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Degradation of the optical properties of ZnO micropowder modified with SiO₂ nanoparticles under sequential irradiation with electrons, protons, and solar spectrum quanta

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The degradation of the integrated emissivity coefficient Δa_s of $m\text{ZnO}$ micropowder modified with $n\text{SiO}_2$ nanoparticles ($m\text{ZnO}/n\text{SiO}_2$) was studied under sequential irradiation with 30 keV electrons (e^-), 5 keV protons (p^+), and solar spectrum quanta (SSP). The additivity coefficient (Cadd) was calculated to quantify the mutual influence of different types of radiation. It was found that, depending on the type of radiation and the irradiation sequence, Cadd can be less than, equal to, or greater than unity.

Keywords: zinc oxide, silicon dioxide, modification, irradiation, electrons, protons, solar spectrum quanta, additivity coefficient.

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Introduction

Thermal control coatings (TCC) based on zinc oxide pigments represent one of the most common classes of materials for spacecraft thermal control systems. Their widespread application is attributed to the combination of a low integrated solar absorptance ($a_s = 0.15\text{--}0.25$) and a high infrared emittance $\varepsilon > 0.85$, which provides efficient radiative heat exchange with the environment [1, 2]. The stability of these characteristics throughout the spacecraft's active lifetime is a key requirement in the design of thermal control systems [3].

In actual space flight conditions, TCCs are exposed to a combination of space environmental factors. The main destabilizing agents in the geostationary orbit are fluxes of electrons and protons from the Earth's outer radiation belt with energies ranging from several to hundreds of keV, as well as solar ultraviolet radiation [4, 5]. In low Earth orbit, the impact of atomic oxygen is added

to the above factors. All these factors induce an increase in a_s , which causes heating of the spacecraft surface and disruption of the thermal balance. Predicting such degradation is a mandatory stage in spacecraft design [3].

Modification of $m\text{ZnO}$ micropowder with $n\text{SiO}_2$ nanoparticles is an effective method for increasing the radiation resistance of TCCs [6, 7]. It has been established that the deposition of $n\text{SiO}_2$ onto the surface of $m\text{ZnO}$ particles reduces the density of intrinsic defects in zinc oxide, which are centers of radiation-induced absorption, and forms a protective layer that partially shields corpuscular radiation [8]. As a result, TCCs based on $m\text{ZnO}/n\text{SiO}_2$ exhibit a significantly smaller increase in a_s under electron and proton irradiation compared to the unmodified zinc oxide powder [9, 10].

However, ground-based simulation of flight conditions is traditionally performed using separate exposure to individual factors, and the resulting degradation is estimated as the sum of

separate contributions. This approach fails to consider the possible mutual influence of factors on each other. For a quantitative assessment of the deviation of the combined effect from simple additivity, the additivity coefficient (K_{add}) is used.

The aim of this paper is to investigate K_{add} for modified $m\text{ZnO}/n\text{SiO}_2$ powders under sequential exposure to electrons, protons, and solar ultraviolet radiation, and to establish the physical mechanisms of the observed effects.

Study Object and Experimental Procedure

The object of study was a zinc oxide micropowder. Silicon dioxide nanopowder was used as the modifying additive. The $m\text{ZnO}$ micropowder was mixed with the $n\text{SiO}_2$ nanoparticles in a 100:3 ratio.

For the modification process, the $n\text{SiO}_2$ was dissolved in distilled water under ultrasonic agitation, then $m\text{ZnO}$ was added and the mixture was stirred with a magnetic stirrer. The mixture was dried, ground, and heated in air at the optimal temperature [11].

The integrated solar absorptance (a_s) was calculated from the diffuse reflectance spectra according to equation (1) [12–14]:

$$a_s = 1 - R_s = \frac{\int_{\lambda_1}^{\lambda_2} \rho_{\lambda} \cdot I_{\lambda} \cdot d_{\lambda}}{\int_{\lambda_1}^{\lambda_2} I_{\lambda} \cdot d_{\lambda}}, \quad (1)$$

where R_s is the integrated reflectance of solar radiation, I_{λ} is the solar spectrum, and $(\lambda_1 - \lambda_2)$ is the wavelength range of the solar spectrum used for integration, equal to 0.2–2.5 μm .

