

# DESIGN AND DEMONSTRATION OF AN AUTOMATED BIDIRECTIONAL REFLECTOMETER FOR LOW-REFLECTIVITY OPTICAL COATINGS

Deepali Shirsekar<sup>1</sup>, N. Q. Vinh<sup>1</sup>, J. R. Mahan<sup>1\*</sup>, Kory J. Priestley<sup>2</sup>

<sup>1</sup>Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061, USA

<sup>2</sup>NASA Langley Research Center, Hampton, Virginia 23682, USA

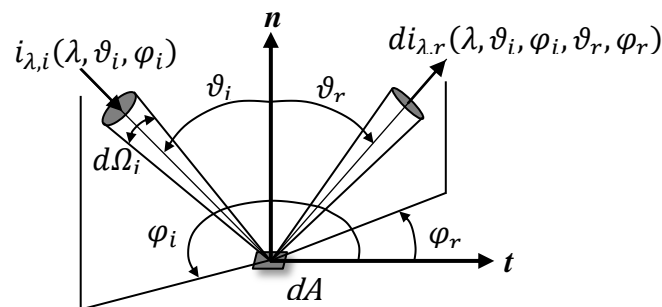
## ABSTRACT

While diffuse emission and diffuse-specular reflection are often adequate models in the conceptual design stage of radiation-dominated systems, more accurate bidirectional spectral models may be required for performance evaluation. Described is an effort leading to empirical models for the bidirectional reflectance distribution function (BRDF) of highly specular, low-reflectivity black coatings typical of those frequently used in aerospace and remote sensing applications. An automated goniometer-based reflectometer has been designed, assembled, and used to obtain the needed measurements at both polarizations. The measurement problem is exacerbated by the fact that the BRDF of such materials varies several orders of magnitude over the range of incidence angles of interest. The BRDF is also influenced by coating thickness and wavelength as well as by the method used to apply the coating. The current contribution describes preparation of the samples and the design and use of the reflectometer. New data are presented for the Z302 BRDF for both the p- and s-polarizations at 305 nm.

**KEY WORDS:** Radiation heat transfer, Bidirectional reflectivity, BRDF, Measurement technology, Z302

## 1. INTRODUCTION

Black surfaces are used in optical and radiation heat transfer applications both to control stray light and to enhance absorption. For effective stray light control, the small amount of reflection that does occur should be specular. The bidirectional reflectance distribution function (BRDF), or bidirectional reflectivity, is a measure of the efficiency and directionality with which a surface reflects incident radiation. Surface reflectance is generally a function of wavelength as well as the incident zenith angle  $\vartheta_i$ , the reflected zenith angle  $\vartheta_r$ , and the reflected azimuth angle  $\varphi_r$ . Most surfaces of practical engineering interest may be considered isotropic; therefore, their BRDF is invariant with the incident azimuth angle  $\varphi_i$ .



**Fig. 1** Nomenclature for bidirectional reflectivity

The BRDF is defined [1, p. 84] as

\*Corresponding Author: jrmahan@vt.edu

$$BRDF(\lambda, \vartheta_i, \varphi_i, \vartheta_r, \varphi_r) = \rho(\lambda, \vartheta_i, \varphi_i, \vartheta_r, \varphi_r) \equiv \frac{di_{\lambda,r}(\lambda, \vartheta_i, \varphi_i, \vartheta_r, \varphi_r)}{i_{\lambda,i}(\lambda, \vartheta_i, \varphi_i) \cos \vartheta_i d\Omega_i}. \quad (1)$$

In Eq. (1),  $i_\lambda$  is the spectral intensity, or radiance ( $\text{W}/\text{m}^2 \cdot \text{sr} \cdot \mu\text{m}$ ), the subscripts  $i$  and  $r$  denote the incident and reflected angles,  $\lambda$  is wavelength, and  $d\Omega_i$  is the incident differential solid angle. The relationships among these quantities are illustrated in Fig. 1. Once known for a given surface, the BRDF can be used to compute the other surface properties such as emissivity, absorptivity, and the remaining reflectivities [1, p. 85].

In the current contribution we seek to measure and model the BRDF of Aeroglaze® Z302 Absorptive Polyurethane Black [2], a coating used widely in optical applications because of its high absorptivity and specularity. While several investigations [3-7] report BRDF measurements for Z302 over a range of wavelengths, published datasets often omit crucial information such as layer thickness. Also, results differ somewhat due to a combination of measurement error and sample-to-sample variations. It is therefore important that information such as high-resolution photographs, AFM surface profiles, and layer thicknesses be included in BRDF data files. Also, when high accuracy is required for subsequent performance modeling of Z302-coated surfaces, it is desirable that the BRDF model used be based on data from samples prepared in a similar manner.

