolength scalar effect becomes strong starting from 100 nm and extending downward in size. The properties of polymers can be tailored for the purpose of engineering novel materials. Composites based on nanoparticles and polymers which modified their properties could offer unique X-ray optical properties. The fraction of particles which are nano- or molecular particles imbedded into organic polymer or resin matrix materials determines the properties of nanocomposites.

In this paper, the CRL parameters are investigated using simple calculations for focusing hard X-ray and for application of CRL to X-ray microscopy. We also demonstrate the feasibility of using the nanometal-polymer composite CRLs for X-ray focusing as well as utilizing them for X-ray microscopy.

II. METHODS AND MATERIALS

The refraction of X-ray beam is weak at the boundary of the surface and the absorption is strong when hard X-rays travel through the medium. The index of refraction for the spectrum window of X-rays in the medium is written as.

$$n = 1 - \delta + i\beta \tag{1}$$

where the dispersive decrement δ is of the order 10^{-5} – 10^{-7} and β is related to the linear absorption coefficient μ. Absorption is minimized by choosing low atomic weight elements such as beryllium (Be) and carbon (C) as lens material since the mass absorption coefficient decreases with the atomic number Z. Fig. 1 shows the dispersive decrement δ of the index of refraction for typical X-ray diffractive lens materials in terms of the X-ray energy [15]. It suggests that the refractive index is close to the unit at higher energy which means less refraction of X-ray beam at the interface of the lens. Beryllium, PMMA and carbon are currently used to fabricate the CRLs for hard X-ray focusing because of their lower absorption of X-ray compared with the conventional optical materials.

Since the refractive index is very small the bending and focusing of X-rays is difficult for a single X-ray lens. And the concave shape of lens focuses the X-ray as expected when the real part of the refractive index is smaller than 1. The small refraction is compensated by aligning many refraction lenses in a stack to use as an X-ray lens. It is known for a visible spectrum optical lens that the focal length of a series of N lenses is reduced by $1/N$. Then the radius of R for each lens of the compound refractive lenses which consists of N lenses has a focal length f is given;

$$f = \frac{R}{2\delta N} \tag{2}$$

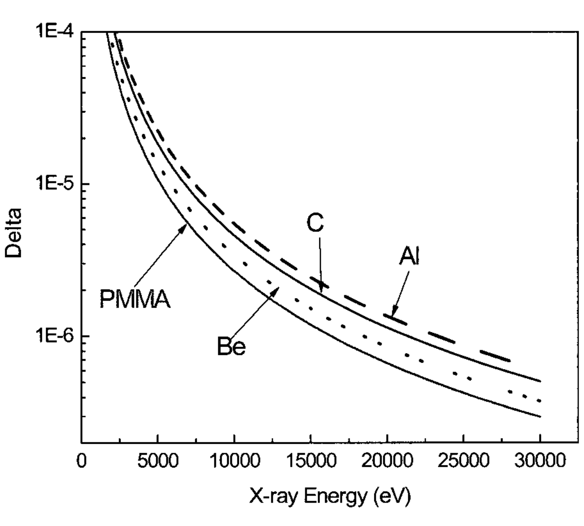

FIG. 1. The dispersive decrement δ in the index of refraction for typical CRL materials in terms of the X-ray energy.

Using the single lens focal length f_s, f also can written $f = f_s/N$. Fig. 2 is shown the schematic diagram of the CRL. The incident plane monochromatic X-ray wave with amplitude E_0 propagated along with the z-axis of a CRL. The CRL thickness d with the minimum thickness h_{min} at its center. Then the thickness of the CRL along the axial direction from the center $h(r)$ is approximated as $h(r) \approx r^2/2R + h_{\min}$ where R is radius of the CRL and r is radial axis.

The cross section of outgoing X-ray beam is spherical and focusing on its focal plane. The intensity distribution in an arbitrary plane located at z>>R is given by [16]

$$I(p) = I(0)\exp\left[-\frac{\pi}{\lambda R}\frac{(1-\delta)}{\beta}p^2\right] \tag{3}$$

And the resolution should be derived according to the Reyleigh critertion for Gaussian intensity distribution, $I(p)=I(0)\exp(-ap^2)$, numerical calculation gives in spherical type lens gives [17]

$$\Delta p = 1.88\sqrt{\frac{\lambda R}{\pi}}\frac{\beta}{(1-\delta)^2} \tag{4}$$

The sintering process, which is the most common technique for fabrication of beryllium X-ray lenses, would create voids in the lens. Beryllium is very brittle which makes it difficult to machine the CRL from the beryllium plate. And beryllium and many nano metal particles are toxic if inhaled. The importance of developing

