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Polishing of polystyrene scintillators

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As a result of research in regularities of influence of material's physical properties and the dispersion system on the polishing performance and roughness of polished surfaces of polystyrene-based plastic scintillators, it was found that the formation of slurry particles of the treated material is a consequence of the Förster resonant energy transfer that occurs in an open resonator formed by two parallel surfaces of the treated material and lapping pad, between the energy levels of polishing powder particles and treated material in the four-mode regime. It is shown that the material removal rate is determined by the total coefficient of volumetric wear, the total lifetime of treated surface clusters in an excited state and the resulting resonator quality factor at all possible frequencies. Roughness of polished surface depends on the spectral resolution and the resonator quality factor and is characterised by a superposition of the parameters Ra, Rq and Rmax, which are specific to each mode. It is shown that polishing of polystyrene-based plastic scintillators using a dispersion system of micro- and nanoparticles allows to reach the values of material removal rate and the roughness of polished surfaces, which satisfy the requirements for the polishing process of optical surfaces.

Keywords: polishing, Förster resonance energy transfer (FRET), material removal rate (MRR), roughness.

Полірування сцинтиляторів з полістиролу *Ю.Д. Філатов, А.Ю. Бояринцев, В.І.Сідорко, С.В. Ковальов, О.В. Колесніков*

В результаті дослідження закономірностей впливу фізичних властивостей оброблюваного матеріалу та дисперсної системи на продуктивність полірування і шорсткість полірованих поверхонь сцинтиляторів з полістиролу встановлено, що утворення частинок шламу оброблюваного матеріалу є наслідком ферстерівського резонансного перенесення енергії, яке відбувається у відкритому резонаторі, утвореному двома паралельними поверхнями оброблюваного матеріалу і притира, між енергетичними рівнями частинок полірувального порошку і оброблюваного матеріалу в чотирьохмодовому режимі. Показано, що швидкість зняття оброблюваного матеріалу визначається сумарним коефіцієнтом об'ємного зносу, сумарним часом життя кластерів оброблюваної поверхні у збудженому стані і результуючою добротністю резонатора на всіх можливих частотах. Показано, що під час полірування сцинтиляторів з полістиролу за допомогою дисперсної системи з мікро- і наночастинок досягаються швидкість зняття оброблюваного матеріалу і шорсткість полірованих поверхонь, які задовольняють вимогам, що висуваються до процесу полірування оптичних поверхонь.

1. Introduction

Traditionally, the polishing of precision surfaces of optoelectronic components and optical systems made of non-metallic materials has been carried out using polishing dispersion systems of non-abrasive micro- and nanopowders. The material removal rate depends on the rheological properties of the dispersion system, the structure of the treated material and

its dielectric and spectroscopic properties [1-3]. An increase in polishing performance is only possible by improving and developing of new polishing dispersion systems, which would ensure sufficient speed of material removal and formation of the required nanoprofile of the polished surface in the absence of any defects on it. Therefore, the study of the regularities of plastic scintillators polishing using micro- and nanopowder dispersion systems is so actual.

The aim of this study is to investigate the regularities of influence of physical properties of treated materials and the dispersion system on the polishing performance and the roughness of polished surfaces of polystyrene scintillators.

2. Methodology

According to the cluster model of polishing of non-metallic materials [1] and the Deryaguin–Landau–Verwey–Overbeek theory [4, 5], the interaction between the workpiece surfaces and the polishing powder particles during polishing is determined by the Van der Waals intermolecular interaction forces, which result from the emergence of local dipole moments at characteristic frequencies. The energy transfer between particles of the dispersed phase and the treated surface is based on Förster resonance energy transfer (FRET) mechanism, and the energy transfer from excited donor to an acceptor occurs without intermediate photon emission [6–11]. Resonance energy transfer in a dispersion system located in an open resonator formed by two parallel surfaces of the workpiece and the lap, in contrast to conventional FRET [12, 13] and the “carnival effect”, which means the reverse energy transfer when donor and acceptor are reversed [14–16], occurs when molecular fragments of the particles of the dispersed phase and the workpiece are both donors and acceptors at the same time [17]. In this case, the FRET efficiency depends on the spectral resolution $\delta\nu = \nu_2 - \nu_1$ and the resonator quality factor $q = \nu_1/(\nu_2 - \nu_1)$ (ν_1, ν_2 are the natural vibrational frequencies of molecular fragments on the treated surface and the surface of the polishing powder particle, respectively), and the material removal rate (polishing productivity) is determined in accordance with the formula [17–19].

$$Q = \eta L_t \frac{\tau}{t_c} q \qquad (1)$$

where η is the volumetric wear coefficient; L_t is the friction path length of polishing powder particle on the workpiece; t_c is the contact time of the polishing powder particle with the surface to be treated; u is the relative velocity between the workpiece and lapping tool, τ is the lifetime of the workpiece clusters in an excited state.

