Functional Materials 10, No.3 (2003) © 2003 — Institute for Single Crystals

Service life forecasting for scintillation tiles of HE calorimeter for CMS

V.G.Senchishin, N.P.Khlapova*, V.N.Lebedev, A.F.Adadurov

Institute for Scintillation Materials, National Academy of Sciences of Ukraine, 60 Lenin Ave., 61001 Kharkiv, Ukraine
*V.Karazin Kharkiv National University, 4 Svobody Sq., 61077 Kharkiv, Ukraine

Received September 16, 2002

Accelerated thermal aging tests have been carried out on UPS-923A and UPS-98RH scintillation materials as well as on their foreign analogues, SCSN-81 (KURARAY) and BC-408 (BICRON). From the test results, the service life duration has been predicted for scintillators operated under normal conditions up to maximum acceptable strength limit reduction (50 %) and the light yield degradation (20 %). The service life of UPS-923A has been shown to be the highest one and to amount 8 to 10 years. The service life of UPS-98RH scintillators is 6 to 8 years while that of SCSN-81, 6 to 10 years. It is just BC-408 scintillators that are shown the maximum long-term stability of mechanical strength and light yield (5 to 6 years).

Проведены ускоренные испытания на тепловое старение сцинтилляционных материалов — UPS-923A, UPS-98RH и их зарубежных аналогов — SCSN-81 (Kuraray) и BC-408 (Bicron). По результатам испытаний сделан прогноз срока службы сцинтилляторов при нормальных условиях эксплуатации для максимально допустимого снижения предела прочности (50 %) и светового выхода (20 %). Показано, что долговечность для UPS-923A является максимальной и составляет 8–10 лет. Для сцинтилляторов UPS-98RH долговечность составляет — 6–8, а для SCSN-81 — 6–10 лет. Минимальная долговременная стабильность по механической прочности и по световому выходу — 5–6 лет оказалась у сцинтилляторов BC-408.

The applicability of plastic scintillators (PS) in high energy physics experiments is often limited by insufficient stability of their properties. Due to external de-stabilizing factors, point defects, micropores, silver cracks (crazes) and microcracks arise in time both at the PS surface and in bulk. As a result, the PS surface becomes dull, the scintillator losses its transparence and its initial light yield becomes reduced. These changes can be caused by internal and surface stresses arising in the PS polymer matrix in the course of synthesis (due to fixed arrangement of its macromolecules that is not the equilibrium one), migration of low molecular mass impurities from the bulk to the surface as well as by oxidation processes involving O_2 occurring both in the scintillator bulk and at its surface.

To reveal and study damage arising in due course in the scintillators intended for the Hadron Endcap (HE) calorimeter of CMS detector, accelerated aging tests of those PS were carried out in air at elevated temperatures. Such tests make it possible to determine the PS heat stability but also, according to the known principle of temperature-time superposition [1], to forecast the scintillator material behavior under normal operating conditions, that is, to evaluate the long-term stability of the PS characteristics.

The HE calorimeter detection system of HMS detector includes up to 1,000,000 thin

scintillating plates, so-called tiles [2]. Since the scintillating tiles will be exposed to continuous ionizing irradiation for long time, their radiation resistance is tested obligatory prior to service. In our opinion, tests of the PS long-term stability are also of critical importance. Such tests are necessary because characteristics of PS do not agree in some cases with forecasts made basing on the radiation resistance test results. This unpredictable behavior of PS seems to be due to the fact that there is a rather long time lag between the scintillator production and its service onset (i.e. its montage in the detecting system). During that period, the PS may be exposed to air oxygen, elevated temperatures, mechanical strains, etc. Thus, the full pre-history of a PS is to be known for a reliable evaluation of its service reliability. Such information is unknown in most cases as well as data on the forecast long-term stability of PS. In such situations, uncontrollable variations of studies are possible that hinder to establish the functional dependence of the PS characteristics on the ionizing radiation parameters.