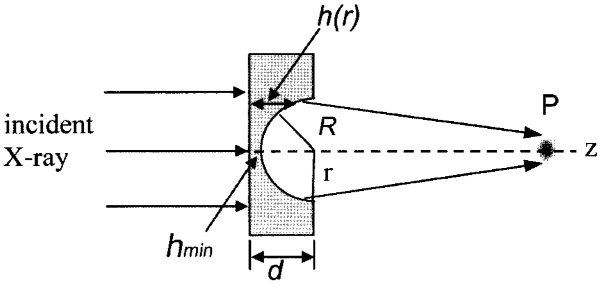

FIG. 2. A schematic of the compound refractive lens.

nanoscale engineering instruments has increased dramatically with the advance of material, synthesis and device processing capabilities. One of the advanced processes to utilize nanoparticles is a self-assembly process for fabrication of the nanometal-polymer composite. However, it is very difficult for metal or inorganic nanoparticles to disperse in the polymer matrix at the nanoscale through conventional mixing because nanoparticles have very high surface energy and will agglomerate during mixing.

A schematic diagram of the process of fabrication of nanometal-polymer composite CRL is shown in Fig. 3. The first step of the process is to avoid the agglomeration of the nanoparticles in the solution of polymer, the ultrasonic dispersion is applied into the mixture of the nanoparticles and polymer solution which is shown in Fig. 3 (a). Ultrasonic frequency of 20 kHz is used for an encapsulating process before the high-G process. Then high gravity force up to 6000 G is applied on the solution using a centrifuge. In this process the dispersing nano particles could be self-assembled on the surface of lower CRL mold by high gravity which is shown in Fig. 3 (b). The upper mold of CRL is inserted in the container in order to form the bi-concave shape lens which is shown in Fig. 3 (c). The thermal treatment of the self-assembled particles is for bonding the particles by polymerization. The thin layers of polymer at each surface of the nanoparticles are linked together by the thermal polymerization. Temperature of the vacuum oven is set to be 70℃ and the chamber of the oven is flushed by Ar gas then remains at 2×10^{-2} Torr during the process. Temperature is increased upto 90℃ over a 10 hour period for finalizing the polymerization. The nanometal-polymer composite CRLs are obtained after removing the molds from the container.

III. RESULTS AND DISCUSSION

Fig. 4 shows the relationship between the radius of CRL and the number of the compound lens. For a focal length $f = 1$ m, the radius of CRL for different X-ray energies as well as different materials have been determined. For example, 3 CRLs with 1×10^{-5} m radius are needed for focusing 20 keV X-rays at the focal point 1 m. However, 22 CRLs are needed for focusing 40 keV at the same focal point. It shows that a smaller CRL is needed for focusing higher X-ray energy at the focal point of 1 m when same number of CRLs is used. For 8 keV of X-ray energy, PMMA-(1), beryllium-(2), carbon-(3), and aluminium-(4) have the range of 100 μm to 300 μm in radius of CRL can be used for focusing. PMMA-(1) has to be smaller curvature than the other 3 materials (2)-(4) which are shown in Fig. 4 due to small value of the dispersive decrement δ. Beryllium which is the most transparent material for X-rays can be fabricated with slightly larger radius than PMMA for focusing at 1 m at the focal spot. The materials which have a higher value of the dispersive decrement δ such as carbon-(3) and alumimium-(4) can be used to obtain short focal length, in other words, less CRL units can be used for the given focal length. However, absorption is another factor that has to be considering for choosing CRL materials. The radius of CRLs are comparied for four different materials PMMA-(1), beryllium-(2), carbon-(3), and aluminium-(4) for X-ray energy of 8 keV in Fig. 4. It shows that aluminum CRL could be fabricated with larger curvature than carbon and beryllium as well as PMMA. In order to determine the radius of CRL and the number of individual CRL units in the X-ray optic system Eq. (2) can be used for focusing at the given focal plane.

The determination of the focal length of CRL is an important step for utilizing the CRL optic system on the laboratory scale because the index of refraction is closed to 1 in the X-ray region which means the angle of refraction is very small compared with the visible optic lens. Fig. 5 shows the focal length of CRL as a function of radius of CRL for X-ray energy at 8 keV, 20 keV, and 40 keV. In order to obtain the reasonable

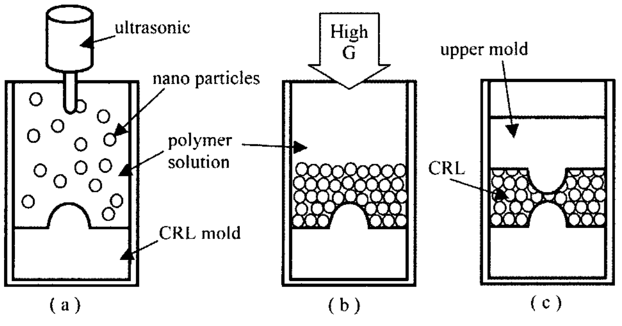

FIG. 3. A schematic diagram of the process of fabrication nanometal-polymer composite CRL.