The lifetime of treated surface clusters in an excited state is inversely proportional to the probability of dipole transitions at a frequency of ν_2 and is determined by the formula [17].

$$\tau = \frac{3N_m}{8\pi c\alpha^3\nu_2}; \qquad (2)$$

where N_m is the number of possible transitions, $\alpha = \frac{e^2}{\hbar c} = \frac{1}{137}$ – the fine-structure constant; ; $\hbar$ = 1,054·10^{-34} J·s – the Planck’s constant; e – the electron charge; c – the speed of light.

According to the theory of molecular vibrations, the number of possible transitions between the quantum energy levels of the treated surface clusters and the polishing powder particle depends on the number of levels and their degeneracy. The number of possible transitions is determined according to the formula $N_m = k_m N_{0m}$; (where N_{0m} is the number of energy levels; $k_m = \prod_{n=1}^{f} \frac{(n+g-1)!}{n!(g-1)!} = 6$ – is the multiplicity of level degeneracy; $f = 2$ and $g = 2$ are the number of freedom degrees of oscillatory motion of treated surface clusters and the number of normal oscillations, respectively).

The volumetric wear coefficient depends on the size $d_1(i)$ of slurry particles and is determined by the formula $\eta = \sum_i \frac{d_1(i)^2}{4\beta(i)t_c}$, where the values of $\beta(i)$ are solutions of the system of transcendental equations [1, 20].

$$\frac{\exp(-\beta(i)^2)}{\sqrt{\pi}\, erf(\beta(i))} = \frac{1}{N(i)\, S_i}\sqrt{L_t d_2 S_c \vartheta}, \qquad (3)$$

where $N(i)$ is the concentration of the ith slurry particle on treated surface; S_i is the surface area of the ith slurry particle; d_2 is the average size of polishing powder particle; S_c is the contact area of the workpiece with the lapping tool surface; $\vartheta = \frac{\lambda T L_t}{p_a u S_c}$; is a dimensionless parameter; λ is the thermal conductivity of the material being treated; T is the temperature in the contact zone between the workpiece and the lapping tool; p_a is the pressure of the workpiece against the lapping tool.

The number of the *i-th* slurry particles on treated surface $N(i)$, which depends on the potential W of the interaction of the polishing powder particle with the treated surface, as well as on the function of the distribution of slurry par-

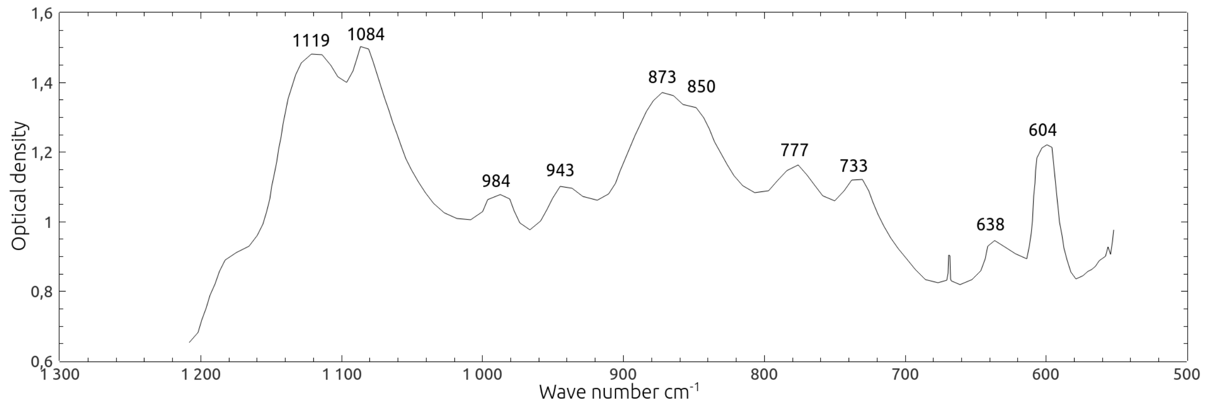

Fig. 1. Infrared absorption spectrum of the polishing powder

ticles on the surface area $P(i)$, is determined in accordance with the formula [1, 4, 5, 21].

