Production of a New Generation of Silica Aerogel and its Application for the KEK B-factory Experiment

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Abstract

Low-refractive index silica aerogel is the most convenient radiator for threshold-type Cherenkov counters, which is used for particle identification in high-energy physics experiments. For the BELLE detector at the KEK B-Factory, we have produced about 2 m^3 of hydrophobic silica aerogels of n = 1.01 - 1.03 using a new production method. The particle identification capability of the aerogel Cherenkov counters was tested and 3 σ pion/proton separation has been achieved at 3.5 GeV/c. Radiation hardness of the aerogels was confirmed up to 9.8 Mrad.

Thanks to the improved transparency, aerogels prepared by the two-step method can be used as radiators for not only threshold-type, but also for Ring Imaging-type Cherenkov counters. The newly developed aerogels will be used extensively for particle identification devices in future high-energy physics experiments.

1. Introduction

In high-energy physics experiments, the identification of particles is very important in the understanding of detailed dynamics of underlying interactions. One method for such particle identification involves detection of the Cherenkov light emitted by charged particles passing through transparent materials. This light is produced only when the velocity of the particle is faster than the velocity of light in the material, this condition is expressed as:

$$n\beta > 1, \tag{1}$$

where n is the refractive index of the material, and β is the velocity of the charged particle in units of c (velocity of light in vacuum). Figure 1 shows $n\beta = 1$ lines for pions and kaons. In this figure, condition (1) is satisfied for the regions above each line; hence, the hatched area is the region where pions emit Cherenkov light and kaons do not. Thus, if we select a material with n = 1.01, then only pions will emit Cherenkov light in a momentum range between 0.98 GeV/c and 3.48 GeV/c. The figure indicates that a material having a refractive index of less than 1.03 is indispensable for pion/kaon separation, at a few GeV/c, with a threshold-type Cherenkov detector. However, it is very difficult to attain such a low refractive index with most materials. The only candidates other than silica aerogels are liquid Helium (n = 1.024) or pressurized CO_2 (n = 1.02

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at 25MP), neither of which allows for easy handling. For this reason, a large number of experiments [1] have used silica aerogels as the radiator for threshold-type Cherenkov counters. The first successful demonstration of this technique was by *Cartin et al.* in 1974 [2]. Before this experiment, a fine grained silica powder, mixed with air, was tried to achieve a low-refractive index material [3]; however, the number of observed Cherenkov photons was quite small compared with later silica aerogel experiments.

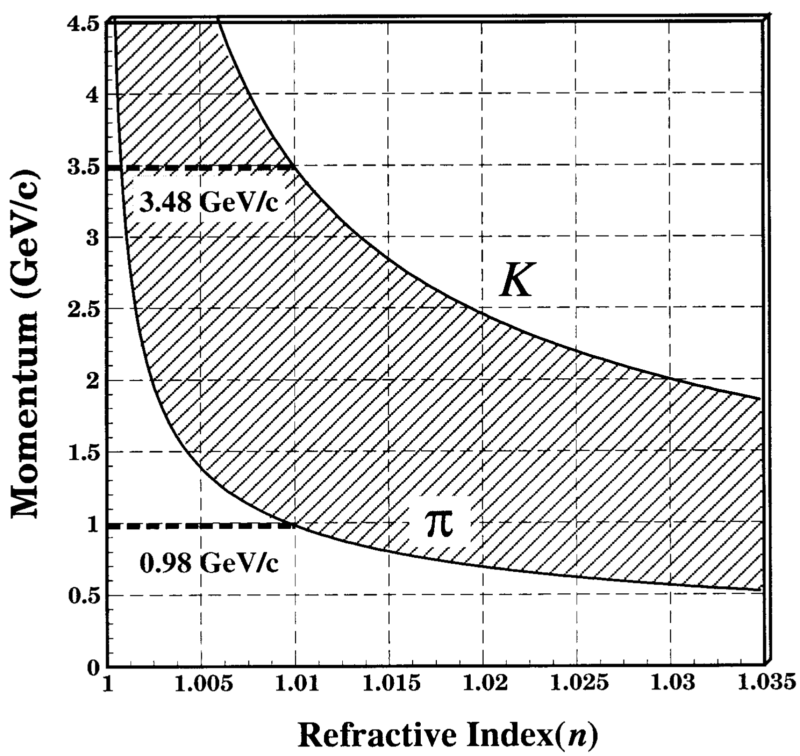

Fig. 1 In the region above each curved line ($n\beta = 1$), particles (pions or kaons) emit Cherenkov light. The hatched area is the region where pions emit Cherenkov light and kaons do not.

