

## ORIGINAL CONTRIBUTION

# Beyond sun protection factor: An approach to environmental protection with novel mineral coatings in a vehicle containing a blend of skincare ingredients

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## Abstract

**Background:** Premature skin aging results from exposure to a range of environmental factors, primarily ultraviolet radiation, but also high-energy visible light in the blue spectrum, infrared radiation, and environmental pollution. These extrinsic factors result in the generation of reactive oxygen species which promote photoaging and DNA damage resulting in skin cancers.

**Aims:** To formulate skincare products utilizing a new coating applied to zinc oxide and titanium dioxide particles and complimentary skincare ingredients to provide broad protection against a range of environmental insults.

**Methods:** A cross-polymer, multifunctional coating of silicate, polyalkylsilsesquioxane, and polydimethylsiloxane moieties increases the photostability and decreases the reactivity of mineral sunscreen agents when interacting with energy sources. These products are also formulated with antioxidants to minimize free radical propagation. Additionally, this coating improves the esthetic feel of mineral sunscreens, while the appearance is enhanced by formulating products with a blend of iron oxides.

**Results:** A series of in vitro and ex vivo studies demonstrated the ability of mineral-based products formulated with the new multifunctional coating to provide protection against ultraviolet radiation, high-energy visible light, infrared radiation, and environmental pollution.

**Conclusion:** Newly formulated mineral-based skincare products provide environmental protection, are ecologically safe, and can replace chemical-based sunscreen ingredients.

## KEYWORDS

free radicals, mineral sunscreen, photoaging, physical sunscreen, zinc oxide

## 1 | INTRODUCTION

The function of our skin is to protect us from trauma and invading bacteria, prevent dehydration, regulate body temperature, and provide the ability to sense heat and cold and different touch sensations. The outer epidermis and associated stratum corneum forms a waterproof barrier and contains pigment-producing melanocytes. The dermis below contains fibrous and elastic tissue which gives skin its strength and flexibility, hair follicles, nerve fibers, and sweat glands. Below this is a layer of subcutaneous fat and connective tissue. Unless preventative steps are taken, environmental factors such as sunlight and air pollution can damage the skin, resulting in signs of premature skin aging.

A novel line of skincare products has been formulated utilizing a new coating on zinc oxide and titanium dioxide particles and complementary skincare ingredients. This formulation is intended to provide broad protection from a variety of environmental and energy aggressors, including ultraviolet (UV) radiation, high-energy visible (HEV) light in the blue spectrum, infrared radiation, and environmental pollution. Importantly, these products have been formulated to be reef-friendly and replace chemical-based active sunscreen ingredients with mineral-based protection.

## 2 | CAUSES OF SKIN DAMAGE

### 2.1 | Effects of sunlight on skin

The skin is subject to constant exposure to the damaging effects of our environment. The most prevalent among these is UV radiation. UV radiation creates free radicals, such as reactive oxygen species (ROS), which damage the skin's extracellular matrix, including collagen and elastic fibers, causing the skin to take on a sallow appearance, lose its elasticity and sag.<sup>1</sup> Long-term exposure to UV radiation can also cause the skin to bruise and tear more easily. This occurs because UV exposure destroys the normal dermal architecture, replacing the superficial collagen-rich dermis with nonfunctional elastic tissue, a process termed solar elastosis. Because UVB (290-320 nm) is of shorter wavelengths than UVA (320-400 nm) and absorbed more effectively, it does not penetrate as deeply as UVA. It primarily affects the epidermis where it is responsible for causing the erythema and inflammation associated with sunburns. UVB and UVA are primarily responsible for the photoaged appearance of skin including the five key signs of skin aging: fine lines and wrinkles, enlarged pores, redness, pigmentation, and sagging skin. Skin cancer is also the result of chronic UV exposure. These changes result from direct DNA damage, primarily from UVB, as well as from free radical damage caused by inflammation and UVA.<sup>2,3</sup>

### 2.2 | Effects of infrared light on skin

Sunlight is also a source of infrared radiation. Although infrared radiation in a therapeutic setting can have beneficial effects on the skin, such as enhanced wound healing,<sup>4</sup> prolonged exposure can generate

ROS which promote photoaging.<sup>5</sup> In addition, infrared light is absorbed mostly in both the epidermis and deeper dermis due to its relatively deep penetration into skin, where it is converted to heat.<sup>6</sup> The damaging effects of heat include degrading proteins, such as collagen and elastic fibers, which contributes to solar elastosis, premature skin aging and skin sagging.<sup>6</sup>

### 2.3 | Effects of high-energy visible light, including blue light

It has been demonstrated that up to one-half of the free radicals produced in the skin may be due to sunlight in the visible regions of the spectrum.<sup>7</sup> While low doses of blue light may be beneficial for treating proliferative skin diseases and acne,<sup>8</sup> high doses and long-term exposure can generate damaging free radicals, such as nitric oxide.<sup>9</sup> It has recently been suggested that HEV light from electrical devices, such as computer screens and smartphones, may also have damaging effects on the skin.<sup>10</sup> This damage has been proposed to occur from the generation of free radicals in the skin resulting in oxidative stress which contributes to photoaging.<sup>11,12</sup> This is significant because most sunscreens do not provide protection against visible and near-infrared radiation.