$$N(i) = \frac{2W}{\pi \hbar S_c} P(i)\,. \qquad (4)$$

The polishing of flat surfaces of polystyrene scintillators with a 60 mm diameter was carried out on the 2ShP-200M machine using a 100 mm diameter polyurethane foam lapping pad at a pressure of pressing the workpiece against the lapping pad of 17.7 kPa, a lapping pad rotation speed of 90 rpm, a stroke offset of 30 mm, a stroke length of 80 mm and an average temperature in the contact zone between the workpiece and the lapping pad of 298 K. For the polishing of polystyrene scintillators (density 1.06 g/cm^3, thermal conductivity 0.12 W/(m·K)), a dispersion system of micro- and nanopowders was used,which was characterised by the frequencies of natural vibrations of molecular fragments of the polishing powder particles determined by IR absorption spectra (Nicolet 6700 Fourier spectrometer): 550, 597, 604, 638, 670, 733, 777, 850, 873, 943, 984, 996, 1084, 1119 (cm^{-1}) (Figure 1). The typical frequencies for polystyrene are 537, 694, 753, 905, 1027 (cm^{-1}) [22–25]. The size of polishing powder particles was determined from the images (Fig. 2) obtained using a Zeiss-EVO 50 Scanning Electron Microscope with an AZtec Microanalysis System. The images show that the maximum length of polishing powder particles is lm = 5.1 μm and their thickness varies from 262.0 nm to 555. 9 nm with an average polishing powder particle size of d_2 = 409 nm. The value of ellipticity is estimated by the value of the angle of curvature of the generating line $\alpha_к = 2(\pi - 2\mathrm{arctg}(d_2/l_m)$ and is between 333,4^o до 341,6^o, indicating the needle-like shape of particles. The removal of treated material was determined by the weight method using an AVD-200 analytical balance in mg/30 min. Roughness parameters of polished surfaces were determined by computer modelling and monitored using a Micron-alpha non-contact interference 3D profilograph. The length of the friction path of the polishing powder particle on the treated surface was L_t = 188 mm, and the speed of relative velocity of the workpiece and lapping tool, averaged over the kinematic parameters of polishing (offset and stroke length), was u = 0.47 m/s. The contact time of the polishing powder particle with the treated surface was $t_c = d_2/u$ = 0.88 μs.

3. Experiment

Given the functional characteristics of the detector, polishing is a way of obtaining certain reflective properties from the detector surface. The condition of the surface has a major influence on light collection processes. Light reflection conditions, in particular diffuse or specular reflection, have a complex effect on the detector's technical parameters. In this work, we have carried out an experimental comparison of the above-mentioned polishing technology using a dispersion system of micro- and nanoparticles (Type 1) with the traditional polishing technology (Type 2), which involves the use of cerium-based micro-abrasive powders.

For comparison, detectors of different shapes and sizes were polished: small scintillators of 10x10x10 mm [34], large detectors of 50x250x1000 mm, ∅ 50 x 1000 mm [35] and organic light guides [36].

All detector types were excited by ^{241}Am and ^{137}Cs sources.

4. Results and discussion

Polystyrene polishing by lapping method, when the system "workpiece surface-dispersion system-lapping surface" is located in an open resonator formed by the contacting surfaces of