Before the invention of the two-step method [4], it was very difficult to produce silica aerogels having a refractive index below 1.02. This restricted the particle identification capability to low momentum regions ($p < 2.5$ GeV/c). Recent *B*-physics experiments, however, require good particle identification on the momentum range of up to 4 GeV/c, and low-refractive index material ($n = 1.007$ to 1.03) becomes vital for such experiments. Thanks to the invention of the two-step method, silica aerogel with low-refractive index can be easily produced. We have produced about 2.0 m^3 of silica aerogel ($n = 1.01$ to 1.03) for the KEK B-factory experiment [5] in a cooperative work with Matsushita Electric Works, Ltd. and KEK.

Here, we report details of our silica aerogel counter, which will be used for the KEK B-Factory experiment. Section 2 provides outline of the silica aerogel application of in the BELLE detector; Section 3 describes the production methods and the quality of the aerogel produced; Section 4 evaluates its performance. Other applications for high-energy experiments are also briefly discussed.

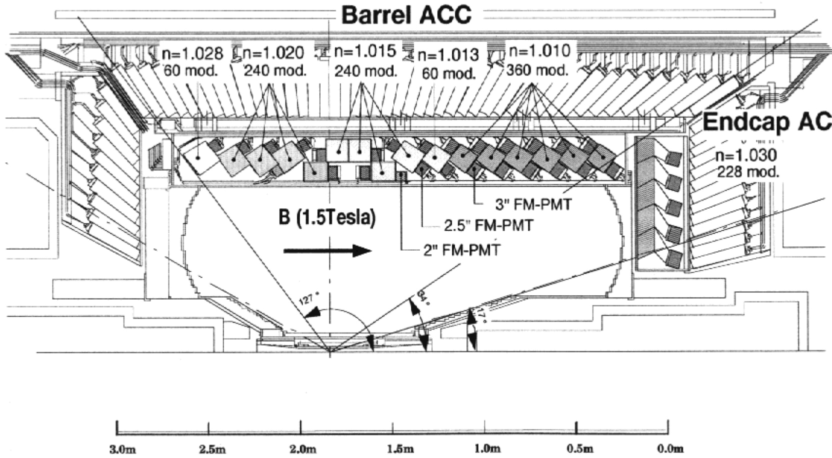

Fig. 2. The arrangement of the ACC at the center of the BELLE detector.

2. Silica Aerogel for the BELLE Detector

One of the most intriguing puzzles of nature is that the universe is composed of only matter, which contradicts cosmological theories, suggesting that an equal amount of particles and antiparticles have been produced in the Big Bang. A simple explanation of this phenomenon requires the violation of matter-antimatter symmetry (so-called CP symmetry), which has so far been only observed in the kaon system [6]. The K-M scheme [7] of the Standard Model, which is the most successful theory in particle physics, predicts a large CP asymmetry in the B-meson system [8]. In order to elucidate this interesting physics, several B-factories have been proposed and are being constructed around the world at such places as CESR (Cornell) [9], SLAC-B (Stanford) [10], KEK-B (KEK), HERA-B (DESY) [11], and LHC-B (CERN) [12], where large numbers (~10^{7-8}/year) of B-meson decays will be examined for the study of CP violation among other things. In such B-factories, separation of pions from kaons is vital for the identification of *B* or *B* mesons and the selection of rare decays. The BELLE group (the experimental group for KEK B-Factory) has decided to use an array of silica aerogel Cherenkov counters (ACC) for such particle identification. Figure 2 shows the configuration of the ACC in a central part of the BELLE detector. The ACC consists of 960 counter modules for the barrel part and 228 modules for the end-cap part of the detector. In order to obtain good pion/kaon separation for the whole kinematics range, the refractive indices of aerogels are selected to be between 1.01 and 1.03, depending on their polar angle region. A view of a typical single ACC module is shown in Fig. 3.