### 2.4 | Effects of pollution on skin

The harmful effects of sunlight may be made worse by the damaging effects of air pollution.<sup>13</sup> Exposure to airborne particulate matter increases ROS and inflammation in skin,<sup>14,15</sup> exacerbating the damaging effects of sunlight<sup>16</sup> and producing the characteristic appearance of photoaged skin.<sup>17</sup>

## 3 | EXISTING SKIN PROTECTION

### 3.1 | Broad-spectrum ultraviolet protection

The extent of UVB protection provided by sunscreens is indicated by their sun protection factor (SPF). Products with a high SPF provide more protection against the injurious effects of UVB than those with a low SPF. The SPF is calculated by measuring the ratio of the minimal erythema dose (MED) of UV radiation on skin with sunscreen applied to it, compared to the MED on unprotected skin. In other words, a product with an SPF 50 will protect the skin until it is exposed to 50 times more UVB radiation than what is required to burn unprotected skin.<sup>18</sup> In the United States, UVA protection is measured by a variety of methods, including a critical wavelength assessment. Other sun protection rating systems have also been developed. The Japanese Cosmetic Industry Association Guidelines use a UVA protection scale from PA+ (low) to PA++++ (high), which has recently been adopted in the United States.

### 3.2 | Chemical sunscreens

Strategies for avoiding the harmful effects of the sun include sun avoidance, protective clothing, such as long-sleeves and broad-brimmed

hats, a UV protective window film on vehicles, and the use of sunscreens. Chemical sunscreens are the most common sunscreens on the market and protect the skin by absorbing UV light which is released as heat. These chemicals include oxybenzone, avobenzone, octisalate, octocrylene, homosalate, and octinoxate, usually formulated in various combinations. Unfortunately, growing evidence suggests there are numerous negative effects associated with the use of chemical sunscreens, including allergic reactions,<sup>19</sup> neurotoxicity,<sup>20</sup> and detrimental hormonal effects.<sup>21-23</sup> In some instances, chemical sunscreens have also been shown to enter the environment<sup>21,24</sup> where they pose a hazard to small marine organisms<sup>21,25</sup> and can even enter the food chain.<sup>21</sup> In 2018, the state of Hawaii signed a bill banning two of the more popular chemical sunscreen agents (oxybenzone and octinoxate) due to their detrimental impact on coral reefs.<sup>26</sup> Since that time, cities such as Miami Beach and the Florida Keys have proposed similar legislation.<sup>27</sup>

### 3.3 | Mineral sunscreens

In contrast to chemical sunscreens, mineral or physical sunscreens are designed to reflect, scatter, and also absorb solar radiation. These products are often formulated with titanium dioxide and zinc oxide.<sup>18</sup> Zinc oxide-containing products attenuate both UVA and UVB light, while titanium dioxide primarily attenuates UVB radiation. Historically, it has been difficult to formulate esthetically elegant formulations of sunscreens using only mineral sunscreen agents due to their opacity; however, with new innovative coating materials and skillful formulation involving the use of iron oxides, it is now possible to create products that have an appealing esthetic with the benefits of mineral protection. Mineral sunscreens offer an additional benefit of being less likely to cause irritation,<sup>28</sup> making them more suitable for sensitive skin and more environmentally friendly. In fact, the FDA has recently stated in the 2019 final monograph for nonprescription, over-the-counter sunscreen products that the mineral sunscreen actives zinc oxide and titanium dioxide are the only sun-blocking ingredients generally recognized as safe and effective (GRASE).<sup>29</sup>

### 3.4 | Other skin protectors

As the appearance of photoaged skin is largely due to oxidative damage caused by ROS, the topical application of antioxidants has been proposed as a means for preventing this skin damage.<sup>30,31</sup> Topically applied antioxidants can also protect against environmental pollution.<sup>32</sup> Among topical agents with proven skin-protecting properties are vitamin C, quercetin, *Aloe vera*, silymarin, chromane (benzodihydropyran), green tea extract, ginseng, and *Polypodium leucotomos* extract, among others.<sup>33-40</sup>

## 4 | DEVELOPMENT OF A NOVEL SKIN PROTECTOR

A line of products (Sunforgettable<sup>®</sup> Total Protection; Colorescience<sup>®</sup>, Inc) has been developed using novel, patented, sun-protecting agents