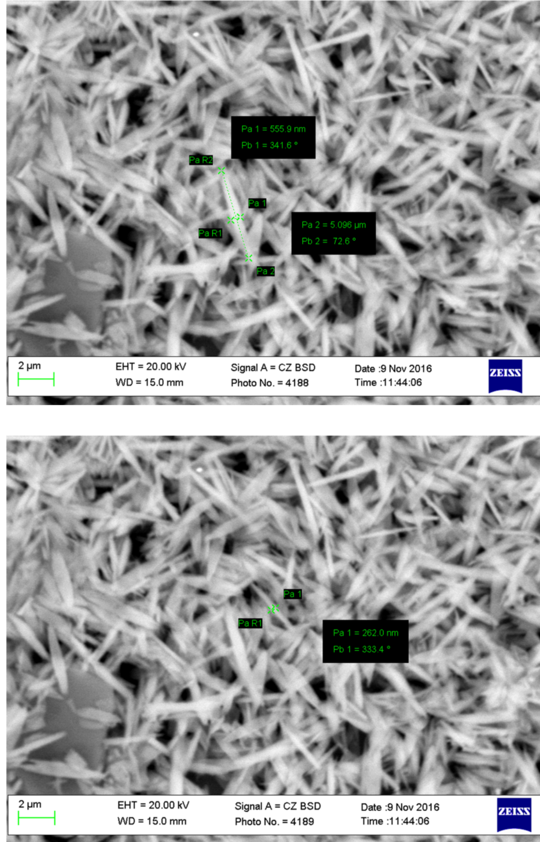

Fig. 2. Size of polishing powder particle

the workpiece and the lapping surface, in the absence of photon generation, as in the case of FRET lasers [26-31], generates slurry nanoparticles of the treated material, wear nanoparticles of polishing powder and wear nanoparticles of lapping material [19], the power of which determines the rate of material removal, the intensity of polishing powder wear and lapping material wear. The removal of treated material is a result of quantum transitions between energy levels of polishing powder particles and treated material according to the scheme shown in Fig. 3. Slurry particles are formed as a result of FRET from polishing powder particles to treated surface, which occurs in the four-mode regime under transitions between energy levels, which are characterised by frequencies: v_{211} = 1084 sm^{-1} → v_{14} = 1027 sm^{-1}, v_{29} = 943 sm^{-1} → v_{13} = 905 sm^{-1}, v_{26} = 777 sm^{-1} → v_{12}=753 sm^{-1} i v_{25} =733 sm^{-1} → v_{11} = 694 sm^{-1}.

The FRET efficiency at each of the transitions, which depends on the spectral resolution δv_i, the resonator quality factor q_i and the lifetime τ_i of treated surface clusters in an excited state, determines the size and number of slurry nanoparticles, and hence the material removal rate and roughness of the polished surfaces

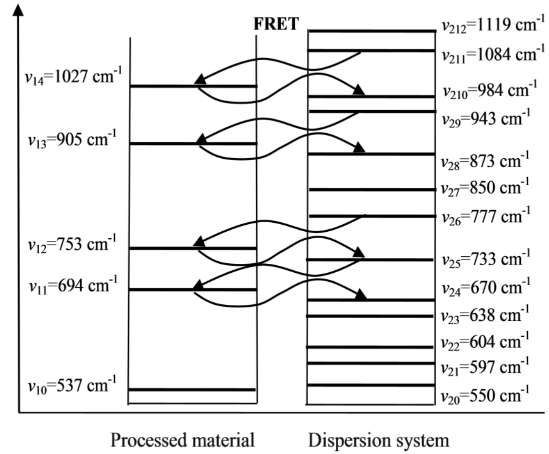

Fig. 3. Diagram of quantum transitions between energy levels of polishing powder particles and treated material

[17–19]. The calculated values of parameters of interaction between the treated surface and the dispersion system as well as the calculated values of slurry nanoparticles characteristics and the polystyrene polishing parameters are shown in the table.

The lifetime of treated surface clusters in an excited state for each mode was determined according to formula (2), taking into account the number of energy levels and the multiplicity of their degeneracy. The total lifetime was $\tau = \sum_i \tau_i$ = 524 ns. The resulting resonator Q-factor for a multimode system with a discrete spectrum of natural frequency was determined as $q = \left[\sum_i q_i^{-1}\right]^{-1}$ = 5.39. During polystyrene polishing in an open resonator, the energy and size of the slurry nanoparticles, as well as their concentration, depend on the spectral separation, the resonator Q-factor and determine the volumetric wear coefficients corresponding to different quantum transitions between the polishing powder particle and the treated surface, which are calculated according to Eqs. (3) and (4). The total volumetric wear coefficient, which determines the material removal rate, was $\eta = \sum_i \eta_i = 6{,}9 \cdot 10^{-12}$ m²/s. The value of the material removal rate during polishing of polystyrene scintillators calculated according to formula (1), which is $Q = 41.6 \cdot 10^{-13}$ m³/s, agrees well with the data of the experimental determination of the polishing performance (7.8±0.3) mg/30 min (5.2 µm/hour, or $40.9 \cdot 10^{-13}$ m³/s) with a deviation of 2%. This indicates the validity of considering the process of formation and removal of slurry nanoparticles from the treated surface as a consequence of FRET in the multimode regime.

