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RADIATION DAMAGE STUDIES ON POLYSTYRENE-BASED SCINTILLATORS

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Abstract

Britvich G.I. et al. Radiation Damage Studies on Polystyrene-Based Scintillators: IHEP Preprint 91-187. - Protvino, 1991. - p. 11, tables 6, fig.1.

The radiation resistance of polystyrene-based scintillators containing various scintillation solutes is reported. All samples were irradiated to ^{137}Cs gamma rays in air at room temperature. The examination of radiation resistance of about thirty fluorescence compounds has been made. The most radiation-hard fluors are X25, X31, 3HF and M3HF.

Аннотация

Бритвич Г.И. и др. Исследование радиационной стойкости сцинтилляторов на основе полистирола: Препринт ИФВЭ 9I-I87. - Протвино, I99I. - II с., 6 табл., I рис.

Исследована радиационная стойкость полистирольных сцинтилляторов с различными сцинтилляционными добавками. Образцы толщиной 5 мм и диаметром 25 мм облучались γ-квантами от источника ^{137}Cs на воздухе при комнатной температуре, в этих же условиях наблюдалось частичное восстановление световыхода облученных образцов. Исследовано более 30 люминофоров, наиболее радиационно-стойкие из них - Х25б Х3Iб 3HF и M3HF.

INTRODUCTION

During last few years the interest in the radiation-resistant scintillators has renewed for the application in scintillator-based particle detectors at new high energy accelerators SSC, LHC and UNK. As has been pointed out elsewhere [1], polystyrene (PS) is not a sufficient solvent for scintillation solutes in terms of high radiation resistance (polyvinyltoluene (PVT) has similar properties). But owing to the fact that polystyrene is the basis for a series of low low cost and efficient plastic scintillators, studies on radiation hardness of polystyrene-based scintillators PSSC) have been the subject of many recent investigations (see review [2]). Scintillation solutes which increase radiation resistance of PSSC were reported in papers [3-6].

Radiation hardness of an organic scintillator and its recovery process are strongly influenced by conditions (temperature and presence of oxygen) under which it is irradiated . This is the reason that in the studies of the radiation resistance the samples were held not only under normal conditions (air, room temperature). For example, the samples used in the experiments reported in [3,4] were heated at 50°C during both the irradiation and recovery periods. In papers [5,6] during irradiation the samples were maintained in nitrogen atmosphere, and in [6] were held at 5°C . In present paper the study of the radiation hardness of PSSC doped with various fluors was carried out under normal conditions. One may expect that a PSSC which is radiation-resistant in air, will be also more stable in oxygen free atmosphere.

Since a decrease in the light output of small plastic samples is mainly due to a decrease in a local scintillation yield of the scintillator rather than to the reduction in the optical transmission of the sample [7], in the search of radiation- resistant PSSC the main attention was paid to the light output measurements. Moreover the optical transmission measurements of the 5-mm thick samples (which were used in this study) may give only qualitative estimation for the decrease in the light absorption lengths of long fibres and bars (tens of cm or a few meters).

1. EXPERIMENTAL

The monomer was deinhibited through a column, distilled in vacuum, placed in a cylindrical glass container, and dyes were added. Then the solution was deoxygenated, the container sealed off and the material polymerized in a silicone oil bath at 170°C for 24 hours. The material was then machined to polished disks of 25-mm diameter and 5-mm thickness.

The samples were irradiated with a ^{137}Cs source (20 krad/h) in air at room temperature. The light output measurements were made using FEU-110 photomultiplier (PM) with tri-alkaline photocathode (maximum of sensitivity at $\lambda = 480 \pm 15$ nm). The test samples were coupled to the PM (without optical contact) and exposed to a ^{90}Sr source, then the anode current of the PM was compared to that obtained for a standard PSSC (2% pTP + 0.025% POPOP). Since the scintillation yield of the PSSC can vary depending upon a set of parameters of polymer matrix such as concentration of residual monomer, molecular weight, structure of polymer chain, etc. (distribution for the scintillation yields has the width = 5-10%), one of the several samples with standard formulation was referred to as the standard sample with the 100% light output.

2. RESULTS

The experimental results are presented in tables 1-6 and in fig. 1. Formulae of the fluors used and their peak absorption and emission wavelengths are listed in the Appendix.