(EnviroScreen<sup>™</sup> Technology) which provides chemical-free, mineral-based protection from the damaging effects of UVA/UVB, environmental pollution, HEV/blue light, and infrared radiation in an esthetically elegant format. These sunscreen formulations contain zinc oxide and titanium dioxide actives encapsulated by a novel coating technology,<sup>41-43</sup> and iron oxides which are blended with a proprietary combination of ingredients including vitamins, antioxidants, and hydrating molecules. Together with the novel coated actives, these formulations are designed to nourish skin, protect against free radicals, and protect against UVA/UVB-induced skin damage, air pollution, blue light, and infrared radiation that contribute to skin damage. This proprietary technology provides broad-spectrum SPF50/PA+++ and PA++++ protection, thereby helping to prevent photoaging by protecting against UV damage and allows damaged skin to repair itself.

### 4.1 | Novel coating background

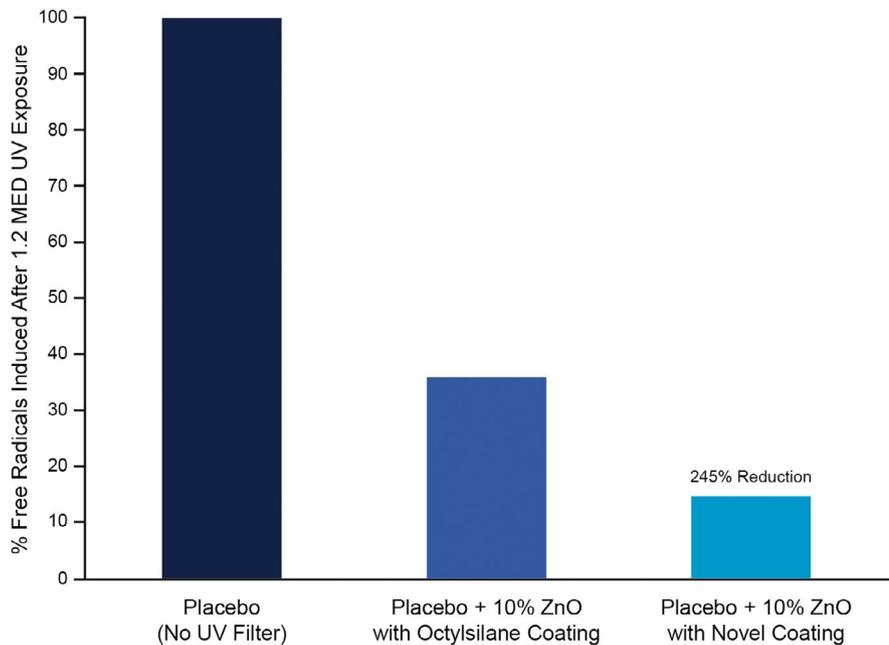
A novel cross-polymer, multifunctional coating consisting of silicate, polyalkylsilsesquioxane, and polydimethylsiloxane moieties has been developed in response to the need to increase the photostability and decrease the reactivity of mineral sunscreen agents when exposed to various energy sources (Solésence LLC). The ability of this multifunctional coating to quench free radicals within the epidermis and dermis following exposure to UVA and UVB has been demonstrated using various techniques, including electron spin resonance spectroscopy (ESR). ESR, which detects unpaired electrons, and other ex vivo methods have been developed to quantify free radicals in the skin following UV exposure.<sup>44</sup> When applied as a topical formulation, this multifunctional coating on zinc oxide particles resulted in a 245% reduction in the number of free radicals generated in the skin following UV exposure compared to the same formula using zinc oxide with a standard coating of octyltriethoxysilane (Figure 1).<sup>45</sup>

The addition of this multifunctional coating allows the esthetic properties of mineral actives to be adjusted to achieve a desired characteristic, such as improved skin feel. In addition, this coating can allow for improved dispersion characteristics and enable greater product hydrophobicity, resulting in better water-resistance. The coating can be further used to adjust the bulk characteristics of sunscreen powders to deliver safe and functionally superior dispensing characteristics.

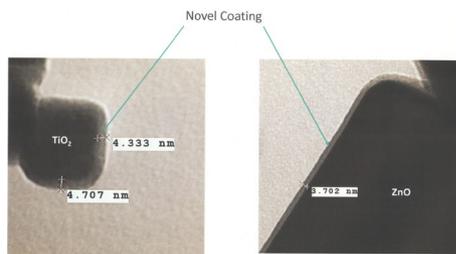
This coating has distinct advantages over more traditional technologies, such as those using octyltriethoxysilane. The bonding of traditional coating chemicals to mineral actives is limited by available surface reactive sites, leading to incomplete particle surface coverage. In contrast, the multifunctional coating creates a complete, dense, and homogeneous encapsulation. This can be verified using transmission electron microscopy (Figure 2). These coating characteristics are critical for enabling the key product attributes, discussed above.