Table 1: Polishing parameters of polystyrene-based plastic scintillators.

Parameter	Transitions FRET			
	$\nu_{211} \to \nu_{14}$	$\nu_{29} \to \nu_{13}$	$\nu_{26} \to \nu_{12}$	$\nu_{25} \to \nu_{11}$
Mode i	1	2	3	4
Spectral resolution $\delta\nu_i$, sm^{-1}	57	38	24	39
Resonator quality factor q_i	18,0	23,8	31,4	17,8
Lifetime of treated surface clusters in an excited state τ_i, ns	113	141	158	112
Particle surface area S_i, nm^2	80	123	175	80
Size of slurry particle $d_1(i)$, nm	4,5	5,6	6,6	4,5
Interaction potential of polishing powder particle with the treated surface W_i, zJ	0,114	0,076	0,048	0,078
Particle concentration $N(i)$, 10^{14} m^{-2}s^{-1}	1,8	1,2	0,8	1,2
Volumetric wear coefficient η_i, 10^{-12} m^2/s	1,1·	1,8	2,7	1,3
Material Removal Rate				
Q, 10^{-13} m^3/s	41,6			
Experiment: mg/min.(μm/h.)	0,26 (5,2)			
10^{-13} m^3/s	40,9 ·			
Calculation error, %	2			
Roughness parameters of polished surface, nm				
Ra_i	9,1±0,2	11,2±0,1	12,8±0,5	8,8±0,3
Rq_i	9,6±0,3	12,4±0,3	13,6±0,7	9,6±0,4
$R\max_i$	15,5±2,4	21,0±1,5	22,7±2,9	16,8±1,6
Ra = 10,1±0,2 nm; Rq = 10,9±0,4 nm; $Rmax$ = 18,2±2,1 nm				

The roughness parameters of polished surface (arithmetic mean Ra and root mean square Rq profile deviation, maximum height of profile $Rmax$), which are determined by computer modelling [3, 32] and depend on the size and concentration of slurry nanoparticles, as well as on the surface area distribution function $P(i)$, increase proportionally with increase in spectral separation between the treated material and the polishing powder particle and with improvement in resonator quality factor formed by the surfaces of the treated material and lapping pad [33]. In this regard, it can be assumed that slurry nanoparticles of size $d_1(i)$ are formed from the polishing powder particles to the treated surface as a result of the FRET, which occurs during the transitions between the energy levels and that are characteristic of each of the modes. The roughness of treated surface is characterised by the parameters Ra_i, Rq_i and $Rmax_i$. The resulting roughness of the polished surface is described by the parameters $Ra = \frac{1}{\sum_i N(i)} \sum_i N(i) Ra_i$ = 10,1 nm, $Rq = \frac{1}{\sum_i N(i)} \sum_i N(i) Rq_i$ = 10,9 nm and $R\max = \frac{1}{\sum_i N(i)} \sum_i N(i) R\max_i$ = 18,2 nm. The errors of their determination $\delta R = \frac{1}{\sum_i N(i)} \sum_i N(i) \delta R_i$ are 0.2 nm, 0.4 nm and 2.1 nm, respectively. Thus, the polished surfaces of polystyrene scintillators, whose roughness is characterised by the parameters Ra = 10.1±0.2 nm, Rq = 10.9±0.4 nm and $Rmax$ = 18.2±2.1 nm, meet the requirements for optical surfaces.

The most significant effect of the polishing process can be seen in the long-size detectors. Thus, the increase in light output for low energy radiation (241Am) on detectors with polishing type 1 (Type 1) compared to polishing type 2 (Type 2) is 14%. For 662keV the increase in light output is 31% (Fig. 4, Table 2). For 10*10*10 mmcb detectors, the increase in light output does not exceed 5%. In our opinion, the greater influence of polishing on long-size detectors is explained by the peculiarities of light collection, namely, a large number of reflections from surfaces.