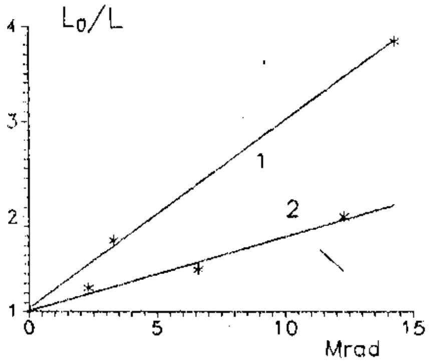

Fig. 1. Dependence of L_o/L on the total dose D [Mrad] for the sample doped with the standard fluors (2% pTP + 0.01% POPOP, sample No. 3 from table 1), curve 1, and for the sample doped with the radiation-hard secondary solute X25 (2% pTP + 0.1% X25, sample No. 3 from table 6).

First of all we have investigated the radiation resistance of PSSCs doped with well known fluors. These results are presented in Tables 1-3. Samples presented in Table 1 were irradiated to two different doses of 3.3 Mrad and 14.3 Mrad. The samples Nos. 7-10 have a PVT base. L_o is the relative light output (in percent) before irradiation, L - after irradiation. L/L_o is the light output normalized relative to the scintillator pre-irradiation value (in percent). Two last columns of Table 1 list the values of L/L_o after 3 months and after 10 months of recovery in air at room temperature. From Table 1 one finds the low radiation resistance of ultraviolet scintillators (Nos. 8-10) and samples containing pyrazolines DBP and mPDP (Nos. 11-13) or benzoxazole derivative BO (No. 14). The same conclusions can be drawn from Table 3.

The light outputs of scintillators containing only POPOP without any primary scintillation solute before and after 14.3 Mrad irradiation dose are given in Table 2. The recovery after 3 and 10 months is also shown. The absence of primary scintillation solute does not change significantly the radiation resistance of PSSC.

Table 1. Relative scintillation light outputs of polystyrene-based and polyvinyltoluene-based scintillators before (L_o) and after irradiation (L). The values of L/L_o (%) after 3 and 10 months of recovery are listed in two last columns

No	Scintillator	L_o %	L/L_o 3.3 Mrad	L/L_o 14.3 Mrad	L/L_o 3 m rec.	L/L_o 10 m rec.
1	2.0% PPO + 0.10 % POPOP	87	55	21	46	44
2	1.5% pTP + 0.01 % POPOP	87	67	23	25	21
3	2.0% pTP + 0.01 % POPOP	82	57	26	45	38
4	PVT + 2.0% pTP + 0.01% POPOP	89	73	29	45	45
5	2.0% PBD + 0.01% POPOP	103	70	32	43	42
6	1.5% PBD + 0.01% POPOP	88	65	30	43	43
7	PVT + 2% PBD + 0.01% POPOP	100	72	37	48	45
8	PVT + 2.0% PBD	68	62	24	32	26
9	PVT + 3.0% PBD	68	62	25	33	26
10	2.0% PPO	58	53	24	30	24
11	1.5% pTP + 0.01% DBP	31	49	23	17	6.5
12	1.5% PPO + 0.01% mPDP	77	48	19	27	24
13	2.0% pTP + 0.1% mPDP	90	52	20	27	23
14	1.0% BO	71	43	20	19	12

The scintillators containing BPO or PPD primary solutes have a smaller radiation resistance relative to the scintillators doped with pTP, PPO or PBD (see Table 3). Sample containing BBOT as the secondary solute has the same radiation resistance as those doped with POPOP. Samples No. 2 and 3 (table 3) with the same concentrations of the same fluors demonstrate fluctuations of radiation resistance.

Table 2. Light outputs of POPOP-containing PSSCs before and after 14.3 Mrad dose, and after 3 and 10 months of recovery

No	Scintillator	L_o %	L/L_o 14.3 Mrad	L/L_o 3 mon. recov.	L/L_o 10 mon. recov.
1	0.1% POPOP	37	36	43	37
2	1.0% POPOP	91	22	52	53

Common polymethylmethacrylate (PMMA) scintillators are known to be radiation-soft [7]. But the PMMA-based scintillator with high naphthalene (N) concentration (15% N + 0.4% POPOP) was observed[1] to have a better radiation resistance than polystyrene sample (2% pTP + 0.1% POPOP). A similar attempt to increase radiation resistance of PSSC by adding N and by increasing the fluors concentration is shown in Table 3 (Nos. 20-24). Scintillators Nos. 20, 21, 23 and 24 have relatively high radiation stability. Sample No. 22 is worse due to the presence of a pyrazoline derivative DSP.