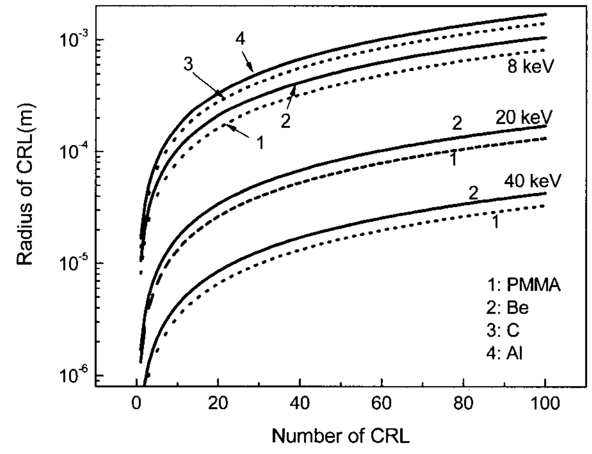

FIG. 4. The relation between the radius of curvature and number of CRL.

focal length for a normal laboratory, a CRL system which consist of 30 individual CRLs are considered in the calculation. Four different materials which are currently under development to fabricate CRL are considered, to determine the focal length for the given radius of CRL at 8 keV of X-ray energy. Of the order of millimeter in radius of the CRL system is used to focus X-ray of 8 keV at the focal length of meter dimension. The focal length reaches to a hundred meter range when incident beam of 40 keV is irradiated on the CRL system. Few tens of meter of focal length will be reached for 20 keV with 1 mm in radius of the CRL system. At the same X-ray energy, longer focal length is reached by the PMMA CRL than by the aluminium CRL system. Such tendency is shown at 8 keV (1)-(4) in the Fig. 5.

The X-ray intensity distribution at the focal spot as a function of X-ray energy and the number of individual CRLs are shown in Fig. 6. The dots lines in Fig. 6 represent the dependence of the number of individual CRL in optic system on the intensity distribution at the focal spot. The intensity distribution at the focal spot by 30 CRLs is 1.7 μm in FWHM, which is about 1/4 that of its single CRL. Then the solid lines in Fig. 6 represent the intensity profiles at the focal spot as a function of X-ray energies that show a tendency to focus at 8 keV and 20 keV. The FWHM of the intensity profiles by a single CRL are 6 μm at 8 keV and 8 μm at 20 keV.

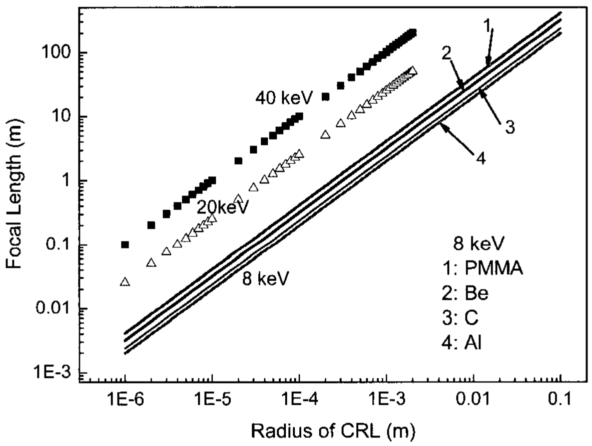

FIG. 5. The focal length of CRL as a function of radius of CRL.

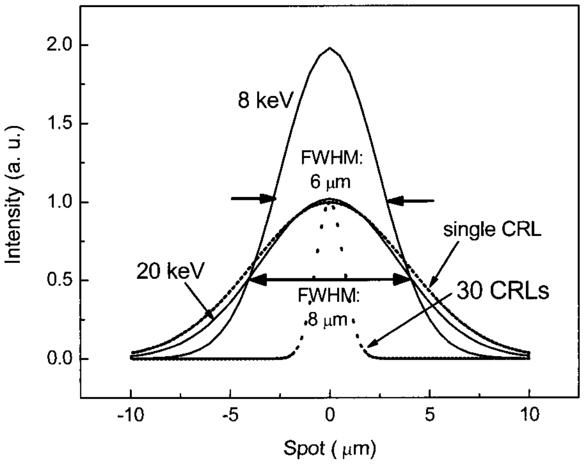

FIG. 6. The dependence of X-ray energy and the number of individual CRL unit on the intensity distribution at the focal spot.