### 4.2 | Final product formulation

A series of carefully selected ingredients were chosen to complement the patented triple-moiety coating into a proprietary blend.



**FIGURE 1** When applied as a topical formulation, multifunctional coating consisting of silicate, polyalkylsilsesquioxane, and polydimethylsiloxane moieties on zinc oxide particles resulted in a 245% reduction in the number of free radicals generated in the skin following UV exposure



**FIGURE 2** Electron microscopy shows the multifunctional coating creates a complete, dense, and homogeneous encapsulation of titanium dioxide (left) and zinc oxide (right particles)

These final formulations were constructed to protect against broad-spectrum light including high-energy visible light (HEV/blue light), infrared radiation, and UVA/UVB radiation.

The formula was also constructed with the intent to provide protection from environmental pollution and other forms of free radical damage. This was achieved through the multifunctional coating technology and the addition of a select blend of antioxidants, including the innovative new ingredient dimethylmethoxy chromanol, which captures both nitrogen and oxygen radicals to limit oxidative stress.<sup>37</sup> In some formulations, the proprietary antioxidant combination also includes natural extracts such as green tea, grapeseed, and oakwood extracts, that provide further skin protection from free radicals produced by pollution and other sources such as UVA.

Defense against HEV light is provided by the multifunctional-coated UV mineral actives and iron oxides. This combination has been demonstrated to significantly attenuate HEV light associated with radical formation in the skin,<sup>46</sup> thereby protecting skin from photoaging associated with blue light and HEV light.<sup>47</sup> To further

protect against infrared radiation, antioxidants including tara tannins and sunflower extract are added.

The formulations also include the key hydrating ingredients hyaluronic acid powder and a combination of *Tremella fuciformis* sporocarp extract, betaine, and glycerin which has been shown to be over four times more hydrating than hyaluronic acid itself.<sup>48</sup> The formulation also contains the skin-calming ingredients niacinamide, bio-available vitamin E, and soothing agents derived from chamomile oil. Ingredients largely comprised of natural extracts were chosen for each specific formulation (powder, face and body) to help enhance the protection against each of the intended elements. Importantly, all complimentary skincare ingredients were added at manufacturers' suggested levels based on efficacy data demonstrated by each manufacturer.

### 4.3 | Final product testing

#### 4.3.1 | UVA/UVB testing

All of the tested formulations achieved maximal UVB protection based on FDA standards set forth in the current draft sunscreen monograph<sup>49</sup> as measured by SPF rating (SPF 50) by a validated third party laboratory.<sup>46</sup> In addition, due to properties associated with the hydrophobic nature of the patented coating, each formulation achieved 40- to 80-minute water-resistance, based on 20-minute increment exposures in a validated water tank with standard 20-minute rest intervals (Table 1).

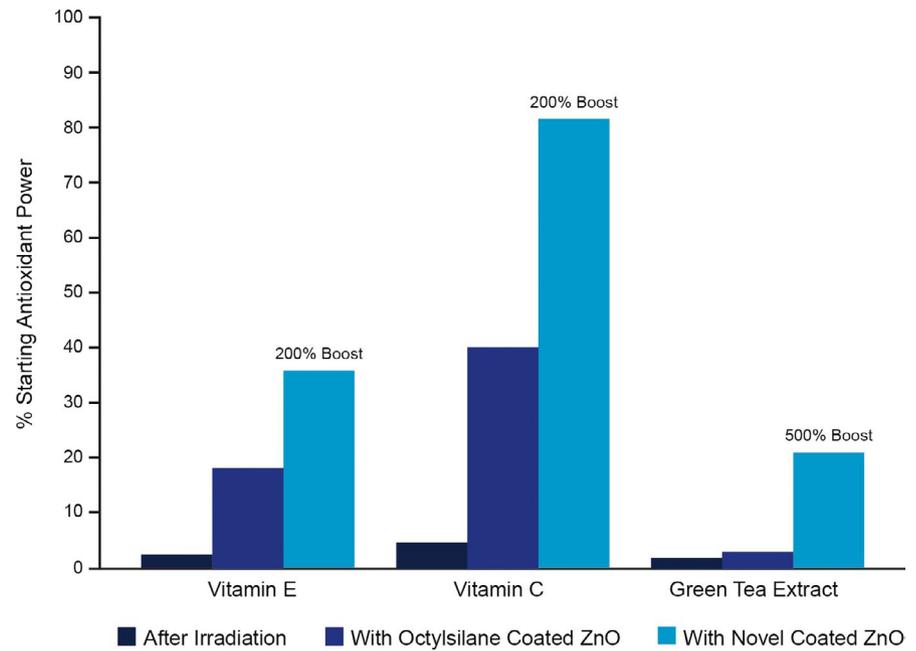
Further, each formula underwent UVA testing using the FDA-approved critical wavelength test procedures and the newer Japanese PA test with each product achieving ratings between PA+++ and PA++++, the highest rating of UVA protection.