In paper [8] for small values of the dose the following dependence of L on the dose D was found to be

$$L_O/L = 1 + K_D D, \tag{1}$$

where K_D is a degradation rate constant. Our experimental results approximately consist with eq. (1) for the dose values up to 14 Mrad, but several samples showed some deviations from linear dependence (1). Fig. 1 displays L_O/L dependence on D for standard PSSC (No. 3, table 1) and scintillator doped with X25 as the secondary solute (No. 3, table 6).

Eq. (1) is similar to the dependence of L_O on the concentration of quencher c_Q [9]. One may suppose that dependence (1) is due to the absorption of PS's or/and primary solute emission by the radiation-induced compounds (products of radiolysis) which cause fluorescent quenching, and the concentration of these compounds is proportional to the dose D.

This consideration suggests that the radiation resistance of PSSC can be improved by addition of a quencher. The quenched scintillation yield is $L_{OQ} = L_O/(1 + K_Q c_Q)$ before irradiation, and after irradiation $L_Q = L_O/(1 + K_Q c_Q + K_D D)$. Now instead of eq. (1) we obtain

$$L_{OQ}/L_Q = 1 + K_D' D, \tag{2}$$

where $K_D' = K_D/(1 + K_Q c_Q)$. In this case one can see a slowed down dependence of L_{OQ}/L_Q on D in comparison with eq. (1) because K_D is replaced with K_D' where $K_D' < K_D$.

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[1] S.A.Malinovskaya. Plastic scintillator on the base of acryl polymers. P.D. Thesis, Institute of Monocrystals, Kharkov, 1970.

Table 3. Light outputs of PSSCs for total doses of 2.3 and 10 Mrad. L_o and L - light outputs before and after irradiation. Last column - the value of L/L_o after 23 days of recovery

No	Scintillator	L_o %	L/L_o 12.3 Mrad	L/L_o 10 Mrad	L/L_o 23 d. recov
1	2.0% pTP + 0.01 % POPOP	85	50	25	50
2	1.5% pTP + 0.01 % POPOP	96	67	23	45
3	1.5% pTP + 0.01 % POPOP	94	56	19	42
4	2.0% pTP + 0.025% POPOP	98	71	23	52
5	2.0% PPO + 0.50 % POPOP	102	31	8	52
6	1.5% PPO + 0.01 % POPOP	85	42	15	43
7	0.5% PPO +.0.50 % POPOP	92	40	9	51
8	2.0% PBD + 0.01 % POPOP	95	54	13	44
9	1.5% BPO + 0.025% POPOP	84	33	8	15
10	1.0% PPD + 0.025% POPOP	75	40	13	35
11	1.5% PPD + 0.025% POPOP	74	42	19	40
12	1.5% pTP + 0.025% BBOT	86	71	22	50
13	2.0% pTP + 0.025% PDdPPS	74	51	18	37
14	0.1% 3HF	18	77	34	46
15	2.0% pTP + 0.025% dStB	45	42	15	29
16	1.5% PPO	59	29	11	28
17	1.5% pTP	43	39	18	22
18	3.0% PBD	68	36	11	32
19	2.0% BO	87	22	12	14
20	10.0% PPO + 0.5% POPOP	98	73	36	67
21	5% N + 2% pTP + 10% PPO + 0.1% POPOP	66	92	58	62
22	(21) + 0.1% DSP	84	52	36	60
23	5% N + 2% pTP + 10% PPO + 0.5% POPOP	71	93	59	69
24	5% N + 2% pTP + 10% PPO + 1.0% POPOP	72	86	57	67
25	2% pTP + 0.025% POPOQP + 1% BAP	22	55	21	48

Table 4. Scintillation light outputs and the recovery process of PSSCs before and after irradiation