The aging tests were carried out using PS samples shaped as $4\times125\times125$ mm^3 plates (tiles) and $5\times4\times150$ mm^3 bars. The PS were prepared by block polymerization in aluminum ampoule (UPS-923A) and in glass molds, using so-called "glass" technology (UPS-98RH) [4]. The samples of PS made by KURARAY (SCSN-81) and BICRON (BC-408) were also tested for comparison.

The thermal aging tests were carried out using two air thermostats at 40 and 75 °C for 200 h. The samples were remover from the ovens and compared with control ones every 50 h. To estimate the damage extent, the tiles were inspected visually and the surface quality was studied using a MIM-7 microscope (50×). The Vickers microhardness (MH) of the samples was measured using a PMT-3 instrument [5]. At each testing stage, the bulk attenuation length (BAL), technical attenuation length (TAL), and the tile light yield were measured according to the scintillation characteristic measurement procedure [6]. The tension strength (σ_t) was determined on a RM-500 testing unit at the pulling speed 5 mm/min using the $5\times4\times150$ mm^3 samples. The test result was expressed as the ultimate tensile strength value averaged for 5 samples [7].

The long-term scintillator stability was determined (according to [2]) as the time period within which the light yield change at normal temperature (20 °C) did not exceed 20 % and the strength limit one, 50 %.

Visual and microscopic observations of PS samples evidence that the UPS-923A and SCSN-81 tiles heat treated at 40 °C for 200 h are substantially free of microcracks both at the surface and in the bulk. In UPS-98RH and BC-408 tiles, the surface silver cracks arise in small amounts but the surface quality is not deteriorated.

In SCSN-81 samples, the surface microcracks appear already after a relatively short heat treatment at 75 °C (50 h). These cracks propagate later over the whole tile surface, so that a polygonal network structure is observed. This deterioration of the surface quality is observable by simple eye as a slight dullness. In UPS-98RH and BC-408 samples, the surface silver cracks arise after 50 h exposure, so the surface becomes dull. The results of microscopic examinations are presented in Table 1.

The silver cracks so formed are distributed in an extremely non-uniform fashion both in the sample volume and on the surface. Wide zones are seen containing numerous diverging short and narrow crazes, although single crazes are observed, too. The wide craze zones at the surface are cen-

Table 1. Changes in tile BAL, TAL, and surface quality after 200 h heat treatment at 75 °C

PS type	BAL (cm)		TAL (cm)		Surface state after test
	Prior to test	Post-test	Prior to test	Post-test	
UPS-923A	250	230	120	40	Single large microcracks
SCSN-81	110	103	70	41	Net of small microcracks
UPS-98RH	110	100	75	25	Large zones of silver cracks
BC-408	300	280	150	17	Craze zones bound together

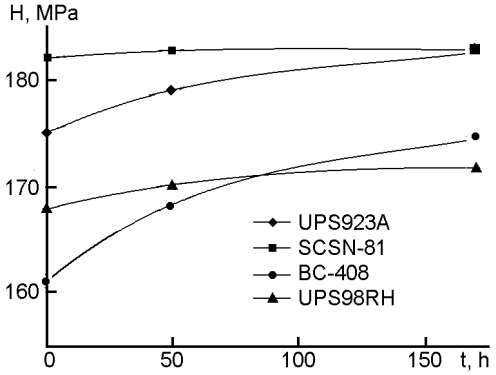

Fig. 1. Microhardness changes of scintillation tiles due to heat aging at 40 °C.

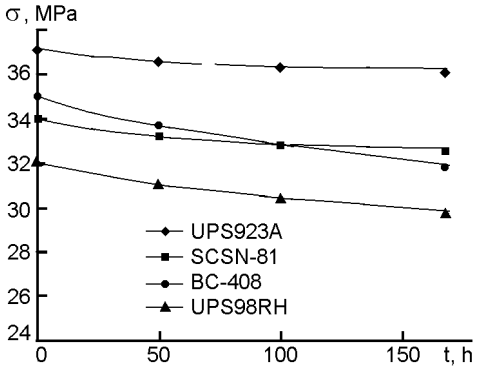

Fig. 2. Ultimate strength changes of scintillation tiles due to heat aging at 75 °C.

tered at various faults such as scars, cleaves at the tile edges, micronotches, contaminations, etc. As to the sample bulk, micropores, low molecular mass inclusions, etc., can be supposed to act as similar centers of silver crack formation. The internal silver cracks are simple crazes while all the surface crazes are bound together, as was observed in [8].

