Development and Analysis of A New Nuclear Emulsion Film for Detection of Neutrinos

Ken-ichi Kuwabara, Shingo Nishiyama, and Tadaaki Tani
Digital Photo-Imaging Res. Labs., Fuji Photo Film Co., Ltd.
Minami-Ashigara, Kanagawa, Japan

Abstract

A new nuclear emulsion film has been developed for the study of neutrinos by a noteworthy experiment named OPERA in particle physics. They are characterized by enhanced sensitivity, extremely reduced fog level, and resettability. The reset of the emulsion film with erasure of noises (i.e., unwanted particle tracks caused by cosmic rays during storage) before use can be realized by adding an appropriate amount of methyl-benzotriazole to the emulsion and keeping it under highly humid condition for several days. An analysis was made on the mechanism of the reset effect and the role of methyl-benzotriazole in it.

Introduction—Background for Development of New Nuclear Materials

The possibility of a finite mass for the neutrino is one of the most intriguing questions of particle physics. A neutrino mass implies physics beyond the Standard Model of elementary particles, representing a fundamental milestone in particle physics, astrophysics and cosmology. The search for a process which can only occur if the neutrino has a finite mass is crucial to asses this issue. The oscillation between a muon neutrino ν_μ and a tau neutrino ν_τ is such a process, bringing about the deficit of ν_μ and the appearance of ν_τ during the travel of ν_μ from the source to the detector.

Recent neutrino experiments using atmospheric, solar, reactor, and accelerator neutrinos have demonstrated that neutrinos change flavor as they travel from the source to the detector, a phenomenon consistent with the hypothesis of neutrino oscillation. The OPERA experiment has been proposed and scheduled to start in June 2006 in the CNGS long baseline beam from CERN SPS to the Gran Sasso Laboratory at the distance of 730 km in order to get the evidence of the ν_τ appearance from the oscillation, where OPERA is an abbreviated word of Oscillation Project with Emulsion tRacking Apparatus[1].

Although it is not possible to directly observe ν_τ, the evidence for the appearance of ν_τ can be given by the direct detection of the particles produced in the charged current (CC) interaction of the ν_τ with the target. Since a τ particle moves to the distance of only several hundreds μm within its lifetime of 0.3 picoseconds, a device with very high spatial resolution is required to directly detect the τ particle, and only nuclear emulsions can meet this requirement.

Nuclear emulsions have ever been playing important roles in particle physics owing to their high spatial resolution. C. F. Powell could discover Yukawa's particle (i.e., Pion) by use of nuclear emulsions[2]. Recently, only the DONUT experiment performed in the Fermi National Laboratory has succeeded in detecting the ν_τ interactions by means of an ECC (Emulsion Cloud Chamber) detector, which has a modular structure made of a sandwich of passive material plates as the targets for ν_τ interspaced with emulsion sheets as the trackers for the τ particles. While the ECC detector planned for the OPERA experiment is conceptually similar to that used in the DONUT one, the target plates in the former are made of Pb, while those in the latter were made of Fe. The performance of the emulsion used in the OPERA experiment is much better than that in the DONUT one owing to the recent progress in emulsion technology. It is expected that a nuclear emulsion will continue to play important roles in the future in particle physics as a unique three-dimentional detector with resolution of sub-micrometer[3].

Nuclear emulsions have been usually coated so far by hands before use, since unwanted particle tracks caused by cosmic rays during their preparation and storage bring about noises in coated nuclear emulsion sheets. Even in the previous experiment named CHORUS, which was aimed to get the evidence of the formation of ν_τ by the neutrino oscillation, the amount of the nuclear emulsion used was still small enough to coat it by hands before use. Since it was believed at that time that the difference in mass between ν_μ and ν_τ was so large to bring about the oscillation of neutrinos during their travel to a short distance, it was planned to detect the τ particles at the place, which was at a distance of only 800 meters from the source of neutrinos. If the neutrino oscillation took place at that place, it could be detected by the detector with weight of 1 ton containing the nuclear emulsion of 100kg, which could be coated by hands before use.

The failure of the detection of the neutrino oscillation by the CHORUS experiment was explained by the result of the Super-Kamiokande experiment, according to which the difference in mass between ν_μ and ν_τ was so small that the oscillation of neutrinos could not take place during their travel from the source to the detector (i.e., 800 meters), and should be detected at the place, which was located at much longer distance from the source of neutrinos.

