X-ray radiographic technique for measuring density uniformity of silica aerogel

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A B S T R A C T

This paper proposes a new X-ray radiographic technique for measuring density uniformity of silica aerogels used as radiator in proximity-focusing ring-imaging Cherenkov detectors. To obtain high performance in a large-area detector, a key characteristic of radiator is the density (i.e. refractive index) uniformity of an individual aerogel monolith. At a refractive index of $n=1.05$, our requirement for the refractive index uniformity in the transverse plane direction of an aerogel tile is $|\delta(n-1)/(n-1)|<0.04$% in a focusing dual layer radiator (with different refractive indices) scheme. We applied the radiographic technique to evaluate the density uniformity of our original aerogels from a trial production and that of Panasonic products (SP-50) as a reference, and to confirm they have sufficient density uniformity within $\pm 1.0$% along the transverse plane direction. The measurement results show that the proposed technique can quantitatively estimate the density uniformity of aerogels.

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1. Introduction

In high-energy and nuclear experiments, silica aerogel, which is a colloidal form of quartz ($\text{SiO}_2$), is important as a radiator in Cherenkov counters. This is because this transparent solid has a tunable refractive index ($n$) and it is easier to handle than liquid or gas radiators. Hydrophobic aerogels with $n=1.066-1.14$ can be consistently manufactured by our conventional method [1]. Furthermore, by using a new method (the pin-drying method), we can produce ultrahigh-refractive-index ($n=1.10-1.26$) aerogels with sufficient transparency [2]. The recent progress toward developing highly transparent aerogels with $n=1.04-1.26$ [3] will open a new window to particle identification (PID) in future experiments, for example, the super $B$-factory experiment at KEK (Belle II experiment [4]) and nuclear experiments proposed at J-PARC.

Our aerogel development is mainly motivated by the Belle detector upgrade program, including the PID device upgrade. For the new PID device to separate kaons from pions in the momentum range around 1–4 GeV/c in the forward end-cap of the Belle II detector, the A-RICH group is developing a proximity-focusing aerogel ring-imaging Cherenkov (A-RICH) detector [5]. In a dual layer radiator scheme (focusing combination) [6], the use of approximately 250 large-area aerogel tiles with $n\sim 1.05$ and dimensions $18\times 24\times 3\text{cm}^3$ is planned. Section 2 describes the maximum allowed refractive index variation over a large transversal area that meets our requirement to ensure a constant-mean single-photon Cherenkov angle distribution.

On the basis of scanning electron microscopy, the typical diameter scale of the silica primary particles and structural pores in aerogels is known to be of the order of 10 nm. In the micrometre range, aerogels are considered to be sufficiently uniform for optical use. However, it is not self-evident that our aerogels are uniform enough in a macroscopic range from millimeters to centimeters. Local non-uniformities might be introduced during all the phases of aerogel production (i.e. wet-gel synthesis, ageing, hydrophobic treatment and supercritical drying). Therefore, in this paper, we propose a method to evaluate the density uniformity of an aerogel monolith using X-rays, and we demonstrate the validity of the method for our standard aerogels as a scale model of the A-RICH radiator.

2. Importance of refractive index uniformity

2.1. Influence of refractive index uniformity on RICH detectors

In a RICH detector, it is crucial to accurately determine the refractive index over the entire tile of an individual aerogel...
because the single-photon Cherenkov angle resolution, in addition to the mean of the single-photon Cherenkov angle distribution, is affected by the effective local refractive index of the radiator. The refractive index uniformity of an individual aerogel monolith is generally a key property of a good Cherenkov radiator, similar to aerogel transparency, because of the impact on detector performance. The refractive index uniformity can be studied along the thickness and transverse plane directions.

Uniformity along the thickness direction is more important because non-uniformities together with chromatic dispersion of the refractive index would directly worsen the Cherenkov angle resolution. In the dual layer radiator scheme, the aerogel of each layer is expected to be sufficiently uniform or to have a limited refractive index gradient [7] along the thickness direction. In previous beam tests [6,8,9], the prototype proximity-focusing RICH counters were composed of two radiator tiles each 20 mm thick and a photon detector array parallel to the radiator face with an expansion distance of 20 cm. As a rule, charged particles impacted on a limited area at the center of the aerogel tiles. Therefore, uniformity in the thickness direction, rather than in the transverse plane direction, contributes most to the Cherenkov angle resolution. With this configuration, the Cherenkov angle resolution \( \sigma_\theta \approx 14 \text{ mrad} \) was obtained. The result agrees with those of an analytical model [10] assuming \( \sigma_{\text{rest}} = 6 \text{ mrad} \), where \( \sigma_{\text{rest}} \) is the contribution to the error in determination of the Cherenkov angle from sources other than the emission point uncertainty and due to the position resolution of the detector (see Ref. [10] for more detail). This contribution from other sources may be partially explained by non-uniformity along the thickness direction. This suggests a fruitful direction for future investigation.