Five aerogel tiles are stacked in a thin (0.2mm thickness) aluminum box ($12x12x12\ cm^3$). In order to effectively detect the Cherenkov light, two fine mesh-type photomultiplier tubes (PMT's) [13], which operate in a magnetic field of 1.5 T, are attached directly to the aerogels at both sides of the box. We use PMT's of three different diameters: 3" (R6683), 2.5" (R6682), and 2" (R6681) for silica aerogel of n =1.01, 1.015 and 1.02, respectively. The inner surface of the box (except for the phototube windows) is lined with a diffuse reflector (Gortex sheet [14]) to obtain a uniform response. The total volume of aerogel needed for the ACC is about 2 m^3. For the study of CP violation, the ACC is required to distinguish pions and kaons with a resolution better than 3σ.

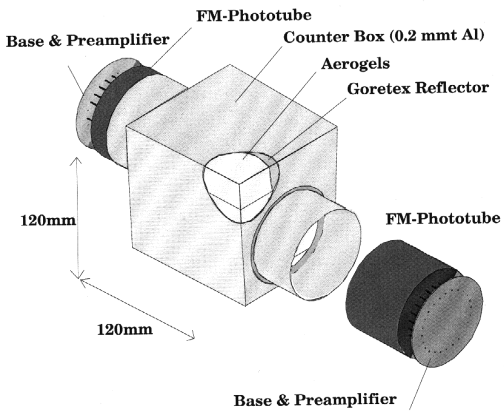

Fig. 3. Schematic drawing of a typical single ACC module: five aerogel tiles are contained in a thin aluminum box lined with a white reflector (Gortex sheet). They are viewed by two fine mesh PMT's.

3. Production of Silica Aerogels

3.1. Preparation of Alcogels

Silica aerogels are conventionally produced by the so-called "sol-gel" process, in which the gels, "alcogels", leading to high-porosity aerogels, are made from the following two reactions (hydrolysis and condensation):

$$nSi(OR)_4 + 4nH_2O \quad \text{----->}\ nSi(OH)_4 + 4nROH \quad \text{(hydrolysis)},$$

$$nSi(OH)_4 \quad \text{----->}\ nSiO_2 + 2nH_2O \quad \text{(condensation)},$$

where OR represents an alkoxyl group such as CH_3O.

These two reactions take place simultaneously in an alcohol solvent with an acid or base catalyst. Then geletion proceeds and syneresis continues to strengthen the structure. Since refractive index of aerogel (n), which is proportional to its density (ρ: g/cc) as $n - 1 = 0.21\ \rho$, is determined by the ratio of SiO_2 and the alcohol. The amount of mixed alcohol can control the refractive index. Hence, if one wants to make a low refractive index aerogel, one needs to add larger amounts of alcohol; however, that induces the reverse reaction of hydrolysis and gelation is prevented. This is why the production of a low-refractive index aerogel with good transparency was difficult.

In order to surmount this problem, a two-step method was proposed by *R. Hrubesh et al.* During the first step, less water then required, to complete the hydrolysis, is used to obtain partially hydrolyzed silica precursor. For instance, the complete hydrolysis of one mol of tetra-alkoxysilane requires 4 mol of water; however, they only added 1.3 mol of water. Later on, all the alcohol is distilled off, and the remaining silica precursor is diluted by a solvent without alcohol, such as acetonitril. In the second-step, the silica oil thus produced is further processed to make a gel by adding extra water with a base catalyst. By adopting such a procedure, the reverse reaction of hydrolysis is inhibited, and we can make a silica aerogel with porosity as high as 99.9% or more. The silica aerogel thus produced has different microstructure from that of conventionally produced ones, namely having much smaller pore size.