In order to prove the oscillation of neutrinos by directly detecting the τ particles produced by τ-nuetrino interaction, it is necessary to significantly increase the distance between the source and the detector of neutrinos, the detector mass (i.e., the masses of the nuclear emulsion and Pb), and the duration for the detection, since the intensity of neutrinos should decrease with increase in the distance between the source and the detector in addition to small probability of the formation of ν_τ from ν_μ(at most several percents) and to small probability of the production of τ particles from ν_τ. Namely, the amount of the nuclear emulsion used for the OPERA experiment should be so large (the detector with weight of 1800 tons containing the emulsion of 180 tons) that the emulsion can not be coated by hands before use. Instead, the emulsion should be coated by a large-scale coater a long time before it will be used for the experiment.

Recently, Super-Kamiokande could determine the mass of neutrino (i.e., the difference of the squared mass eigenvalues Δm^2 as $2.4\times10^{-3}eV^2$) and the wavelength of the oscillation of neutrinos (i.e., 1000km/GeV). These results indicate that the probability of $\nu_\mu\rightarrow\nu_\tau$ event, which takes place during the travel of ν_μ with energy of 20 GeV from CERN SPS to the Gran Sasso Laboratory at the distance of 730 km, is about 1.2 %. This means that the detector with weight of 1800 tons will detect about 10 τ events in five years. It is now planned to increase the $\nu_\mu\rightarrow\nu_\tau$ sensitivity by increasing the source beam intensity at CERN so that the detector will detect 16 τ events in five years.[3]

As stated before, sheets of coated emulsions have a problem to accumulate unwanted particle tracks caused by cosmic rays on their preparation and storage. Kuwabara and Nishiyama have developed the technology to reset sheets of coated nuclear emulsions by eliminating the noises produced by cosmic rays without deteriorating the sensitivity of the emulsions, and made it possible to carry out the OPERA experiment with large amount of the nuclear emulsion, which is coated on film sheets by a large-scale coater a long time before the use for the OPERA experiment[4]. Tani studied the mechanism of the reset effect for the above-stated nuclear emulsion.

The fact that silver halide emulsions are still playing such important roles in particle physics is significant, and is worthy of being presented at AgX 2004 with the theme to look for new possibilities of silver halide imaging.

Development of New Nuclear Materials and Reset Treatment

Recent progress in emulsion technology could realize significant improvements in nuclear emulsions. Namely, the grain diameter of the new nuclear emulsion for the OPERA experiment (0.2 μm) was much smaller than that of the previous one (0.27μm) used for the DONUT experiment, while the sensitivity of the former for a particle in terms of the number of developed grains per unit length of its track is nearly two times higher than that of the latter[4]. Furthermore, the cost of the new emulsion was reduced as compared with that of the previous one as a result of the reduction in the amount of silver halide of the former as compared with that in the latter. The new emulsion grains composed of silver iodobromide were doped with Fe(Π) complex ions in the thin shell, and sulfur-plus-gold-sensitized on the surface in order to enhance their sensitivity and to achieve extremely low fog level (less than 100 ppm by grain number). The emulsion was then coated four times alternately on both sides of a TAC film sheet. A TAC sheet was 200 μm thick, and the emulsion layer on one side was as thick as 44 μm. Gelatin layers with thickness of 1 μm were coated on the surfaces of the emulsion layers in order to prevent them from directly contacting with Pb plates.

Noises (i.e., unwanted particle tracks) produced by cosmic rays during their preparation and storage in the sheets of the above-stated emulsion could be eliminated by the reset treatment. In order to enhance the reset effect of the coated emulsion, about 1/1000 mole of methyl-benzotriazole per mole of silver halide was added to the above-stated emulsion. Then, the reset treatment was carried out by keeping the sheets of the coated emulsion in the atmosphere with high humidity (e.g., 100 %RH at 25-30°C) for several days. It was found that methyl-benzotriazole made it possible to reset the sheets of the coated emulsion by enhancing the elimination of the noises without deteriorating sulfur-plus-gold sensitization centers under highly humid condition[4].

In order to get the knowledge of the dissociation and adsorption of methyl-benzotriazole to emulsion grains together with oxidizing ability of gelatin, pH, redox potential, and silver potential of liquid emulsions and aqueous gelatin solutions were measured at 40°C by use of a glass electrode, a Pt electrode, and an Ag electrode, respectively, with reference to a saturated calomel electrode.