**TABLE 1** Characteristics of novel coating formulation powder, face, and body products

	SPF	Water-resistance	PA rating	Ingredients
Powder product	50	80 minutes	PA++++	TiO <sub>2</sub> 22.5%, ZnO 22.5%
Face product	50	40 minutes	PA+++	ZnO 12%
Body product	50	80 minutes	PA+++	ZnO 12%

Abbreviations: SPF, sun protection factor; TiO<sub>2</sub>, titanium dioxide; ZnO, zinc oxide.

**FIGURE 3** The antioxidant boosting effect of multifunctional-coated zinc oxide is substantially greater than octylsilane-coated zinc oxide

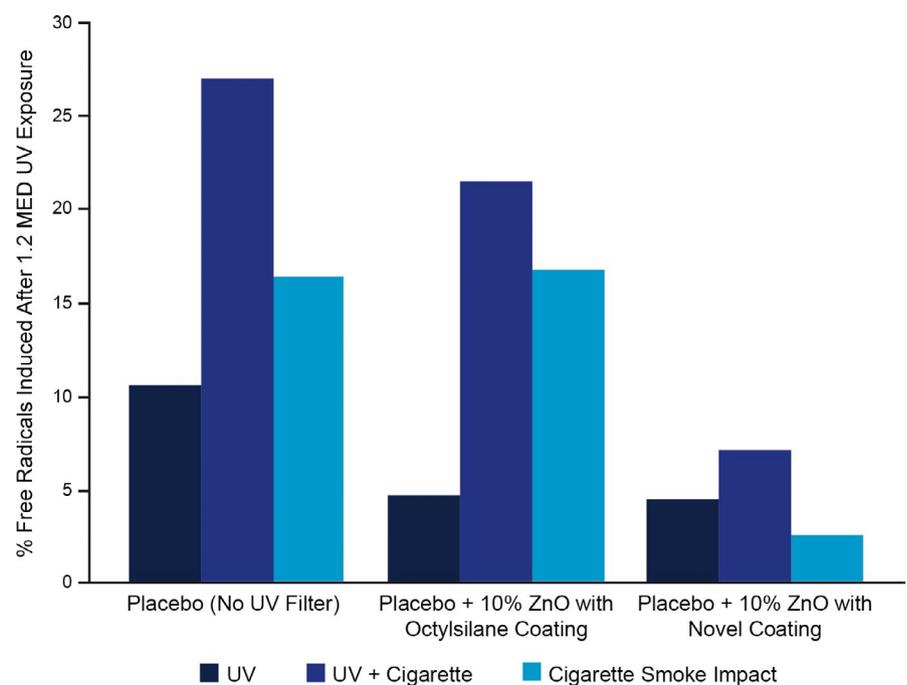


### 4.3.2 | Protection from free radicals and support for antioxidants

Antioxidants have been a staple of skincare formulations. They are intended to reduce the impact of free radicals which are formed as

a result of photoreactivity and energy exchange. A historical challenge in formulas containing antioxidants has been maintaining their stability when exposed to environmental aggressors, particularly UV radiation. Traditional antioxidants frequently break down in less than 90 minutes when exposed to UV radiation. This was demonstrated

**FIGURE 4** Free radical formation caused by UV radiation exposure and air pollution (cigarette smoke) is greatly attenuated by multifunctional-coated zinc oxide relative to octylsilane-coated zinc oxide



**TABLE 2** Characteristics novel coating formulation powder product vs three competitive sunscreens

Product	SPF	Water-resistance	Ingredients
Powder Product	50	80 Minutes	TiO <sub>2</sub> 22.5%, ZnO 22.5%
Competitive Product A	45	None	TiO <sub>2</sub> 15.7%, ZnO 20.0%
Competitive Product B	30	None	TiO <sub>2</sub> 15.0%, ZnO 12.0%
Competitive Product C	45	None	TiO <sub>2</sub> 20.75%, ZnO 19.6%

Abbreviations: SPF, sun protection factor; TiO<sub>2</sub>, titanium dioxide; ZnO, zinc oxide.

