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REVIEW

Total Absorption Counter and Viewing Shield by The Use of Heavy Liquids†

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Present status of our development of using heavy liquids as a radiator of total absorption counters in particle physics experiment is described together with their uses as a viewing shield for radiation protection. The data of transmission curves are presented for the solutions of ZnI_2, of $Tl(HCO_2)$, of the mixture of $Tl(HCO_2$ plus $CH_2(COOTl)_2$ and of $SbCl_3$. Discussions are given on the problems to be solved for using them as practical tools as well as on the future possibilities.

KEY WORDS : Total absorption counter/ Viewing shield/ Heavy liquids/ Particle physics experiments/

Some hologenized salts or complex salts of metal indicate high solubility in water or in other specific solvents. The density of the solution reaches 2~4 g/cm^3 and the solution is called a heavy liquid. These heavy liquids have been known since old time and used in the field of mineralogy for density separation of ores, either floating or sinking in the heavy liquid. For this application, a physical property, such as viscosity or surface tension, becomes more inportant than the transmission, in addition to the density.

Another application of heavy liquids is for a transparent radiation shielding material if it is well transparent. A water solution of zinc bromide ($ZnBr_2$) was used as a viewing shield at hot laboratories two or three decades ago, some of which has been used still at the present. It will be described again later.

We have investigated a possibility of using a heavy liquid as a radiator for total absorption counter by examining the transmission of heavy liquids and testing them as a Cherenkov counter with a tagged electron beam. We summarize here the essential part of the results obtained[4,5,13] and discuss the problems to be solved and the future possibilities of development. We assume that the majority of readers are physicists or chemists working in the fields other than particle physics as many of the problems and the possibilities are related with solid-state physics or chemistry.

After the incidence of high-energy particle or photon on the radiator of total absorption counter,[1,2] the energy is dissipated in the radiator through the processes of electromagnetic cascade shower, of hadronic cascade shower or both. The energy

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dissipated in the radiator is converted to the light through Cherenkov or scintillation processes and the light is further converted to an electric pulse through a photomultiplier. If the leakage of cascade particles and the loss of light in the radiator are small, the size of the electric pulse should be in proportion with the energy of the incident charged particle or photon.

From the mechanisms of the counter described above, necessary requirements for a radiator of total absorption counter are, (1) high density, (2) short radiation length and (3) high transmittance for light of wave-length corresponding to the sensitive region of a photomultiplier. The dimension of a radiator of total absorption counter in practical experiments is required to be 20~30 radiation lengths (r. l.); thus, the size becomes frequently of several m^3. Long-term stability and economy are also the important factors for designing a total absorption counter together with requirements from the physics aims.

As the materials to fulfill those requirements, (a) sodium iodide (NaI) crystal, (b) block of lead glass and (c) combination of plastic scintillator and heavy metal, such as iron, lead, tungsten, are used frequently for the radiator of total absorption counters. Among these materials (a) gives the scintillation light and thus the amount of light is large resulting in a high resolution. The drawback of NaI crystal is a limitation in size and shape, fragility and high price as it is a hygroscopic mono-crystal. (b) is the most common Cherenkov radiator used for many experiments. However, this material has still a restriction in size and shape. (c) is the least expensive and many combinations are possible. However, most of the energy is dissipated in the heavy metal and the counter measures the energy on a basis of sampling. Thus, in general, the energy resolution is poor due primarily to statistical fluctuations in the cascade processes.

Use of liquid as the radiator of a Cherenkov counter has been made for a threshold Cherenkov counter[3] by adjusting the refractive index of the liquid by means of the concentration of the solvent in such a way that a charged particle of velocity exceeding a certain value gives a signal. However, very few works have been reported for using a liquid as the radiator of a total absorption counter.

At the time around 1956[6] when a block of lead glass was tried for use as a Cherenkov counter, Fidecaro[7] proposed to use a water solution of lead perchlorate ($Pb(ClO_4)_2$) as a radiator of total absorption counter. The density of this solution was $d=2.77\,g/cm^3$, the radiation length $X_0 \simeq 4.1$ cm and the transmission was fairly high in comparison with that of lead glass. However, this heavy liquid has a serious drawback that due to the perchlorate it becomes explosive by a little shock when it is dried.[8] For this season, the lead perchlorate solution has not been used widely[9] from the point of view of safety. Matano *et al.*[10] used a water solution of lead nitrate ($Pb(NO_3)_2$) as a total absorption counter for measurements of extensive air-shower in 1957. Due to a limited solubility of this material, the radiation length obtained was 11~13 cm, about 4 times larger than that of lead glass at that time.