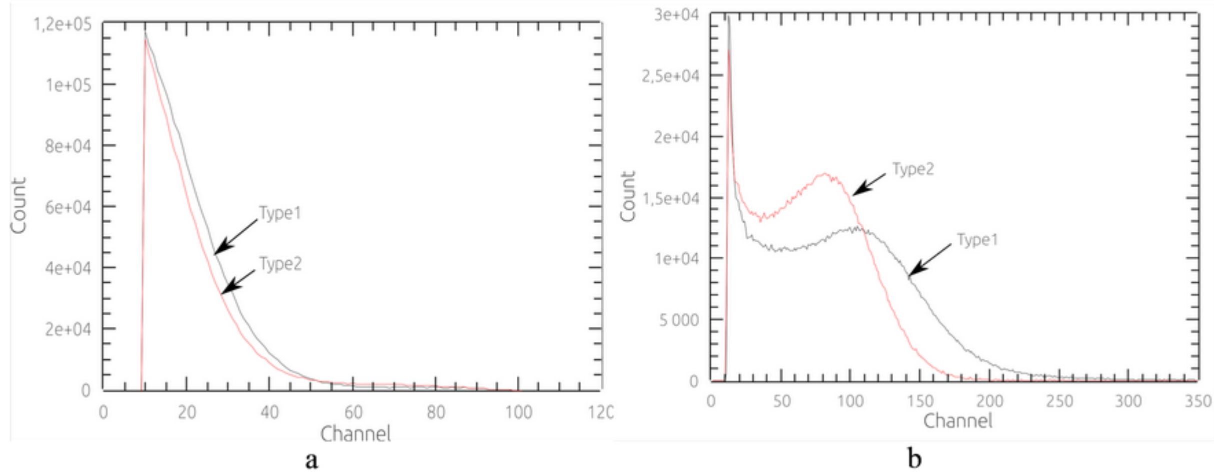

Fig. 4. Amplitude spectra of the 50x250x1000 mm cubes detector using polishing type 1 and polishing type 2 under excitation by ^{241}Am (a) and ^{137}Cs (b).

Table 2: Ratio of light output of different geometry detectors polished with a dispersion system of micro- and nanoparticles and a detector polished with traditional technology C_{Type1}/C_{Type2} for different radiation sources

Detector	Light output ratio C_{Type1}/C_{Type2} for sources	
	^{241}Am	^{137}Cs
50mm x 250mm x 1000mm	1.14	1.31
∅ 50mm x 1000mm	1.17	1.29
10mm x 10mm x 10mm	1.03	1.05

4. Conclusions.

As a result of investigation of the mechanism of precision surfaces polishing of polystyrene-based plastic scintillators by means of dispersion systems of micro- and nanoparticles of polishing powders it has been established that the formation of slurry particles of the treated material is a consequence of FRET, which occurs in an open resonator formed by two parallel surfaces of the treated material and the lapping pad, between the energy levels of polishing powder particles and the treated material in the four-mode regime.

It is shown that the material removal rate is determined by the total coefficient of volumetric wear, the total lifetime of treated surface clusters in an excited state and the resulting resonator quality factor of the system " workpiece surface–dispersion system–lapping surface" at all possible frequencies of the discrete spectrum.

It is established that roughness of polished surface depends on the spectral separation between the material to be treated and the polishing powder particle, as well as on the resonator quality factor at all allowed frequencies. Roughness is characterized by the parameters *Ra*, *Rq*, and *Rmax*, which are a superposition of these characteristics for each of the possible modes.

It has been shown that polishingof polystyrene-based plastic scintillators using a dispersion system of micro- and nanoparticles achieves the material removal rate and the roughness of polished surfaces that meet the requirements for the polishing process of optical surfaces.

It has been determined that the use of polishing technology for polishing polystyrene scintillators using a dispersion system of micro- and nanoparticles increases the light output of long-size detectors. At the same time, both technologies demonstrate comparable results for small detectors.

References

1. Yu.D.Filatov, *J. Superhard Mater*. **42**(1), 30 (2020).
2. Yu.D.Filatov, V.I.Sidorko, S.V.Kovalev et al., *J. Superhard Mater*. **43**(1), 65 (2021).
3. O.Yu.Filatov, V.I.Sidorko, S.V.Kovalev et al., Functional Materials, **23**(1), 104 (2016).
4. N.Sato, Y.Aoyama, J.Yamanaka et al., *Sci. Reports,* **7**(6099), 1 (2017).
5. G.Lin, D.Guo, G.Xie et al., *Colloids Surf. A: Physicochem. Eng. Aspects*., **482**, 656 (2015).