The tile surface silvering observed both by simple eye and in microscope does not affect essentially the BAL value. The maximum BAL reduction after 200 h exposure at 75 °C is about 4 to 5 per cent.

In contrast to BAL, a reduction of the technical light attenuation length (TAL) attaining 5 % is observed in BC-408 and UPS-98RH samples already after the first testing (heat treatment at 40 °C). The test temperature elevation up to 75 °C causes a TAL reduction in all PS. After the 200 h exposure, it amounted 42 % for SCSN, 60 % for UPS-923A and UPS-98RH, and 90 % for BC-408 (see Table 1). The dramatic TAL drop for UPS-98RH and BC-408 is due to the deterioration of the tile surface reflectance caused by silvering [9].

The microhardness (MH) data for untreated and treated tiles are presented in Fig. 1. The microindention (MI) method was selected to control changes in mechanical properties of scintillation plates for the following reasons. First, it is the most convenient simple and non-destructive method to obtain the required data immediately for finished products. Second, the MI testing makes it possible to monitor the changes in the area (subsurface tile layers) where the defect concentration is the highest.

It is seen from Fig. 1 that 150 h exposure at 40 °C causes no essential MH changes in UPS-923A and UPS-98RH samples. For SCSN-81, the MH increase by 5 % has been observed while this increase amounted 10 % for BC-408. The heat treatment at 75 °C resulted in more considerable MH changes attaining 10 % for UPS-923A, UPS-98RH, and SCSN-81, and almost 45 % for BC-408.

The effective MH increase resulting from the heat treatment evidences increased brittleness of the tile material. The same is confirmed by rolls and microcracks arising at the indentation edge and localized along the faces. In some cases with BC-408, the indentation shape distortion was observed and relief, stripes, and microcracks arose at its edges. The number of "poor" indentations for BC-408 differs from those for other materials, thus, this material can be considered as a more brittle one with an imperfect surface. This is confirmed by MH measurements at the tile outer side and at the sawing surface. The microhardness values differ by 30 % for BC-408, by 20 % for UPS-98RH, and as slightly as by 3 % for UPS-923A and SCSN-81.

These differences seem to be due to the tile production method. In fact, the PS from KURARAY (SCSN-81) is a cross-linked plastic produced by rolling. Such a material has a dense quenched defect-free surface layer that is hardly degradable. The UPS-923A tiles cut out of a polymerized block and diamond-polished have an ideal smooth (damage-free) surface providing the following high resistance of the tile. In contrast to UPS-923A and SCSN-81, the BC-408 and UPS-98RH tiles have been produced using the glass technology. This production method itself predetermines tensile stresses in the plastic surface layer due to the mass being polymerized to silicate glass hindering the free shrinkage. As a result, the tensile stresses are fixed at the tile surface. In due course, the surface silver cracks are formed and propagated at such a surface. It

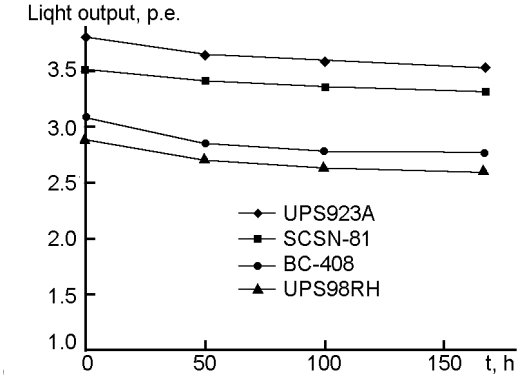

Fig. 3. Light yield changes of scintillation tiles due to heat aging at 75 °C.

is to note that the surface tensile stresses intensify dramatically the aggressive oxygen effect on the polymer. The intense oxidation occurring in this case favor the propagation of the existing cracks and thus generation of a new surfaces.