No	Scintillator	L_o %	L/L_o 4 Mrad	L/L_o 4d rec.	L/L_o 10 Mrad	L/L_o 23d rec.	L/L_o 6m.1 rec.
1	2.0% pTP + 0.025 % X25	78	72	73	53	58	57
2	2.0% pTP + 0.025 % X31	85	65	70	47	55	52
3	2.0% pTP + 0.025 % POPOP	90	42	49	29	48	49
4	1.5% pTP + 0.025 % sfPDdmaP	85	40	46	27	40	36
5	1.5% PPO + 0.025 % sfPDdmaP	83	34	38	25	34	39
6	2.0% pTP + 0.025 % M-NBI	32	49	52	40	53	44
7	2.0% pTP + 0.025 % dmaNBI	27	45	41	42	48	37
8	2.0% pTP + 0.025 % Coum. 30	76	32	36	23	33	28
9	2.0% pTP + 0.025 % Coum. 7	63	34	38	23	35	29
10	2.0% pTP + 0.025% mPDP	85	40	46	24	34	30

Such possibility to improve the radiation resistance of PSSC was checked up by using benzaleacetophenone (BAP, $\lambda_{abs} = 310$ nm) as a quencher. The radiation resistance of quenched sample No. 25 (table 3) is not affected by the quencher and close to the one of unquenched sample No. 4 (table 4). This fact means that the ratio L_{OQ}/L_Q is given by

$$L_{OQ}/L_Q = 1 + K_D D, \tag{3}$$

instead of eq. (2), and hence $L_Q = L_O/[(1 + K_D D)(1 + K_Q c_Q)]$. Thus we conclude that the decrease in the scintillation yield after irradiation is mainly due to the destruction of the polymer base rather than to the quenching products of the radiolysis.

Table 5. Scintillation light outputs of PSSC before and after irradiation and the recovery process

No	Scintillator	L_o %	L/L_o 2.3 Mrad	L/L_o 5 m. rec.
1	1.5% pTP + 0.01 % POPOP	92	49	76
2	2.0% pTP + 0.025% 3HF	50	77	82
3	1.5% pTP + 0.01 % DP	97	52	69
4	1.5% pTP + 0.01 % mBIdeaBO	106	45	59
5	1.5% pTP + 0.01 % R931	95	54	71
6	1.5% pTP + 0.01 % R932	78	53	67
7	2.0% mPBD + 0.025 % POPOP	103	27	75
8	2.0% pTP + 0.01 % PEP	92	48	57
9	2.0% 1MN + 0.025 % POPOP	60	50	65
10	(9) + 0.3 % PPO	88	50	73
11	0.3% PPO + 0.025 % POPOP	80	35	63

In tables 4-6 some new PSSCs as well as known PSSCs (for the sake of comparison) are presented. Low values of L_O for samples Nos. 6 and 7 (table 4), Nos. 7 and 12 (table 6) are due to low photocathode sensitivity for the red emission of these samples. Examined best of all are X25, X31, 3HF and M3HF fluors.

From table 4 one finds low radiation resistance of PSSC containing coumarin derivatives. It should be noted that pyrazoline derivatives are not photostable, while coumarins are photostable. There is no explicit correlation between photostability and radiation resistance of a fluorescence compound.

The oxazole, oxadiazole and naphthoylen-benzimidazole derivatives exhibit the same or less radiation resistance than POPOP (see tables 4-6). Some pyrazoline and benzoxazole derivatives (DP, R931 and R932) have the radiation resistance not worse than POPOP-containing PSSC unlike other pyrazoline and benzoxazole derivatives.

Table 6. Scintillation light outputs of PSSC before and after irradiation and their recovery.

No	Scintillator	L_0 %	L/L_0 2.3 Mrad	L/L_0 6.6 Mrad	L/L_0 12.3 Mrad	L/L_0 3.5m rec.
1	2.0% pTP + 0.025% M3HF	59	77	60	41	55
2	2.0% pTP + 0.05% X25	75	81	62	43	52
3	2.0% pTP + 0.10% X25	71	80	69	50	59
4	0.1% X25	35	69	62	37	52
5	0.3% X25	37	75	66	41	51
6	2.0% butyl-PBD + 0.025% POPOP	104	43	33	20	50
7	2.0% pTP + 0.025% dmaPD-NBI	38	42	45	21	58
8	2.0% pTP + 0.025% bis-dmaP-D	82	46	34	16	31
9	2.0% pTP + 0.025% sfPOdmaP	73	60	44	24	47
10	2.0% pTP + 0.05% MI-DMAN	75	64	59	34	45
11	2.0% pTP + 0.05% omPdaPO	88	60	42	21	43
12	2.0% pTP + 0.05% DP-PNI	24	53	54	25	42