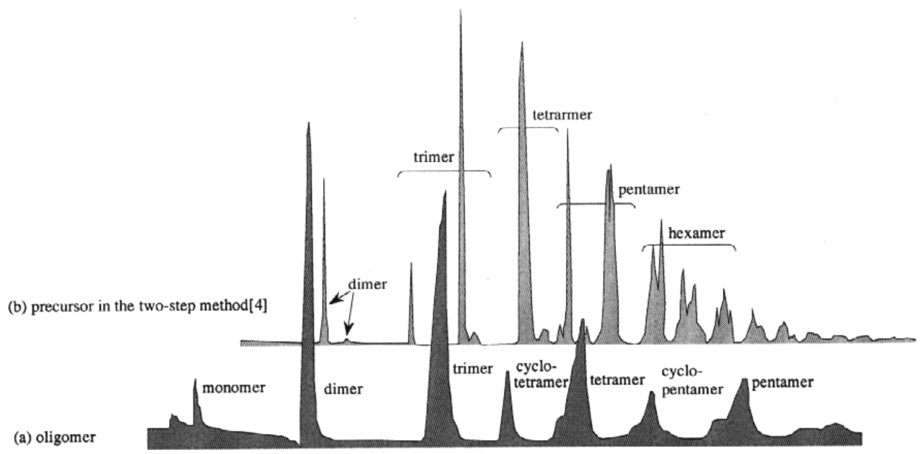

Fig. 4. Charts for the (a) oligomer and the (b) precursor measured by a gas chromatograph mass spectrometer. The horizontal and vertical directions correspond to the response of time and the content of each composition, respectively.

We adopted a modified two-step method for the preparation of alcogels, in which we used a methylalkoxide oligomer as the precursor instead of using the partially hydrolyzed silica precursor. This oligomer contains 51% silica (SiO_2) in percentage weight and is commercially available ("Methylsilicates-51" by Colcoat Co., Ltd.). Using a gas chromatograph and mass spectrometer, the composition of this oligomer was compared with the precursor that is used in the standard two-step method, and was found to be very similar to the latter, as shown in Fig. 4. This oligomer is hydrolyzed and polymerized in a basic catalyst (NH_4OH) with a solution of methanol or ethanol. Although this method is not exactly the same, we can take the advantages of the standard two-step methods and can produce a low-refractive index aerogel. This method is suitable for mass production of aerogel, since we can eliminate the complicated process in the first step of the standard two-step method: alcohol distilled off of and diluted with acetonitril, etc.

The average size of the alcogels that we produced are 120 x 120 x 24 mm^3. They are formed in aluminum molds coated with a thin PTFE film. The molar mixing ratios of the oligomer, methanol (ethanol), water, and ammonia are optimized so that the transmittance of dried aerogels are as high as possible. The molar mixing ratios of the materials are summarized in Table 1. Typical gelation time ranges from a few minutes to 10 minutes depending on the densities. For aerogels of density greater than 0.1g/cc, we adopted methanol for the solution, instead of ethanol, for better transparency.

Table 1. The molar mixing ratios for the preparation of alcogels of four different indices (n = 1.01, 1.015, 1.02, and 1.028). Here, we assume that the molecular weight of MS51 is 470. We used methanol as the solution for n = 1.028 alcogels.

Refractive index	MS51	ethanol	water	NH_3
1.01	1	139.7	18.25	2.466
1.015	1	89.66	21.57	0.8324
1.02	1	57.56	25.48	0.2810
1.028	1	56.95 (methanol)	25.06	0.1578

3.2. Surface Modification for a Hydrophobic Property

Silica aerogels have been used in several experiments; however, their transparencies became worse within a few years of use [15]. This phenomenon may be attributed to the hydrophilic property of the silica aerogels. In order to prevent such effects, we have made our silica aerogels highly hydrophobic by changing the surface hydroxyl groups into trimethylsilyl groups [16]. This modification can be done at any one of the three stages: before the drying, while drying, or after the drying; we adopted the first option for simplicity. After one week of aging, the alcogels are submerged for three days into a solution, which is a mixture of hexamethyldisilazane and ethanol with a volume ratio of 1:8. As a result of this treatment, our silica aerogels are still as transparent as they were when they were produced about two and a half years ago. Since such processed aerogels are highly hydrophobic, these aerogels are easily shaped into any form by using a water-jet cutter without any damage to the transparency.

3.3. Drying

After three weeks of aging, including the surface modification, the alcogels are dried with CO_2. Figure 5 shows a flow sheet of our drying facility. The autoclave volume is 140 litres (550 mm in diameter and 590 mm in height), and can produce about 38 litres of silica aerogel in one batch. After seven months of operation (two batches/week), we have completed the production of about 2 m^3 of silica aerogels.