The tensile stresses in the surface layer can be reduced by a short annealing. For example, the UPS-98RH tiles were annealed at 65 °C for several hours (10 to 20) to remove the surface silver cracks. The anneal resulted in removal of the cracks and the light yield increased by a factor of 1.5 to 1.7. Alternatively, the surface tensile stresses can be reduced by quenching generating compressing stresses [10].

Fig. 2 presents the ultimate tensile strength values for the PS samples. Comparison of these data with the tile MH changes (Fig. 1) shows that the strength decreases as the elevated temperature exposure duration increases, while the MH rises. This relationship evidences the increase of brittleness and deformability reduction of the polymer PS materials.

The light yield data for the tiles are presented in Fig. 3. The temperature effect is seen to be manifested as a monotonous decrease of light yield being considerable at the teast temperature of 75 °C. The Figs. 2 and 3 show that the light yield change over the whole aging period (200 h at 75 °C) did not exceed 10 % to 15 % while the tensile strength dropped by 15 % to 25 %.

From the time dependences of the ultimate strength and light yield determined in the aging tests, the time values have been calculated corresponding to the tile light yield change by 20 % and the strength

Table 2. Long-term stability of plastic scintillators defined by light yield and mechanical strength

PS	τ_{20}, years (20 % light yield reduction)	τ_{20}, years (20 % τ_{20} reduction)
SCSN-81	10	6.2
BC-408	6	5.1
UPS-98RH	8	5.8
UPS-923A	10	8.2

limit, by 50 %. The long-term stability τ_{20} was determined by extrapolating the obtained time values to normal conditions (20 °C). The data so obtained are presented in Table 2.

It is just the UPS-923A scintillators that exhibit the highest heat resistance. The long-term stability as determined by the 50 % change of the ultimate strength (σ_t) is 8 years for those tiles. The minimum stability in mechanical strength (about 5 years) is characteristic of BC-408. The maximum stability as determined by light yield (10 years) has been shown for UPS-923A and SCSN-81.

References

1. O.N.Karpukhin, *Plast. Massy*, No. 11, 27 (1978).
2. CMS. The Hadron Calorimeter. Technical Design Report, CERN/LHCC 97-31, CMD TDR 2, June 20, 1997.
3. K.F.Johnson, H.L.Whitaker, *Nucl. Instr. Meth.*, **A 301**, 372 (1991).
4. V.G.Senchishin, Abstr. Dr. Sci. Thesis (Tech.), ISC Publ., Kharkiv (2001).
5. M.S.Kats, V.R.Regel, *Mekhanika Polymerov*, No. 1, 22 (1973).
6. V.G.Senchishin, V.L.Vasilchuk, A.Yu.Borisenko et al., *Quantum Electronics & Optoelectronics*, **3**, 223 (2000).
7. GOST 11262-80. Plastics: Tensile Testing Methods, Izd. Standartov, Moscow (1986) [in Russian].
8. I.Narisova, Strength of Polymer Materials. Moskow, "Chemistry" publishing house, 1987, p. 399.
9. G.Kettering, *Nucl. Instr. Meth.*, **131**, 451 (1975).
10. K.N.Streltsov, Technology of Plexiglas Products, Mashinostroenie, Leningrad (1984) [in Russian].

Прогнозування строку служби сцинтиляційних тайлів HE калориметра для CMS

В.Г.Сенчишин, Н.П.Хлапова, В.Н.Лебедєв, А.Ф.Ададуров

Проведено прискорені випробування на теплове старіння сцинтиляційних матеріалів — UPS-923A, UPS-98RH та їхніх закордонних аналогів — SCSN-81 (Kuraray) та BC-408 (Bicron). За результатами випробувань зроблено прогноз строку служби сцинтиляторів при нормальних умовах експлуатації для максимально допустимого зниження межі міцності (50 %) і світлового виходу (20 %). Показано, що довговічність для UPS-923A є максимальною і складає 8–10 років. Для сцинтиляторів UPS-98RH довговічність складає — 6–8, а для SCSN-81 — 6–10 років. Мінімальна довготермінова стабільність за механічною міцністю та світловим виходом — 5–6 років виявилась у сцинтиляторів BC-408.