The value of decrease in transparency of the samples was measured in the following manner. The test sample was inserted between the non-irradiated sample and the photomultiplier window. Let the anode current in this case before irradiation of the test sample be I_0 and I is the anode current after irradiation. Then the value of I/I_0 is the measure of optical losses of the irradiated sample. These losses are small for doses of 2-4 Mrad, for example $I/I_0 = 97\%$ for sample No. 1 from table 5 (dose of 2.3 Mrad) while $L/L_0 = 76\%$. For larger dose of 12.3 Mrad (see table 6) $I/I_0 = 93\%$ for sample No. 1 ($L/L_0 = 55\%$), $I/I_0 = 80\%$ for sample No. 3 ($L/L_0 = 59\%$) and $I/I_0 = 80\%$ for sample No. 6 ($L/L_0 = 50\%$). Note that actually the optical losses in the light output measurement are smaller than $1-I/I_0$ because β-particles penetrate the sample (theirs ranges is 0-5 mm) and the average light path in the sample is less than that in the described above transparency measurement. But the value of $1-I/I_0$ allows one to estimate the optical losses in the sample.

An important characteristics of a scintillator is its emission decay time constant τ. The shape of the scintillation pulses of the PSSCs were measured using usual single photon counting technique. The scintillators, containing 3HF, X25 and X31 fluors were observed to have the decay constants $\tau = 7$ ns, $\tau = 12$ ns and $\tau = 10$ ns respectively. The M3HF doped scintillator has two components: $(p_1/\tau_1)\exp(-t/\tau_1) + (p_2/\tau_2)\exp(-t/\tau_2)$ with $p_1 = 0.6$, $\tau_1 = 3.5$ ns, $p_2 = 0.4$ and $\tau_2 = 9$ ns.

CONCLUSIONS

The primary scintillation solutes pTP, PPO, PBD, mPBD and bPBD showed no considerable difference in terms of radiation resistance of polystyrene scintillators doped with these fluors. Primaries BPO, BO and PPD are less stable.

Secondary solutes X25, X31, 3HF and M3HF are the most radiation-resistant among all other examined secondaries.

The radiation resistance of a scintillator can be increased by increasing the primary fluor concentration to high extent (10%) and by adding naphthalene.

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Appendix

The list of fluors used in this study:

N - naphthalene, $\lambda_{abs} \leqslant 310$ nm, $\lambda_{em} = 325$-340 nm;
1MN - 1-methylnaphthalene, $\lambda_{abs} = 320$ nm, $\lambda_{em} = 400$ nm;
pTP - para-terphenyl, $\lambda_{abs} = 290$ nm, $\lambda_{em} = 360$ nm;
PPO - 2, 5 - diphenyloxazole, $\lambda_{abs} = 310$ nm, $\lambda_{em} = 365$ mn;
PBD - 2-phenyl-5-(4-biphenylyl)-1,3,4-oxadiazole, $\lambda_{abs} = 305$ nm, $\lambda_{em} = 365$ nm;
mPBD - 2-(4'-methylphenyl)-5-(4''-biphenylyl)-1,3,4- oxadiazole, $\lambda_{em} = 365$ nm;
bPBD - 2 - (4'-t-butylphenyl) -5- (4''-biphenylyl) -1,3,4 - oxadiazole, $\lambda_{abs} = 310$ nm, $\lambda_{em} = 365$ nm;
BPO - 2-biphenylyl)-5-phenyloxazole, $\lambda_{abs} = 330$ nm, $\lambda_{em} = 390$ nm;
PPD - 2, 5 - diphenyloxadiazole, $\lambda_{abs} = 280$ nm, $\lambda_{em} = 350$ nm;
BO - 2 - (4'- dimethylaminophenyl) - benzoxazole, $\lambda_{abs} = 330$ nm, $\lambda_{em} = 400$ nm;
BAP - benzaleacetophenone (quencher, $\lambda_{abs} = 310$ nm);
POPOP - 1,4-bis-[2-(5-phenyloxazolyl)]-benzene, $\lambda_{abs} = 365$ nm, $\lambda_{em} = 420$ nm;
BBOT - 2,5-di-(tert-butyl-2-benzoxazolyl)-thiophene, $\lambda_{abs} = 380$ nm, $\lambda_{em} = 436$ nm;
dStB - 4,4'-distyrylbiphenyl, $\lambda_{em} = 430$ nm;
mPDP - 1-(o-methoxyphenyl)-3,5-diphenyl-2-pyrazoline, $\lambda_{abs} = 360$ nm, $\lambda_{em} = 460$ nm;
DSP - 1,5-diphenyl-3-styryl-2-pyrazoline, $\lambda_{abs} = 390$ nm, $\lambda_{em} = 460$ nm;