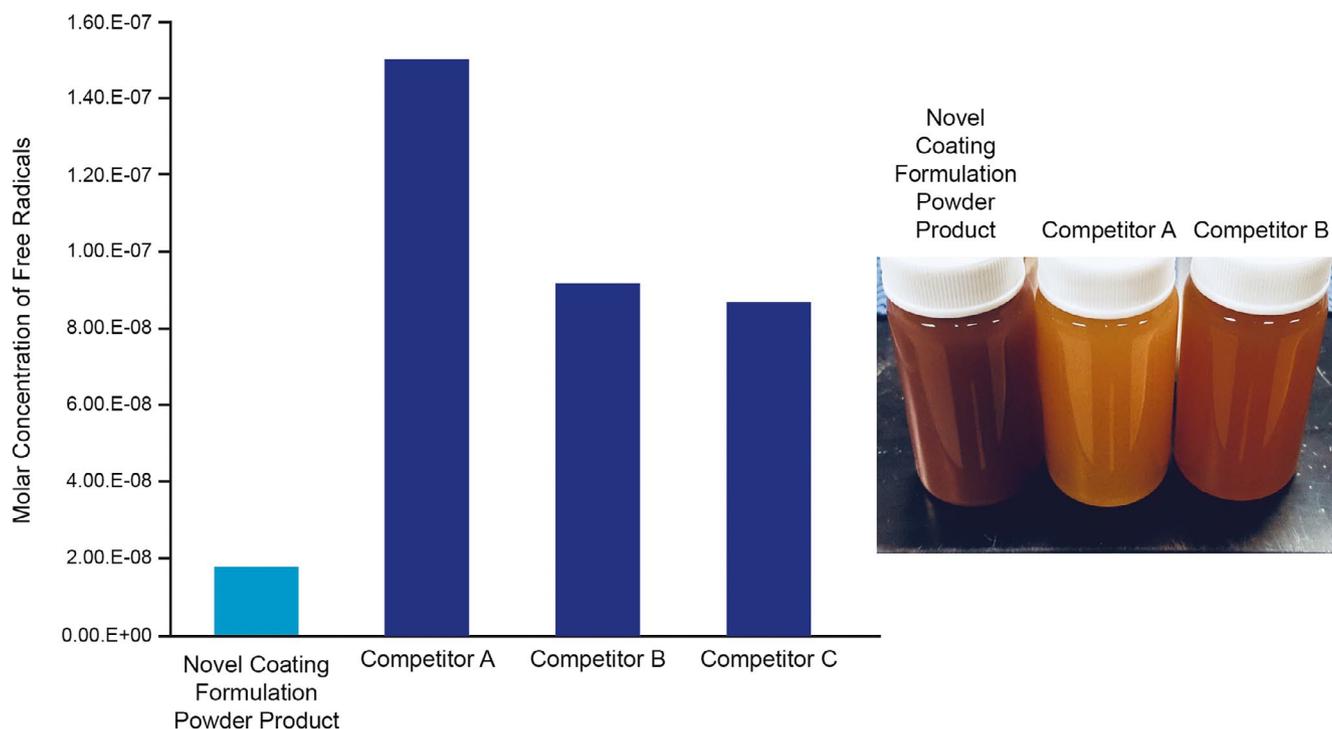
by adding individual antioxidants to a prototype formula containing no UV filter and initially measuring their antioxidant power (AP) using ESR. The formulas were then exposed to UV radiation and the AP values were again determined using ESR and expressed as a percentage of their original values. Moderate UV exposure resulted in the near complete depletion of AP. This experiment was then repeated by adding standard zinc oxide and the multifunctional zinc oxide UV filters to the antioxidant containing preparations (Figure 3). Both standard zinc oxide and the multifunctional zinc oxide improve the retained AP; however, due to the ability of the multifunctional coating to quench free radicals, significant 200%-500% boosts in AP occurred in the formula containing multifunctional zinc oxide, demonstrating the ability to substantially improve the efficacy of

frequently used topical antioxidants, which are known to provide skin health benefits.