Heat treated nanoparticles-polymer composite which is sandwiched between the CRL molds is solidified by means of polymerization. The solidified CRL may not need further polishing. The concave surface which is the actual working area as the X-ray lens has to be in good quality in the SEM image in Fig. 7. The influence of roughness on the CRL (r.m.s. roughness σ) of about 0.1 mm would not affect the performance of CRL. The fabricated CRLs in our laboratory are within that range. Micro Vickers hardness of the microstructured composites are measured as range of 20-30 Hv.

Fig. 8 shows the FWHMs as a function of the number of individual CRL of the nano aluminum-PMMA composite and carbon-PMMA composite CRL. X-ray energy of 8 keV is used which is generated from the Cu target X-ray tube (0.5 mA, 20.0 kV) in order to measure the FWHM of beam diameter. The fabricated CRLs with radius of 2 mm is mounted on the 5-axis translation stages. Incident X-ray is supplied through a colliminator of 2.0 mm I.D which is mounted on the X-ray tube unit. The CRL is placed 50 mm at front of the metal colliminator. Then the exit beam shapes are taken by the X-ray CCD which converts X-ray energy to visible light by a scintillator at the front of CCD. The FWHM of intensity is reduced to 25.0% of intensity without CRL by the10 individual aluminum-PMMA composite CRLs compare with without CRL and 13.0% reduced by the 10 carbon-PMMA CRLs. It can be predicted that 720 μm of FWHM will be obtained when the number of individual CRL, N=30 are included in a CRL system at 50 mm from the CRL. And the focal spot size will be down to about 70 μm at the focal length of 1 m. The insertion in Fig. 8 shows

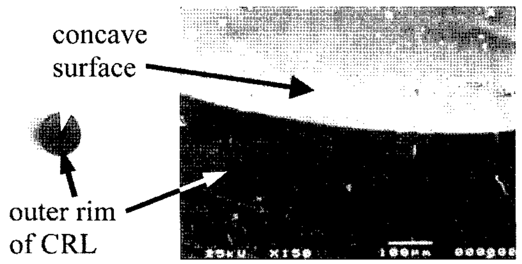

FIG. 7. The pictures of the fabricated CRL unit (a) and a SEM image of CRL (b).

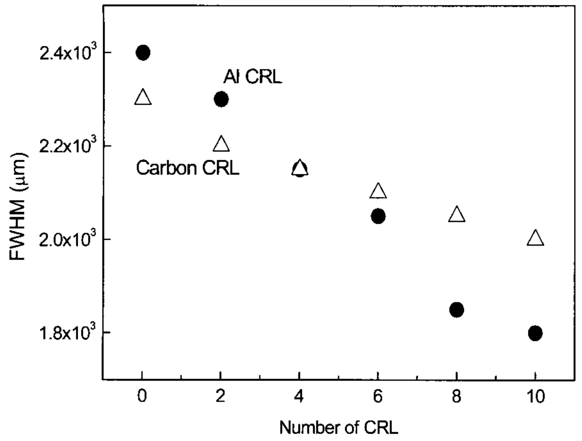

FIG. 8. The FWHM of the intensity as a function of the number of the individual CRL of aluminum- and carbon-PMMA composites.

the intensity reduction by increasing the length between the CRL exit and the CCD.

IV. CONCLUSIONS

The compound refractive X-ray lens (CRL) for focusing hard X-rays is investigated to determine the parameters such as the focal length, the focal spot size, and spatial distribution at the focal spot using simple theoretical calculations and fabricated CRLs by the self-assembly method. The energy of X-ray of 1 to 40 keV is used in the calculations.

The composite CRL system made of individual unit lenses of concave shape is demonstrated for focusing hard X-rays at 8 keV. CRL X-ray lenses are fabricated by nanoparticles-polymer composite. The method of direct self-assembly is applied to form the nanoaluminum-PMMA composite and carbon-PMMA. composite. Aluminum nano-particles of 100 nm and carbon microparticles of up to 10 μm in diameter are diffused in the polymer solution, then the high gravity of 6000 G is applied on the solution using a centrifuge. The nanoparticles in the polymer solution are self-assembled by the high gravity on the bottom mold during the process. Then the upper mold is inserted in the tube to form the biconcave lens shape in between the molds. The nanocomposite materials produce reduced scattering of the X-ray beam because the voids in the conventional sintered lens materials are eliminaited during the high gravity application process. The polymer chains which have nanoscale thickness are bonded to each particle and fill in the space between the particles. The FWHM of intensity for the fabricated nano-aluminium composite CRL system, N=10 is measured as 1.8 mm that would given about 70 μm in FWHM at 1 m of the focal length. The difference between the calculation and measurement of the spot size may be caused by the diverging X-ray source. A colliminated incident X-ray beam is assumed for calculations. Even though the metal colliminator is used in the measurement the exit beam diverges with angle of 3.57×10^{-3} degree.

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