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Fabrication of hard-coated optical absorbers with microstructured surfaces using etched ion tracks: Toward broadband ultra-low reflectance





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1. Introduction

Broadband ultra-low reflectance materials have attracted significant attention owing to their potential relevance in many applications, such as solar thermal energy converters [1,2], high efficiency solar cells [3,4], blackbody thermal radiators [5], optical absorbers for photo-thermal detectors [6–8], and stray light elimination [9]. Vertically aligned carbon nanotube (VACNT) black [8,10–13], black Si [1,4,14,15], gold black [16], and laser-ablated metal surfaces [2,17] exhibit low reflectance around or below 1% in the visible to near infrared (mid or far infrared in some cases) range of wavelengths.

Among these materials, VACNT blacks exhibit extremely low reflectance, below 0.1%, owing to their quite low density which makes the effective refractive index close to unity and suppresses surface reflection [18]. The low density, however, makes VACNT blacks too fragile and susceptible to dust blow off as well as to mechanical contact. Condensed carbon nanotubes are significantly more robust than as-grown VACNTs [19–22], while high-density carbon nanotube aggregates exhibit higher reflectance than

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ABSTRACT

Broadband low reflectance materials have various applications in the field of optical energy management; however, materials with ultra-low reflectance (below 0.1%) have been considered as mechanically delicate. We have developed a novel hard-surface optical absorber with microstructured, diamond-like carbon coated ion tracks on CR-39 plastic substrate. The spectral reflectance of the first prototype was below 2% for wavelengths ranging from 400 nm to 1400 nm; moreover, the optical absorber had mechanically hard surface and exhibited temporal durability. Choosing the appropriate design of the surface structure and coating layer is likely to reduce the reflectance to the order of 0.1%. This technique yields easy-to-handle broadband ultra-low reflectance absorbers.

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VACNT blacks [23]. Achieving both ultra-low reflectance and mechanical stability is a critical issue, affecting the practical usability of this material for broad applications.

Recently, the authors have investigated the mechanism of ultra-low reflectance from nickel phosphorous (NiP) black surface [24], which is one of the darkest materials in the visible to near infrared range of wavelengths [25,26]. NiP black surfaces have microporous morphology of conically shaped pits with an optical absorption layer of oxidized nickel on them, where the incident light experiences multiple reflections, thus effectively enhancing the light absorption. By using numerical simulations, it was determined that the crucial parameters for ultra-low reflectance were the high aspect ratio of the conically shaped pits and the thickness of the optical absorption layer [24]. However, NiP black surfaces are also delicate and customization of their mechanical properties is not easy to achieve [27].

In this paper, we demonstrate, for the first time, a hard-coated low-reflectance optical absorber with microstructured surface, which is inspired by the NiP black surface. In our method, etched ion tracks on CR-39 plastics [28,29] were used for patterning the surface with conically shaped pits of a high aspect ratio, and diamond-like carbon (DLC) [30,31] was deposited on the surface to serve as a hard optical absorption layer. Here, we show the results of spectral reflectance measurements of the first prototype of the proposed optical absorber, and discuss the feasibility of broadband ultra-low reflectance with durability for easy handling.

2. Materials and methods

2.1. Design and fabrication of the microstructured optical absorber

For designing the DLC-coated microstructured optical absorber, the geometrical parameters of NiP black surface serve as a good example because DLC (a-C:H) has a complex refractive index [32] similar to that of oxidized nickel (NiO_{1+x}) [33]. Fig. 1 shows the geometrical model of the surface of the novel microstructured optical absorber with DLC black layer, considered in this study. In the case of NiP black, the preferred pit aspect ratio (defined as d/r and shown in Fig. 1(b)) should be equal to or above 3, and the pit diameter should be larger than the incident light wavelength, for obtaining ultra-low reflectance below 0.1% [24].

To fabricate the pits with such a large aspect ratio on CR-39 substrates (BARYOTRAK, Fukuvi Chemical Industry Co., Ltd., Japan), we performed heavy ion beam irradiation with azimuthally varying field (AVF) cyclotron at the Takasaki Ion Accelerators for Advanced Radiation Application (TIARA), Japan Atomic Energy Agency (JAEA). This ion beam fabrication technique allows easily obtaining the etch pits with desired aspect ratio and surface density appropriate for target reflectance and wavelength. These pits are randomly distributed favorably for preventing the resonant reflection arising owing to grating-like periodical patterns. The fabrication procedures are schematically summarized in Fig. 2. In this first demonstration, we used a 335 MeV oxygen ion beam, for the following reasons. The calculation code of the stopping and range of ions in matter (SRIM) [34] suggest sufficient penetration range of >1 mm in CR-39 plastics; thus, the \sim 0.8-mm-thick CR-39 substrates that we used can be penetrated. The beam also exhibits a large linear energy transfer (LET) of 300 keV/µm (in water) on the backside of the CR-39, whose ion tracks are expected to be etched into pits with a high aspect ratio of ~ 5 [29,34]. A long penetration range and large LET of the incident ion beam enabled us to simultaneously achieve large opening diameters (above several tens of microns) and a high aspect ratio. Such a large opening texture provides lower reflectance even in the longer wavelength range, such as in mid infrared [24]. The irradiation densities were



Fig. 1. (a) Three-dimensional surface geometrical model of the novel microstructured optical absorber with a DLC (a-C:H) black layer developed in this study. (b) The cross-sectional view of a single unit cell of (a).



Fig. 2. Schematic of the fabrication procedures of the microstructured optical absorber in this study.

 $1\times 10^6\,ions/cm^2$ and $1.2\times 10^5\,ions/cm^2$, corresponding to the averaged area per ion track of 100 μm^2 (for the opening diameter of 11 μm) and 833 μm^2 (for the opening diameter of 33 μm), respectively.

After the ion beam irradiation, the CR-39 substrate was etched in 6.5 N NaOH solution at 70 °C for 12–36 h, to ensure that the etch pits filled the entire surface. Afterwards, the etched CR-39 surface was coated with a 100-nm-thick layer of Ni and a 50-nm-thick underlayer of Cr by ion plating [35] to prevent the transmission of light.

The micropatterned substrates were then coated with DLC (a-C:H) by plasma-based ion implantation and deposition (PBIID) [30,31]. The target thickness *l* was about 5 μ m, which is slightly larger than the oxidized nickel layer's thickness in NiP black [26]. A flat CR-39 plate with pre-coated Ni/Cr was also coated with DLC and was used as a control sample.

2.2. Evaluation of the fabricated microstructured optical absorber

The hemispherical spectral reflectance of the developed optical absorbers was measured by using a LAMBDA 900

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