6. G.A.Jones, D.S.Bradshaw, *Front. Phys.* **7**(100), 1 (2019).
7. A.Singldinger, M.Gramlich, C.Gruber et al., *ACS Energy Lett.*, **5**, 1380 (2020).
8. Ch.Abeywickrama, M.Premaratne, D.L.Andrews, *Proc. SPIE,* **113451B**, (2020).
9. A.N.Poddubny, A.V.Rodina, *J. Exp. Theor. Phys.* **122**(3), 531 (2016).
10. G.A.Jones, D.S.Bradshaw, *Front. Phys.* **7**, 100 (2019).
11. L.C.Cristian, J.Zubin, *Optics Express*. **26**(15), 19371 (2018).
12. F.Gordon, S.Elcoroaristizabal, A.G.Ryder, *Biochimica et Biophysica Acta (BBA)-General Subjects*. **1865**(2), 129770 (2021).
13. M.Du, L.A.Martinez-Martinez, R.F.Ribeiro et al., *Chem. Sci.* **9**, 6659 (2018).
14. X.Zhong, T.Chervy, L.Zhang et al., *Angew. Chem. Int.* **56**(31), 9034 (2017).
15. D.Dovzhenko, M.Lednev, K.Mochalov et al., *Chem. Sci.* **12**, 12794 (2021).
16. I.Nabiev, *PhysBioSymp*. 2019. *J. Physics: Conf. Ser.* **2058**, 012001 (2021).
17. Yu.D.Filatov, V.I.Sidorko, A.Y.Boyarintsev et al., *J. Superhard Mater.* **44**(2), 117 (2022).
18. Yu.D.Filatov, V.I.Sidorko, A.Y.Boyarintsev et al., *J. Superhard Mater.* **44**(5), 358 (2022).
19. Yu.D.Filatov, *J. Superhard Mater.* **45**(2), 140 (2023).
20. Yu.D.Filatov, V.I.Sidorko, *J. Superhard Mater.* **27**(1), 53 (2005).
21. Yu.D.Filatov, V.I.Sidorko, A.Y.Boyarintsev et al., *J. Superhard Mater.* **44**(4), 276 (2022).
22. J.F.Fang, Y.M.Xuan, Q.Li, *Sci. China Technol. Sci.* **53**(11), 3088 (2010).
23. F.H.Al-K.Mahasin, S.R.Zahraa, R.S.Sanaa, *J. Radiat. Res. Appl. Sci.* **9**(3), 321 (2016).
24. L.Qionglin, Z.Shunqin, S.You et al., *RSC Adv.* **8**, 17151 (2018).
25. V.Herman, H.Takacs, F.Duclairoir et al., *RSC Adv. J. Name.* 63, 51371 (2015).
26. Q.Chan, Z.Chunhuan, Z.Zhonghao et al., *CCS. Chemistry*. **4**(1), 250 (2022).
27. J.Saha, D.Dey, A.D.Roy et al., *J. Luminescence.* **172**, 168 (2016).
28. J.A.Rivera, K.V.Desai, J.G.Eden, *AIP Advances*. **11**, 125033 (2021).
29. L.Cerdán, E.Enciso, V.Martín et al., *Nature Photonics*. **6**(9), 621 (2012).
30. Q.Chen, A.Kiraz, X.Fan, *Proc. SPIE*. **9725**, 97250I–1 (2016).
31. M.Aas, Q.Chen, A.Jona´s et al., *IEEE J. Select. Topics Quantum Electron.* **22**(4), 7000215 (2016).
32. O.Yu.Filatov, V.I.Sidorko, S.V.Kovalev et al., *J. Superhard Mater.* **38**(3), 197 (2016).
33. Yu.D.Filatov, V.I.Sidorko, S.V.Sokhan' et al., *J. Superhard Mater.* **45**(1), 54 (2023).
34. A. Boyarintsev, A. De Roeck, S. Dolan et al., *JINST* **16** P12010 (2021).
35. A. Yu. Boyarintsev, A.V.Kolesnikov, S.N. Kovalchuk et al., *Functional Materials* **28**(4) (2021)
36. A. Yu. Boyarintsev , N.Z. Galunov , B.V. Grinyov et al., *NIMA* **930** (2019).