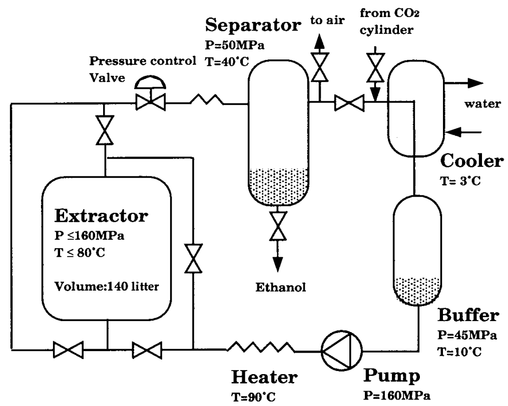

Fig. 5. Flow sheet of the extraction facility at KEK for the production of silica aerogel tiles by super critical CO_2.

The standard process pressure and temperature are shown in Fig. 6. The substitution of carbon dioxide for ethanol was performed at super critical condition of CO_2–ethanol system, which is realized, for example, at a pressure of $p > 9.35$ MP and temperature T = 325.2K [17]. This substitution takes 14 hours, and the extraction of alcohol proceeds at 15.8 MP and 80°C for 20 hours. Confirming that the ethanol composition in CO_2 is less than 0.3% in the extractor, the pressure of the extractor is gradually decreased to atmospheric pressure in 10 hours. Thus the whole process takes 47 hours. By employing the super-critical substitution, the probability of having cracks in the aerogels is greatly reduced and almost 100% of aerogels have no cracks if the thickness is less than 20 mm.

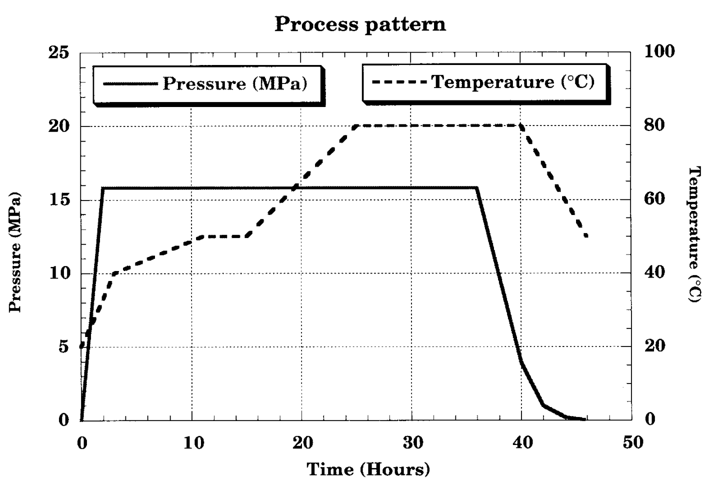

Fig. 6. Process sequence for the pressure and temperature of the CO_2 substitution and drying. The CO_2 substitution is proceeded in a super critical condition of the CO_2–ethanol system.

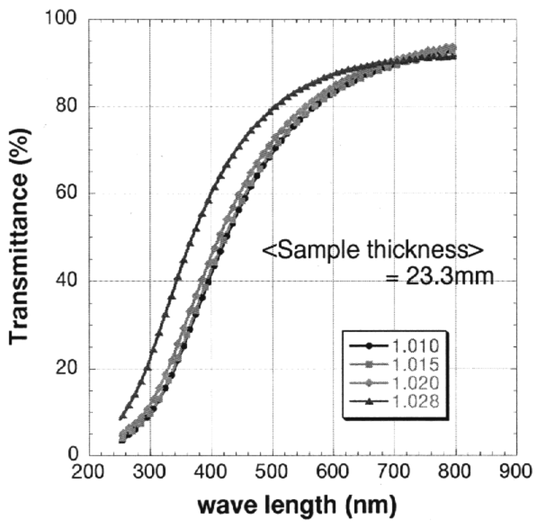

Fig. 7. Light transmittance spectra for the silica aerogels (thickness = 24 mm) of n = 1.01, 1.015, 1.02 and 1.028. The silica aerogels of n = 1.028, which were prepared by using methanol as the solution, have shown best transmittance.