PDdPPS - 4-(5-phenyloxadiazolyl)-4'-(3,5-diphenylpyrazolinyl-1)-stilbene, λ_{abs}= 450 nm, λ_{em}= 560 nm;
X25 - naphthalic anhydride derivative, λ_{abs}= 400 nm, λ_{em}= 500 nm, τ = 12 ns;
X31 - naphthalimide derivative, λ_{abs}= 400 nm, λ_{em}= 500 nm, τ = =10 ns;
3HF - 3-hydroxyflavone, λ_{abs}= 350 nm, λ_{em}= 530 nm, τ = 7 ns;
M3HF - 2-(4'-methoxyphenyl)-3-hydroxyflavone, λ_{abs} = 350 nm, λ_{em}= = 430 nm, τ_1= 3.5 ns (60% of light), τ_2= 9 ns (40%);
sfPDdmaP - 2-(4-sulfofluorophenyl) -5- (4'-dimethylaminophenyl)-1,3,4-oxadiazole, λ_{abs}= 370 nm, λ_{em}= 495 nm;
sfPOdmaP - 2-(4-sulfofluorophenyl)-5- (4'-dimethylaminophenyl) - 1,3-oxazole, λ_{abs}= 400 nm, λ_{em}= 513 nm;
Coum. 30 - 7 - diethylamino - 3 - (3'-methylbenzimidazolyl-2') - coumarin, λ_{abs}= 420 nm, λ_{em}= 480 nm;
Coum. 7 - 7 - diethylamino - 3 - (benzimidazolyl-2') - coumarin, λ_{abs}= 460 nm, λ_{em}= 510 nm;
dmaNBI - 4-dimethylamino-1,8 -naphthoylen-1',2' - benzimidazole, λ_{em}= 600 nm;
M-NBI - 4 - morpholino - 1,8 - naphthoylen-1',2' - benzimidazole, λ_{em}= 575 nm;
PEP - 4,4'-bis-(2-(5-phenyloxazolyl))-stilbene, λ_{abs}= 385 nm, λ_{em}= 450 nm;
DP - 1,3-diphenyl-2-pyrazoline, λ_{em}= 450 nm;
mBIdeaBO - 2 - (1'-methylbenzimidazolyl-2')-6-diethylamino--benzoxazole, λ_{em}= 460 nm;
R931 - benzoxazole derivative, λ_{em}= 490 nm;
R932 - benzoxazole derivative, λ_{em}= 490 nm;
bis-dmaP-D - 2,5-bis-(4-dimethylaminophenyl)-oxadiazole, λ_{abs}= =350 nm, λ_{em}= 400 nm;
dmaPD-NBI - 5-(4-dimethylaminophenyl)-2-(1,8-naphthoylen-1', 2'-benzimidazolyl-5)-oxadiazole, λ_{abs}= 440 nm, λ_{em}= 565 nm;
MI-DMAN - 4 - dimethylaminonaphthalic acid N-methylimide, λ_{em}= = 480 nm;
cmPdaPO - 2-(4'-carbomethoxyphenyl)-5-(4''-dimethylaminophenyl)-oxazole, λ_{abs}= 353 nm, λ_{em}= 460 nm;
DP-PNI - 4-(1,5-diphenyl-2-pyrazolinyl-3)-N-phenylnaphthalimide λ_{em}= 580 nm.

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