As a first step, we now focus on uniformity in the transverse direction, which is the main subject of this study. The measurement of the refractive index uniformity in the transverse plane direction is mandatory for the A-RICH detector because it has a large area of 3.5 m². To guarantee a constant-mean single-photon Cherenkov angle distribution over an entire aerogel tile, it is crucial to evaluate uniformity along the transverse plane direction. Our final goal is to measure the uniformity of large-area aerogel tiles with dimensions of \( 18 \times 18 \times 2 \text{ cm}^3 \) produced by both conventional and pin-drying methods. Measurements of uniformity will provide a comprehensive evaluation that includes the contribution of uniformity in the thickness direction. The maximum tolerance of the refractive index non-uniformity is \( |\delta(n-1)/(n-1)| < 4\% \), which corresponds to \( n = 1.05 \pm 0.002 \). This acceptable variation is based on the model calculation in Ref. [10], where the degradation of the Cherenkov angle resolution in the dual layer radiator scheme is very small. This was empirically verified in a dedicated beam test [11].

2.2. Previous methods

A direct method for measuring the refractive index uniformity of aerogels by a laser (the gradient method) was first introduced in Ref. [12], and was also applied in Refs. [13–15]. This method is based on the deviation of the laser due to the transverse gradient of the refractive index along its optical path. If aerogels have a variation in thickness or a bend (i.e. surface tilt or meniscus effects), this method will not be suitable for measuring the refractive index uniformity because the tilted surface refracts the laser even if the aerogel is sufficiently uniform. In Ref. [15], the thickness variation within an aerogel was mapped using a mechanical comparator, and the result of the gradient method was corrected by the effects of thickness variation. The thickness variation will affect the optical characteristics of aerogels and the RICH detector performance. In Ref. [16], the thickness variation was evaluated using a laser-based sensor to investigate aerogels for use in a RICH radiator.

Another method for evaluating the refractive index uniformity of aerogels by X-rays was applied in Ref. [17]. The first study in which X-rays were used to examine density variations in aerogels for use in a RICH radiator was presented in Ref. [18] using a digital radiographic device [19]. In Ref. [18], X-ray images of an aerogel block (top and side views) were used. A multiple-layer aerogel block was also measured by X-rays in a later study [20].

Cherenkov imaging using a RICH detector is the most thorough method for evaluating the local refractive index of aerogels in a beam test. In Ref. [21], the variation in the Cherenkov angle over an aerogel surface was measured by focusing the beam impact position on the aerogel; however, the measurement area was limited to 36 segments of \( 5 \times 5 \text{ mm}^2 \) each.

2.3. Proposal for novel X-ray radiographic technique

Here we present an X-ray radiographic technique as another method to investigate the density uniformity within an individual aerogel monolith. In this technique, X-ray absorption in the aerogel material is measured more quantitatively. By conducting an experimental procedure as described in the next section, we can determine the absolute density of local areas of aerogels. The refractive index of aerogels is proportional to their density \( \rho \), \( n-1 = k \rho \), where \( k \) is a wavelength-dependent coefficient. This is an empirical (Lorentz–Lorenz based [22]) relationship [1]. Measuring the density uniformity is thus a means of evaluating the refractive index uniformity of an individual aerogel tile. A characteristic of our method is that it measures the aerogel thickness as accurately as possible. Thickness measurements have important implications for the quantitative evaluation of density of aerogels because the X-ray absorption is sensitive to changes in their thickness. Thus, we expose the cross-sectional surface by cutting aerogels in order to measure the thickness. The potential disadvantage is that our technique destroys the aerogels. However, by measuring the aerogel thickness precisely, our X-ray radiographic technique has an important advantage because the measurement of the density of aerogels does not depend on the particular condition of their surface. A preliminary experimental result for our aerogel using the X-ray technique was briefly reported in Ref. [23]. In the following pages, a complete investigation of the density uniformity of basic (conventional) aerogels with small dimensions is reported. For novel aerogels produced by the pin-drying method, the results of characterization studies are presented in a separate paper [24].

3. X-ray radiographic technique

3.1. Measurement concept

X-ray absorption by materials obeys the exponential attenuation law

\[
I/I_0 = \exp(-\mu x)
\]

(1)

where \( I_0 \) is the incident X-ray intensity, \( I \) is the transmitted X-ray intensity, \( \mu_m \) is the X-ray mass absorption coefficient and \( x \) is the mass thickness of the material. This equation is valid for a narrow monoenergetic X-ray beam. The mass thickness is defined as

\[
x = \rho t
\]

(2)

where \( t \) is the thickness of the material. The photon mass absorption coefficients for elements are available as a function of the photon energy from the database in Ref. [25]. We found that the material density is reproduced well when we use the