3.4. Quality

All the aerogel tiles thus produced have been checked for optical transparency, refractive index, density, size, etc. Figure 7 shows a typical transmittance curve obtained by a photo-spectrometer [18] for aerogels of four different refractive indices. The n = 1.028 aerogels have better transmittance than other ones. Their average transmission length (Λ) at 400nm is 46mm, while others are around 25mm. Here, we define Λ as: $T/T_0 = \exp(-d/\Lambda)$, where T/T_0 is the transmittance, and d is the thickness of the aerogels. These aerogels were produced from the alcogel, which was prepared by using methanol as the solution.

The refractive indices are well controlled at $\Delta n/(n\text{-}1) \sim 3\%$, which is almost the same as the measurement error. The surface treatment for attaining hydrophobic property also helps to reduce the shrinkage after drying. Even at n = 1.01 aerogels (density $\rho \sim 0.04$g/cc), the volume shrinkage was only 4%.

3.5. Radiation Hardness

Detectors will be subject to a high radiation environment in future high-energy experiments to be carried out at high beam intensity machines. Hence, the radiation hardness of the detectors is a critical issue for such experiments. Radiation damage implies, for instance, that transparent glasses will turn brown by forming color centers after irradiation. At hadron machines such as HERA-B, a radiation dose of 10 Mrad/yr. is expected in the vicinity of the beam pipe.

For this reason, we carried out a test to ensure the radiation hardness of aerogels by placing them in high intensity γ rays from a ^{60}Co source at the irradiation facility in the National Tsing Hua University (Taiwan)[19]. Transparencies and refractive indices of aerogels were measured at several irradiation stages. Up to 9.8 Mrad, which corresponds to more than ten years of running at the KEK B-Factory, no deterioration on the transparency and no change in the refractive indices were observed, as shown in Fig. 8. In order to cancel out effects other than the radiation dose, we use transparency ratio, pulse height of the irradiated sample, illuminated by a blue LED normalized by that of nonirradiated sample (reference), which was kept in the same condition (humidity, temperature, etc.) as the irradiated one.

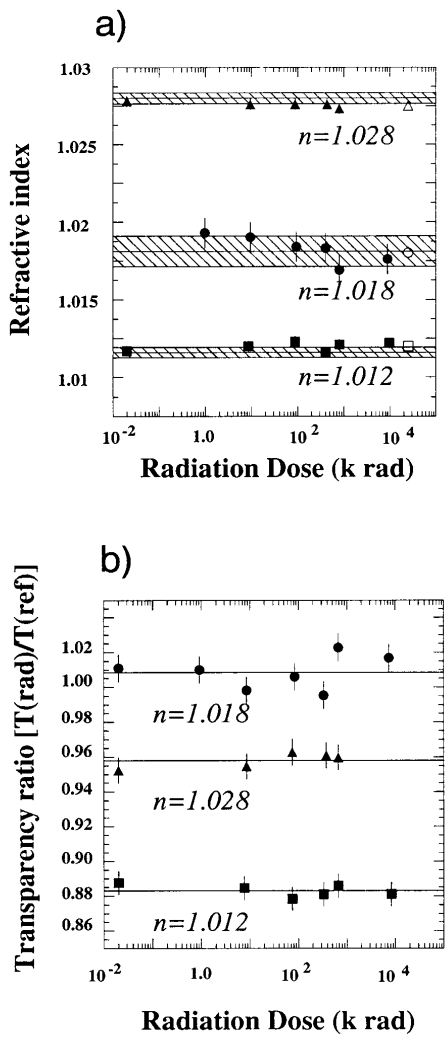

Fig. 8. Effects of radiation dose on aerogels were examined for (a) refractive indices and (b) transparency. In figure (a), the solid lines indicate the refractive indices at the production time and the shaded areas are $\pm 1\sigma$ regions obtained from the measurements at the production time.