### 4.3.3 | Protection from environmental pollution

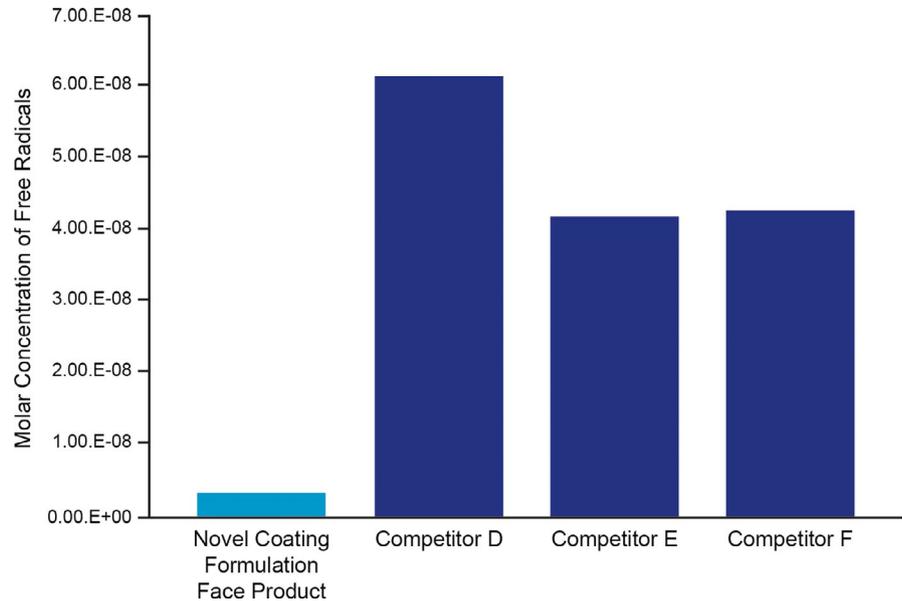
Environmental pollution is increasing dramatically around the world and poses a major health challenge, affecting the ability to maintain healthy skin by causing irritation, blocked pores, and accelerating photoaging.<sup>13,50</sup> As discussed previously, prior work has examined the performance of zinc oxide mineral actives coated with the multifunctional coating compared to that of zinc oxide with a standard octyltriethoxysilane coating in protecting against the combined effects of pollution and UV exposure (Solésence LLC).<sup>45</sup> In that work, electron paramagnetic resonance spectroscopy was used to detect free radicals at the surface of the skin using an ex vivo porcine skin model, and cigarette smoke was used as a proxy for environmental pollution as the particle profile (PM 2.5) is similar to that observed for diesel soot.<sup>51</sup> Formulas containing the multifunctional-coated zinc oxide and standard zinc oxide, and a placebo formula containing no UV filter were applied to the surface of porcine skin and then exposed to 1.2 MED of UV light. For the case of pollution exposure, samples were exposed to the smoke of one cigarette prior to UV exposure. Free radicals were then detected using ESR following UV exposure.

Using the placebo formula as a reference, the added pollution had a substantial impact, more than doubling the concentration of free radicals generated at the skin surface when the environmental



**FIGURE 5** DPPH free radical assays of commercial powder sunscreen formulas following UV radiation exposure. The formula described in this work, incorporating multifunctional-coated mineral UV filters and selected antioxidants, greatly outperforms the competitor formulas as further illustrated by the greater persistence of the purple color of the DPPH dye

**FIGURE 6** DPPH free radical assays of commercial face lotion emulsion sunscreen formulas following UV radiation exposure. The formula described in this work, incorporating multifunctional-coated mineral UV filters and selected antioxidants, greatly outperforms the competitor formulas



aggressors are combined. When the samples were not exposed to pollution, the UV protective effect of zinc oxide was evidenced by a reduction of free radicals. When a pollutant was added prior to UV exposure, the placebo and standard zinc oxide formulas showed large increases in free radical generation, while the multifunctional-coated zinc oxide formula showed only a slight increase.

The UV-only and pollution plus UV signals can be subtracted to isolate the effect of the UV-activated pollution. While the standard zinc oxide formula provided UV protection, it provided no benefit in stopping free radical propagation compared to a placebo formula containing no UV filter (Figure 4). In contrast, the multifunctional zinc oxide formula provided a 7-fold reduction in the effect of the pollutant. This study demonstrated that products developed with the multifunctional coating technology represent the only UV protective actives with demonstrated anti-pollution properties.

It has also been demonstrated that quenching free radicals by UV blockers with multifunctional coatings leads to significant boosting of antioxidant activity and substantial protection from environmental pollution. As discussed above, these actives are combined in a proprietary blend of supportive ingredients. These final formulations have been tested for their ability to suppress free radical formation and show superior performance when tested against comparable commercial products.

**TABLE 3** High-energy visible light shielding novel coating formulation face product vs four competitive sunscreens

Product	Percent block		
	415 nm	440 nm	465 nm
Face product	71	64	60
Competitive product D	65	58	52
Competitive product E	30	26	24
Competitive product F	60	52	47
Competitive product G	33	30	27