Groom[11] tried to use a ternary solution of lead acetate ($Pb(CH_3COO)_2$), acetic acid plus water and obtained a heavy liquid having the radiation length $X_0 \simeq 3.5$ cm. The liquid was likely so sensitive to temperature and easily crystallized by some unknown mechanism that it was not used for a practical experiment.

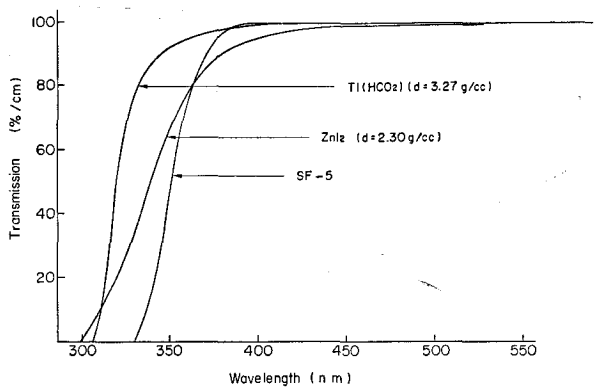

Fig. 1. The transmission (per unit length) of light through $Tl(HCO_2)$ solution in comparison with that of SF-5 lead glass as a function of the wave-length of light.

Varforomeev *et al.*[12] tried to use the Clerici solution, described in detail later, as the radiator of a total absorption counter. The Clerici solution is a heavy liquid used for mineral separation and shows a density $d \simeq 4.2\,g/cm^3$, a radiation length $X_0 \simeq 1.9$ cm. The liquid has an extremely high density and seems to be very attractive as a radiator. However, the transmission is not sufficiently high for the radiator of a total absorption counter. Consequently, the energy resolution obtained by these authors was poor ($\Delta E/E \sim 45\%$ for $2 \sim 4$ GeV/c e), that it has not been used for practical experiments.

We have developed a heavy liquid of thallium formate ($Tl(HCO_2)$)[5,13] as the radiator of total absorption counters. The liquid has a density $d \simeq 3.27\,g/cm^3$ at room temperature, a refractive index $n = 1.57$ and a corresponding radiation length $X_0 \simeq 2.57$ cm. The transmission is $T = 99\%$ at $\lambda = 400$ nm and it extends to shorter wave-length than that of SF-5 lead glass as shown in Fig. 1. The results of a test using the test beam T1 of the KEK 12 GeV proton synchrotron revealed that the characteristics of thallium formate solution are comparable or even superior to those of SF-5 lead glass as the radiator of a total absorption counter. In particular, the resistivity of thallium formate solution against radiation is much greater (perhaps more than 10^3 times) than that of the SF-5 lead glass.

In past, thallium metal has not been used widely and regarded as an almost useless metal having a limited application as a rodenticide and insecticide so that the production rate of this metal has been low. The cost is comparable to that of SF-5 lead glass of the same weight if we take the price of reagent grade of thallium formate. It can be expected that the price will be lowered by a factor for a large scale production. However, both the supply and demand are difficult to predict and this fact reflects the price unstable. Another drawback of thallium complexes is its toxity. The toxity of thallium compounds is, in general, a bit less than that of mercury or arsenic in terms of the LD_{50} values. Nevertheless, thallium compounds appeared as poisons in criminal affairs and sufficient precaution and control have to be exercised before its practical application, particularly with large quantities. The vessel should be designed to be perfectly water-

tight and many technical problems should be solved before utilizing it as a practical detector. In addition, long-term stability is required for use as a practical detector but little data has appeared in literature[14] on physical and chemical properties of thallium complexes. However, we believe that most of these problems could be solved by present-day chemical engineering techniques applied at many plants in the chemical industries.

The high solubility of thallium carboxylates, including thallium formate, were known since old time shortly after its discovery by W. Crooks in 1860's. As far as the density is concerned, about 1800 g of the mixture of approximately equal amounts of thallium formate ($Tl(HCO_2)$) and thallium malonate ($CH_2(COOTl)_2$) can dissolve in 100 cm^3 of water at room temperature and forms a liquid of density $d=4.24\,g/cm^3$. The liquid was found by an Italian mineralogist, E. Clerici[15] in 1907 and has been used as a heavy liquid for mineral separation with the name of the Clerici solution. The solution appears to be of low viscosity and is almost as colourless as water although of much higher density. The calculated radiation length is $X_0 \simeq 1.92$ cm, much shorter than that of SF-5 lead glass, $X_0 \simeq 2.54$ cm. However, the transmission of the Clerici solution is unfortunately not high enough to be used as the radiator of a total absorption counters as was reported by Varforomeev et al., described before. The results of our measurements[13] using tagged electrons from the test beam Tl of the KEK proton synchrotron also verified the same for the transmission as shown in Fig. 2 and for the energy resolution.