4. Performance of the Detector

4.1. Performance of the Single Counter Modules

The performance of single ACC modules has been tested using a 3.5 GeV/c negative pion beam at KEK PS (π 2 beam line). The number of photoelectrons obtained for 3.5 GeV/c pions are 18.2, 20.3, and 20.3 for n = 1.01, 1.015, and 1.02 silica aerogels, respectively. Using equation 3, our photon detection system has higher than 70% efficiency. Typical pulse height distributions for 3.5 GeV/c pions and protons observed by an aerogel counter (n =1.015 with two 2.5" PMT's) are shown in Fig. 9. Pions (above threshold) and protons (below threshold) are clearly separated by more than 3σ. This separation is maintained even in a high magnetic field (1.5 T). We also found that cracks in the aerogel do not make a difference in the light yield. Although the precursor for the preparation of alcogels is different, our silica aerogels have shown performance as good as the aerogels produced by the Jet Propulsion Lab [20]. In terms of the performance in aerogel counters, we believe that there is no significant difference between the aerogel produced by our method and one produced by the normal two-step method in Caltech.

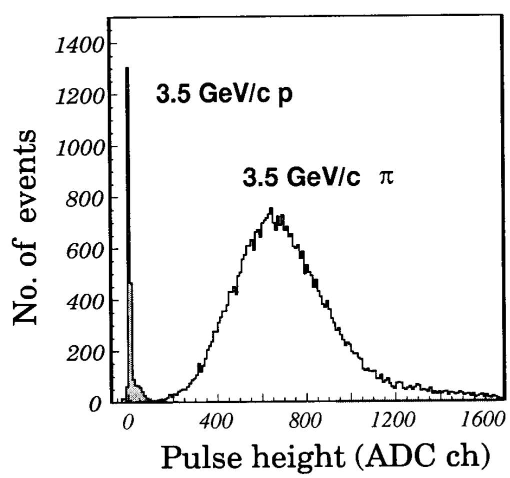

Fig. 9. Pulse height spectra for 3.5 GeV/c pions (above threshold) and protons (below threshold) obtained by a single module of ACC, in which n = 1.015 silica aerogels were stacked. Pions and protons are clearly separated by more than 3σ.

4.2. Monte Carlo Simulation

In order to arrive at a better understanding of the performance of the aerogel counters, we have developed a Monte Carlo program [21], which can simulate the behavior of Cherenkov photons in the aerogel as realistically as possible. All considerable effects such as Rayleigh scattering, absorption by the aerogel, reflection by the Gortex walls, absorption by the wall, and the response of the PMT's are taken into account as a function of wavelength. The only unknown factor is the absorption in the aerogel, which we have treated as a free parameter and later determined by comparing the results from the simulation with the test beam data. Incident position dependence of the pulse height is compared with the simulation. The absorption length thus determined is about 7 m at λ = 400nm and increases almost exponentially as a function of λ.

5. Other Applications

Since our successful mass-production of silica aerogels, several experimental groups have decided to use silica aerogels for their particle identification devices. For example, the cosmic anti-matter search experiment, the LHC-B experiment, and the internal polarized target experiment (HERMES) [22].

The BESS detector [23], which was launched in a balloon to an altitude of around 10,000m, has successfully completed its role for the observation of anti-particles in primary cosmic rays. Antiprotons are separated from other negatively charged particles by using information from the aerogel Cherenkov counter. A schematic drawing of the BESS detector is shown in Fig. 10. A superconductive solenoid combined with a tracking system determines the momenta of charged particles, and the signal on the aerogel counter, where the charged particles are expected to pass, is checked. Recently, a space station experiment, Anti-Matter Spectrometer (AMS), has been proposed [24] for a search of anti-matter nuclei in space, and it was decided that a silica aerogel Cherenkov counter would be used for isotope separation.