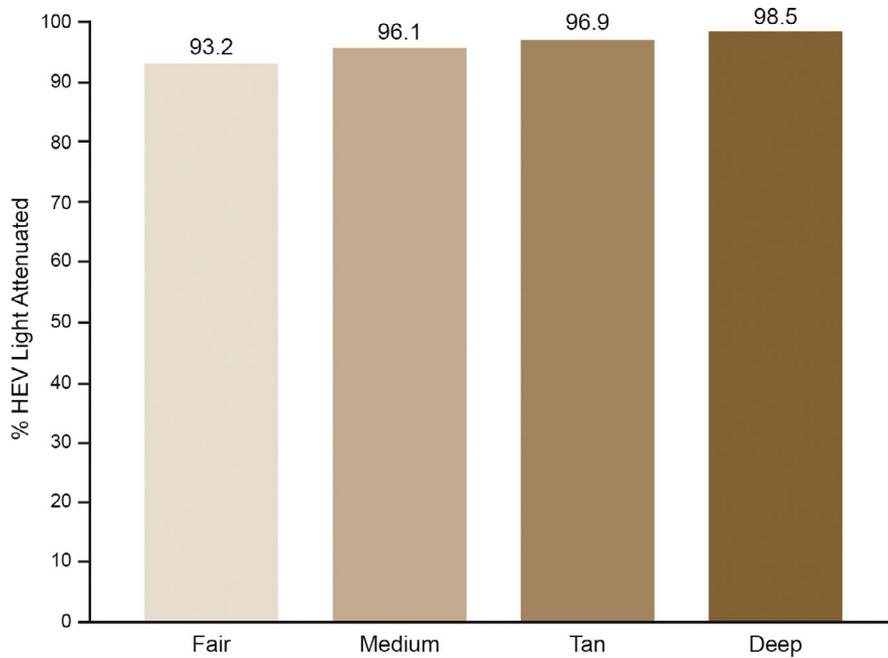
In these experiments, the formulas themselves were exposed to UV radiation and assayed for free radicals using a modification of the well-established 2,2-diphenyl-1-picrylhydrazyl (DPPH) antioxidant assay.<sup>52</sup> DPPH is a purple dye composed of stable free radical molecules which react and combine with other free radical species, thereby acting as scavengers. This reaction is accompanied by a color change from purple to yellow and may be quantitatively monitored spectrophotometrically through the decrease of the DPPH absorbance signal at 517 nm.

In one experiment, one powder formula was tested against three commercial powder sunscreens (Table 2). All four products were combined with a DPPH solution and irradiated with 0.44 MED of UV radiation. The results are shown in Figure 5 together with an image illustrating color changes in the DPPH following UV exposure. The new formulation showed a small free radical signal near the limit of detection as indicated by maintaining the dark color of the assay solution. In contrast, the other powder sunscreens showed substantial free radical generation with corresponding change in solution color. This notable difference in their performance demonstrates the superiority of this formulation in quenching the free radicals that can lead to oxidative stress in the skin.

This assay has also been performed on other dosage forms, such as a facial emulsion, that utilizes this same proprietary technology. Similar results have been verified independently using the ESR method. As shown in Figure 6, the new emulsion formulation showed little detectable free radical signal, whereas the tested commercial comparator products in the same dosage form show substantial generation of free radicals following UV exposure.

#### 4.3.4 | High-energy visible light

Environmental exposure to HEV light is primarily from the sun, but also from smartphones, tablets, and computer screens, and recently reports have demonstrated HEV light can be destructive to the skin.<sup>53</sup> The ability of products containing the multifunctional



**FIGURE 7** High-energy visible light blocking performance of tinted powder sunscreen formulas of differing shades described in this work. The addition of iron oxides increases the effectiveness of multifunctional-coated UV mineral actives, blocking 93%-98% of these wavelengths

technology to block HEV light was measured directly using the diffuse transmittance method. The emulsion formulation intended for the face blocked more HEV light at all measured wavelengths compared to an array of other commercial products (Table 3).

It is well known that iron oxides play a critical role in the protection against HEV exposure and enhance the already substantial HEV protection provided by the UV actives with the multifunctional coating. The powder formulations have been developed in four different shades, with the darker shades containing a higher percentage of iron oxides. All shades of powder demonstrated the ability to shield at least 93% of HEV, with the darker shades increasing the shield potential against HEV to over 98% (Figure 7).

## 5 | CONCLUSION

With a greater need to protect human skin from ever-increasing environmental insults including the damaging effects of solar radiation, pollution, and other sources of free radical damage that lead to photoaging and skin cancer, it is increasingly more important to better protect people with improved all-mineral sunscreen formulations. We have presented data supporting a series novel formulation of chemical-free, all-mineral sun protection incorporating novel coatings combined with carefully chosen skincare ingredients to address the key environmental and climate insults that can damage skin. These formulations provide the maximum SPF/PA scale protection and 40- to 80-minute protection with water exposure. Further, such products have been demonstrated in vitro and ex vivo to show a significant reduction in free radical formation in models simulating the impact of environmental pollution and HEV exposure as compared to standard mineral coated sunscreen formulations.

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**How to cite this article:** Bernstein EF, Sarkas HW, Boland P, Bouche D. Beyond sun protection factor: An approach to environmental protection with novel mineral coatings in a vehicle containing a blend of skincare ingredients. *J Cosmet Dermatol.* 2020;19:407–415. <https://doi.org/10.1111/jocd.13007>