The change in the absorption coefficient a_s was calculated from its values before and after irradiation. For each irradiation condition, the additivity coefficient K_{add} was calculated by equation (2) [15, 16]:

$$K_{\text{add}} = \frac{\sum(\Delta a_{s1} + \Delta a_{s2})}{\Delta a_{s(\text{COBM})}}, \quad (2)$$

where $\sum(\Delta a_{s1} + \Delta a_{s2})$ is the sum of changes under separate irradiation; $\Delta a_{s(\text{COBM})}$ is the change in value under simultaneous irradiation.

Experimental Results and Their Discussion

Table 1 contains the experimental values of K_{add} for the $m\text{ZnO}/n\text{SiO}_2$ powder under sequential irradiation by protons and solar spectrum quanta: $p^+ \rightarrow \text{solar spectrum quanta}$.

Table 1

Values of K_{add} for the $m\text{ZnO}/n\text{SiO}_2$ powder after irradiation by protons and solar spectrum quanta

Sequential irradiation $p^+ \rightarrow \text{solar spectrum quanta}$ (Φp^+ , cm^{-2} ; t_{SSQ} , h)					
$\Phi p^+ \rightarrow t_{\text{SSQ}}$	0.63×10^{16}	1.08×10^{16}	2.09×10^{16}	3×10^{16}	4.07×10^{16}
Δa_s	0.039	0.060	0.084	0.107	0.124
$\Sigma \Delta a_{s(\text{sep.})}$	0.028	0.043	0.068	0.093	0.113
K_{add}	0.71	0.72	0.82	0.87	0.91
$\Phi p^+ \rightarrow t_{\text{SSQ}}$	$4.07 \times 10^{16} \rightarrow 2$	$4.07 \times 10^{16} \rightarrow 4$	$4.07 \times 10^{16} \rightarrow 6$	$4.07 \times 10^{16} \rightarrow 8$	-
Δa_s	0.138	0.145	0.152	0.158	-
$\Sigma \Delta a_{s(\text{sep.})}$	0.129	0.133	0.138	0.145	
K_{add}	1.05	1.06	1.07	1.10	

1. Sequential irradiation $p^+ \rightarrow \text{solar spectrum quanta}$ was carried out in the following mode:

– First, separate proton irradiation was performed at fluences from $0.63 \times 10^{16} \text{ cm}^{-2}$ to $4.07 \times 10^{16} \text{ cm}^{-2}$, and the values of Δa_s were recorded;

– then, after the proton irradiation, the samples were sequentially exposed to solar spectrum quanta, and Δa_s values were recorded after each irradiation period;

– then, separate irradiation by solar spectrum quanta was performed for 2, 4, 6, and 8 hours;

– next, the sum of changes in the absorption coefficient under separate irradiation by protons and solar spectrum quanta was calculated as $\Sigma \Delta a_{s(\text{sep.})}$;

2. finally, K_{add} was calculated from values $\Sigma \Delta a_{s(\text{sep.})}$ and values Δa_s obtained under sequential irradiation $\Delta a_{s(\text{seq.})}$, which were close to unity.

Sequential irradiation solar spectrum quanta $\rightarrow p^+$ was carried out in the following mode:

– first, separate irradiation by solar spectrum quanta was performed for 6 hours, yielding $\Delta a_s = 0.069$;

– then, sequential irradiation by protons at fluences from $0.52 \times 10^{16} \text{ cm}^{-2}$ to $3.93 \times 10^{16} \text{ cm}^{-2}$ was performed, yielding Δa_s from 0.094 to 0.164;

– the sum of changes of Δa_s after separate irradiation by solar spectrum quanta and protons in the same conditions is significantly lower than the values obtained under sequential irradiation: with proton fluence increasing to $3.93 \times 10^{16} \text{ cm}^{-2}$ $\Sigma \Delta a_{s(\text{sep.})}$ increases from 0.049 to 0.135, but remains significantly lower than $\Sigma \Delta a_{s(\text{seq.})}$.