We tried to improve the transmission of Clerici solution by many trials including purification. However, the problem was solved by the simplest way. By adding a small amount of water into the saturated Clerici solution and lowering the density from 4.24 g/cm^3 to 4.00 g/cm^3, it was observed that the transmission improved significantly as shown in Fig. 2.[13] The radiation length corresponding to the diluted solution was still $X_0 \simeq 2.1$ cm and the value was consistent with the performance as a total absorption counter with tagged electrons. The results of measurement indicated that the transmission of diluted Clerici solution came close to fulfilling the requirement of total absorption counter.

The reason why the slight dilution improved the transmission has not been clear

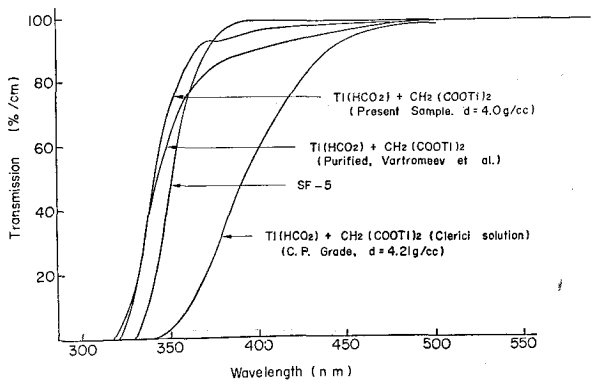

Fig. 2. The transmission (per unit length) of light through the Clerici solutions of various densities and grades in comparison with that of SF-5 lead glass as a function of the wave-length of light.

yet. It can not be explained by a simple arithmetic or geometric mean of transmissions of the saturated Clerici solution and of pure water. At this point, we must confess that we are laymen on those problems and we would appreciate suggestions from specialists in this field.

It can be shown easily that the system of the Clerici solution consists of approximately each one molecule of thallium formate, thallium malonate and water. For the case of thallium formate solution, one molecule of thallium formate combines with only two molecules of water. Thus it appears to us that the solution is far from a normal concept of solute and solvent but rather a sort of eutectic complex behaving as if it were water except for the density. The author searched the literature for works related to these problems but he was not successful in finding them.

To the extent of our experience, both thallium formate solution and the Clerici solution are very stable solutions. By leaving these solutions without special protection against air, light etc., no change has been observed in physical and chemical properties of these solutions for more than one year. The viscosities are similar to that of water and the solutions are neither explosive nor volatile. We have no difficulty in handling these heavy liquids except for their potential toxities. However, we admit that, due to toxities, there should be much restrictions and problems to be solved prior to practical application of these solutions. From the point of view of practical application, we have been developing other heavy liquids, such as zinc iodide (ZnI_2) solution, antimony trichloride ($SbCl_3$) solution etc.[4,13]

We have obtained a zinc iodide (ZnI_2) solution of a density $d \simeq 2.7\ g/cm^3$, corresponding to a radiation length $X_0 \simeq 4.0$ cm. The transmission of the processed zinc iodide solution is, at present, about to fulfill the requirement of a radiator of total absorption counter, as shown in Fig. 3.

A pure water solution of zinc iodide will be discoloured by light irradiation to yellowish brown. This is caused by oxidation resulting from light irradiation and it can be suppressed by adding an adequate reducing agent. The difficulty lies that some reducing agent makes a chemical reaction to zinc iodide. We tried acetone first, however

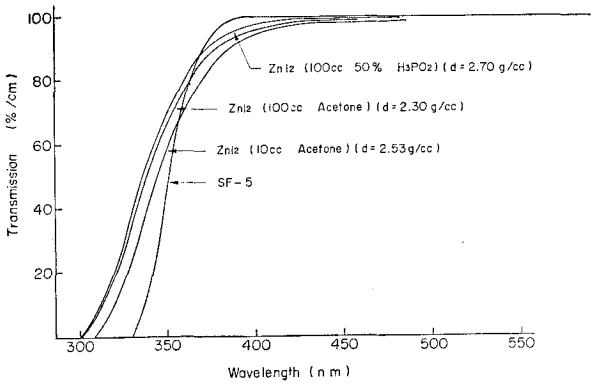

Fig. 3. The transmission (per unit length) of light through ZnI_2, solutions of various densities in comparison with that of SF-5 lead glass as a function of the wave-length of light.