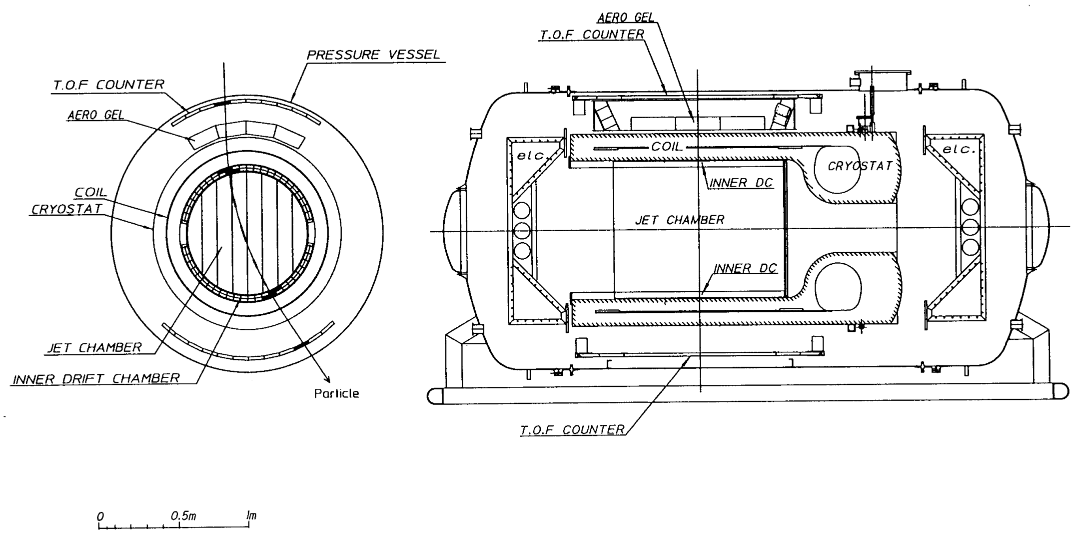

Fig. 10. Layout of the BESS detector. The superconducting solenoidal magnet and the JET-type tracking chamber determines the momenta of charged particles. The TOF and the silica aerogel counters are used for particle identification.

LHC-B and HERMES use aerogels as the radiator for their RICH detectors. Since the scattering length has been improved from the previous one-step aerogels, a clean image of the Cherenkov ring could be observed [25]. We have also succeeded in observing an image of the Cherenkov Ring from our aerogel ($n = 1.03$) radiator of 5 cm thickness, which is shown in Fig. 11. The Cherenkov light was focused on an Image Intensifier (Hamamatsu Photonics Co.) by a concave mirror and recorded on video tapes.

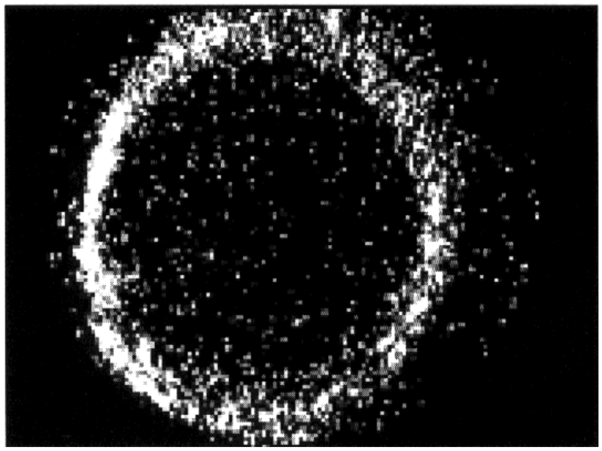

Fig. 11. Ring image of Cherenkov light radiated by 3.5 GeV/c negative pions in the silica aerogel of $n = 1.03$, with a thickness of 5 cm. The image is observed using an image-intensifier. The Cherenkov light was focused by a concave mirror.

6. Conclusion

Silica aerogel is a unique material for threshold-type Cherenkov counter radiators. There are, however, two major difficulties for their application to pion/kaon separation at a momentum range of a few GeV/c. One of the difficulties is achieving a low refractive index ($n < 1.02$) aerogel with high transparency, and the other is long-term stability. The invention of the two-step method and the surface modification to the hydrophobic agent have helped surmount these difficulties. With these advances, good pion/kaon separation is possible up to about 4 GeV/c.

For the BELLE detector at the KEK B-Factory we have produced about 2 m^3 of silica aerogels of $n = 1.01$-1.03 using a new production method. The particle identification capability of the aerogel Cherenkov counters was tested by using real beams. Pion/proton separation of three standard deviations has been achieved. The radiation hardness of aerogels was tested up to 9.8 Mrad. Neither deterioration of transparency nor change in the refractive index was observed, which give us confidence in particle identification with aerogels in a high-radiation environment.

Thanks to their improved transparency, namely longer scattering length, aerogels prepared by the two-step method can be used as radiators for not only threshold-type but also Ring Imaging Cherenkov counters. Thus the newly developed aerogel has opened a new era for particle identification in high-energy experiments.

Acknowledgments

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