As a result, K_{add} has values of 0.8 over the entire studied range from $\Phi_1 = 0.52 \times 10^{16} \text{ cm}^{-2}$ to $\Phi_2 = 3.93 \times 10^{16} \text{ cm}^{-2}$; its values are less than unity, which indicates the presence of synergistic effects.

3. Sequential irradiation $e^- \rightarrow$ solar spectrum quanta was carried out in the following mode:

– first, irradiation by electrons at a fluence of $7 \times 10^{16} \text{ cm}^{-2}$ was performed, yielding $\Delta a_s = 0.04$;

– then, irradiation by solar spectrum quanta was performed sequentially after electrons for 2 and 4 hours, yielding Δa_s values equaling to 0.086 and 0.103, respectively;

– then, separate irradiation by solar spectrum quanta was performed for 2 and 4 hours, yielding Δa_s values equaling to 0.044 and 0.057, respectively.

The sum of changes of Δa_s under separate irradiation by electrons and solar spectrum quanta was 0.084 and 0.097, while under sequential irradiation $e^- \rightarrow$ solar spectrum quanta it was 0.086 and 0.103, respectively, and K_{add} was **0.977 and 0.941**. Its values are close to unity; no synergistic effects are observed, and in ground testing of this pigment, sequential irradiation can be replaced by separate irradiation by electrons and solar spectrum quanta.

Sequential irradiation solar spectrum quanta $\rightarrow e^-$ was carried out in the following mode:

– first, irradiation by solar spectrum quanta was performed for 6 hours, yielding $\Delta a_s = 0.067$;

– then, sequential irradiation by electrons at fluences of 1×10^{16} and $2 \times 10^{16} \text{ cm}^{-2}$ was performed, yielding $\Delta a_s = 0.059$ и 0.064, respectively;

– finally, separate irradiation by electrons at the same fluences of 1×10^{16} and $2 \times 10^{16} \text{ cm}^{-2}$ was performed, yielding $\Delta a_s = 0.017$ и 0.024, respectively.

The sum of changes of Δa_s under the separate irradiation by solar spectrum quanta and electrons was 0.084 and 0.091, and K_{add} was 1.43 and 1.42. These values are significantly higher than unity, indicating the presence of synergistic effects under this sequential irradiation mode. Solar spectrum quanta $\rightarrow e^-$, and in ground-based testing of this pigment, sequential irradiation cannot be substituted by separate irradiation.

Conclusion

The degradation of the integrated solar absorbance Δa_s of the mZnO micropowder modified with nSiO₂ nanoparticles (mZnO/nSiO₂) was studied under sequential irradiation by electrons (e^-) at 30 keV, protons (p^+) at 5 keV, and solar spectrum quanta (SSQ). The additivity coefficient (K_{add}) was calculated to quantitatively assess the mutual influence of these types of radiation on the formation and accumulation of defects and absorption centers.

It was established that for the sequential and separate irradiation of the mZnO micropowder modified with nSiO₂ nanoparticles, the additivity coefficient in two of the four sequential irradiation modes ($p^+ \rightarrow$ SSQ and $e^- \rightarrow$

SSQ) is close to unity; in the $\text{SSQ} \rightarrow \text{p}^+$ mode it equals 0.8, whereas in the $\text{SSQ} \rightarrow \text{e}^-$ mode it reaches 1.42–1.43.

To clarify the mechanisms of the processes occurring in the powders during irradiation, further studies are required, which are planned to be carried out by decomposing the induced absorption spectra under these types and modes of irradiation and analyzing the parameters of the elementary absorption bands. Such studies can be performed using the “Spectrum” setup, a unique apparatus in the Russian Federation, that allows for the in situ registration of diffuse reflectance spectra and induced absorption

spectra after each irradiation period directly within the irradiation chamber.

The practical significance of this study lies in the fact that the further advancement of space exploration relies on the development of technologies for producing spacecraft materials that ensure high reliability over long-term orbital missions (15–20 years).

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