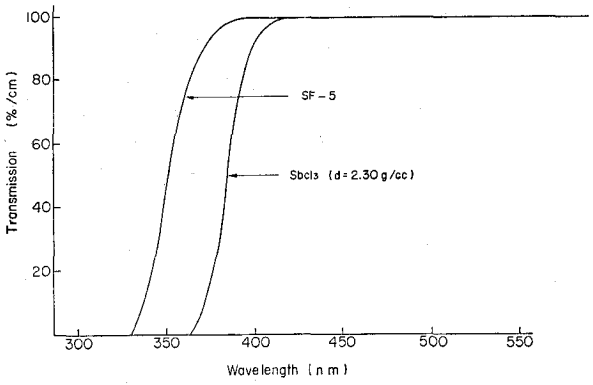

Fig. 4. The transmission (per unit length) of light through $SbCl_3$ solution in comparison with that of SF-5 lead glass as a function of the wave-length of light.

acetone attacks the organic materials used in the component of vessel. At the present, we use hypophosphorous acid (H_3PO_2) as a reducing agent and the solution gives a satisfactory result as shown in Fig. 3. However, this material should also be tested for a long-term stability against the materials used for the vessel.

The solution of antimony trichloride ($SbCl_3$) has a density $d \simeq 2.3\,g/cm^3$, corresponding to a radiation length $X_0 \simeq 5$ cm. The solvent should not be pure water as commonly appears in handbooks since pure water hydrates antimony chloride. We obtained a satisfactory result by using 10 N hydrochloride (HCl) solution as the solvent. The transmission is close to fulfilling the requirement for a radiator however the cut-off wave-length is around $\lambda \simeq 360$ nm, much longer than those of other heavy liquids and that of SF-5 lead glass as shown in Fig. 4.

At room temperature, antimony trichloride is in a crystalized state and the melting point is as low as 80°C. If the melted antimony chloride in a flask is cooled rapidly to the liquid nitrogen temperature by dipping the glass flask in a liquid nitrogen reservoir, the melt is frozen to a transparent solid of glassy state. By taking out the flask from the liquid nitrogen reservoir and leaving it at room temperature, the transparent glassy solid changes to a milky liquid of high viscosity and then becomes again the crytallized solid form. We wonder whether this transition from the transparent solid state to the crystallized solid state via the milky liquid of high viscosity is so-called "glass transition" or not. Again, we are laymen on these problems and we do not know even whether it is normal or abnormal.

Nevertheless, we consider that if there is a material of high density, of relatively low melting point, becoming a transparent solid or gel state at room temperature, the material would be very attractive and useful as a radiator of total absorption counter.

The cost of zinc iodide or antimony trichloride is approximately 2/1～1/4 of lead glass of the same weight and these are readily obtainable. The toxity of these materials is almost non or, at most, about the same order as that of lead; however sufficient care is required for handling them as these solutions are saturated and cause burns to the skin by direct contact.

So far we described the properties of heavy liquids for their uses as a radiator of total sbsorption counter; however, these heavy liquids can be used as transparent shielding materials.[16] By considering the composition, the density and the radiation length of each material described above, the shielding capability of the thallium formate solution and of the Clerici solution are comparable to or even superior to that of baryte concrete and those of zinc iodide solution and antimony trichloride solution to that of ordinary concrete.

As described before, a solution of zinc bromide ($ZnBr_2$) was used extensively at hot laboratories as a transparent (viewing) shield two or three decades ago,[17] and some have been used even at present. The radiation and transmission properties of the liquids described here are superior to those of zinc bromide solution. However, for a practical use, long term stability has to be proved in the same way as for the case of total absorption counter.

One of the attractive possibilities of using a heavy liquid as the radiator of a total absorption counter is to increase the amount of light from the radiator by mixing a scintillating or wave-shifting material with the heavy liquid. If it becomes possible, it could afford a useful material for radiation detectors. However, our knowledge of the structure of heavy liquids is very limited and many investigations have to be done before ad-hoc trials. It is likely that the structure of heavy liquid is closely related to those of melts and metal complexes in solutions. These investigations are growing so rapidly at present[18,19] that the authors are anxious to solicit their interest on heavy liquids which we can expect to have many useful applications in particle physics or in radiation protection.

The author is afraid that he would commit many mistakes in describing this note as he deals with problems outside of his speciality, i. e., of experimental particle physics. He is grateful if the reader points out or corrects the errors inadvertently made in this note and the related articles by him.

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