Roles of Benzotriazole in Reset Treatment

Noises accumulated in the sheets of the coated emulsion during their preparation and storage are composed of silver clusters. It is considered that a sulfur-plus-gold sensitization center is composed of a dimer of substitutional sulfide ions associated with an interstitial silver ion and an interstitial gold ion[5]. It is therefore probable that gelatin phase around silver halide grains oxidizes the silver clusters without deteriorating sulfur-plus-gold sensitization centers under highly humid condition in the reset treatment. The oxidation potentials of silver clusters and substitutional sulfide ions in silver bromide grains, which constitute the framework of a sulfur-plus-gold sensitization center, were already estimated in the literature[6]. Under the condition corresponding to that in the reset treatment, the electrochemical redox potential of an aqueous gelatin solution was more positive than the oxidation potential of silver clusters, while it was less positive than the oxidation potential of substitutional sulfide ions in silver bromide grains. This result supports the idea that the gelatin phase in the coated emulsion could oxidize silver clusters constituting the noises formed during storage, while it could not oxidize the substitutional sulfide ions constituting sulfur-plus-gold sensitization centers. It is therefore proposed that the reset effect of the coated nuclear emulsion was attributed to the oxidation of the noises without deterioration of chemical sensitization centers by gelatin phase in the emulsion under highly humid condition.

Since the pKa value of methyl-benzotriazole is as high as 8.50, methyl-benzotriazole dissociates to form its anion with high adsorptivity to silver halide when pH of emulsions is higher than 8.50, while it does not dissociates and has little adsorptivity to silver halide when pH of emulsions is lower than 8.50[6]. In accord with this knowledge, it was experimentally confirmed that methyl-benzotriazole was strongly adsorbed to silver bromide grains at pH higher than 8.50, and hardly adsorbed to the grains at pH lower than 8.50. Since a methyl-benzotriazole anion is adsorbed to a silver halide grain by strongly combining with a silver ion on the grain surface[6], it should principally enhance the oxidation of silver particles and retards the development process by decreasing silver potentials of emulsions. After methyl-benzotriazole is added to an emulsion, the pH of the emulsion is kept to be lower than 8.50 at various stages including the reset treatment, and becomes to be higher than 8.50 for the first time when the coated emulsion are introduced into a developer. It is therefore thought that the retarding effect of methyl-benzotriazole can be effective only at the development process, and is not effective at any other processes including the reset treatment process.

It is known that the oxidation potentials of latent sub-image centers and small latent image centers are more positive than those of large silver clusters[6]. Although the complete oxidation of silver paricles results in the disappearance of all the clusters present, partial oxidation preferentially oxidizes large clusters, and converts them into small ones. Since the oxidizing power of gelatin is too small to achieve the complete oxidation of silver clusters, it is considered that only small silver clusters with size comparable to latent sub-image centers and small latent image centers are left on emulsion grains after the reset treatment.

It is considered that silver clusters as formed by cosmic rays are so large that they can initiate development much more quickly than small latent image centers and latent sub-image centers. It is known that the retardation of the development by such antifoggants as methyl-benzotriazole is not effective when the development is initiated quickly, while it is effective when the development is initiated slowly.

On the basis of the above-stated results and considerations, the role of methyl-benzotriazole in the reset treatment is proposed as follows. During the reset treatment process, gelatin phase in the coated emulsion brings about the partial oxidation of silver clusters constituting the noises produced by cosmic rays during storage and converts them into small ones, which initiate development so slowly as to be effectively retarded by a methyl-benzotriazole. A methyl-benzotriazole, which is introduced into emulsions, has little influence on the processes other than development, since it does not dissociate to form its anion with adsorptivity to silver halide under the conditions on these processes. On the other hand, a methyl-benzotriazole dissociates and effectively depresses the development process, which is slowly initiated by small silver clusters, which are converted from large clusters during the reset treatment, while it hardly depress the development, which is quickly initiated by large silver clusters formed after the reset treatment.

Conclusion

A new nuclear emulsion film has been developed for the direct detection of the neutrino oscillation by thr OPERA experiment. Since the amount of the emulsion is too large to be coated by hands before use, the technology has been developed to mass-produce the sheets of the emulsion by a large-scale coater a long time before their use and reset them. The reset can be realized by treating the sheets of the emulsion containing methyl-benzotriazole under highly humid condition for several days. The following mechanism has been proposed for the reset effect and supported by experiments. Namely, gelatin phase in the emulsion preferentially oxidizes large silver clusters constituting the noises caused by cosmic rays during storage, and converts them into small ones without deteriorating sulfur-plus-gold sensitization centers. While a methyl-benzotriazole has little influence on processes other than development, it depresses the development slowly initiated by the above-stated small clusters without depressing the development quickly initiated by large silver clusters formed after the reset treatment.

References

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Biography

Tadaaki Tani received his B.S. degree in Synthetic Chemistry from the University of Tokyo in 1963 and a Ph.D. in Applied Chemistry from the same University in 1968. Since 1968 he has worked in Research Laboratories in Ashigara, Fuji Photo Film Co., Ltd. His work has primarily focused on the mechanisms of photographic processes including latent image formation, physical properties of silver halide grains, chemical and spectral sensitizations. He is a member of the SPSTJ, IS&T, RPS.