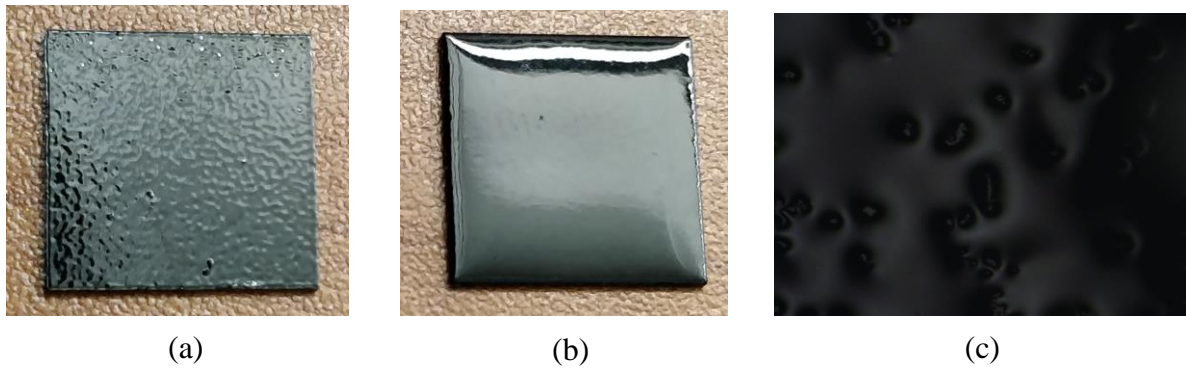
## 2. CREATION OF AEROGLAZE® Z302 SAMPLES

Z302 is typically delivered in a well-sealed one-gallon can. It is toxic in its liquid form and must be handled with care; the instructions in the Material Safety Data Sheet shipped with the product should be scrupulously adhered to when preparing Z302 samples. Specifically, application of the coating should be done in a ventilated hood while wearing chemical-resistant gloves, eye protection, and a mask. It is recommended that coatings be applied on clean, dry surfaces at a temperature of between 13°C (55°F) and 35°C (95°F). When still in liquid form Z302 degrades with long-term exposure to humid atmospheric air. Therefore, once opened, the unfilled volume of the can should be purged with an inert gas such as dry nitrogen before resealing. The cure time depends on the conditions under which the coating is stored. The samples prepared for the effort reported here were prepared and stored in a chemical hood with good air circulation. The cure time for handling is around 12 hours at room temperature. The recommended time between applying a recoat is 3 to 8 hours.

We applied our initial coatings to polished aluminum substrates representative of typical applications. However, we soon realized that the substrate surface topography influences the coating topography unless the layer is quite thick. To assure that the measured BRDF is free of such artifacts, the substrate must be as smooth and uniform as possible. By a process of trial-and-error, we settled on using glass microscope slides and vapor-deposited silicon wafers as substrates. Both are sufficiently flat that they do not impose surface features on the Z302 layer regardless of coating thickness, thereby promoting repeatability in BRDF measurements.

The method of application of the coating also plays a vital role. Initially we used an air brush with an adjustable spray-head to apply the liquid Z302 by hand. We found that the combination of spray-head adjustment and spray-head-to-sample distance was critical. In-flight curing of the individual droplets can produce a grainy surface if the spray is too fine and/or the distance too great. It is important that the droplets arrive at the sample surface in liquid form to allow coalescence and surface tension to spread the paint uniformly. After sufficient practice using multiple recoats—usually three—we were able to produce extremely smooth, uniform coats on glass slides and silicon wafers.

Figures 2(a) and (b) are photographs of our early efforts to create Z302 samples [6]. The sample in Fig. 2(a) has a single 30- $\mu\text{m}$ -thick layer, and the sample in Fig. 2(b) has a 95- $\mu\text{m}$ -thick layer applied in three recoats. Both samples were created by hand spraying on a silicon wafer chip. Figure 2(c) is an atomic force measurement (AFM) image of the 30- $\mu\text{m}$ -thick single-layer sample. This latter layer clearly exhibits the graininess alluded to in the previous paragraph. The three-layer sample in Fig. 2(b) was created by holding the spray head nearer to the sample surface during recoating.



**Fig. 2** Two early samples of Z302 coatings, (a) one 30- $\mu\text{m}$  thick and (b) the other 95- $\mu\text{m}$  thick [6], and (c) an atomic force measurement (AFM) image of a portion of the 30- $\mu\text{m}$ -thick sample.

One of the chief advantages of Z302 in aerospace applications is its post-cure hardness and durability. Within 24 hours of application, the coating can be handled and manipulated without fear of marking or otherwise damaging its surface. This admirable property can be a disadvantage in creating multiple samples over a period of several weeks. Our original procedure was to use a bulb pipette to suck the liquid product from the open can and to transfer it to the small reservoir in the air brush. Upon completion of the spraying process, the bulb pipette and air brush must be thoroughly cleaned, a procedure that creates a significant amount of presumably toxic waste. If this is not promptly done, the paint hardens into a nearly indestructible residue that cannot be easily removed. Because creation and disposal of toxic waste in a university environment can be a bureaucratic nightmare, this essential step tends to be put off or even disregarded, resulting in a thoroughly clogged and inoperable air brush. Our solution to this dilemma was to build a rig for submerging the substrate in the can of liquid Z302 while maintaining it horizontal during dipping and subsequent curing. This procedure was found to create thick and extremely smooth samples with a single dipping while avoiding creation of toxic waste.

A photograph of two samples created using the dipping method appears in Fig. 3. Although these mirror-like samples appear to be wet, they are in fact completely cured. Apparently liquid Z302 is unable to “wet” either pure silicon or glass, as evidenced by the uncoated patches. Once cured the hardened Z302 forms a strong bond with both substrate materials. Even though the layer is globally non-uniform, large patches of locally uniform Z302 having a thickness of about 100  $\mu\text{m}$  are available for testing.

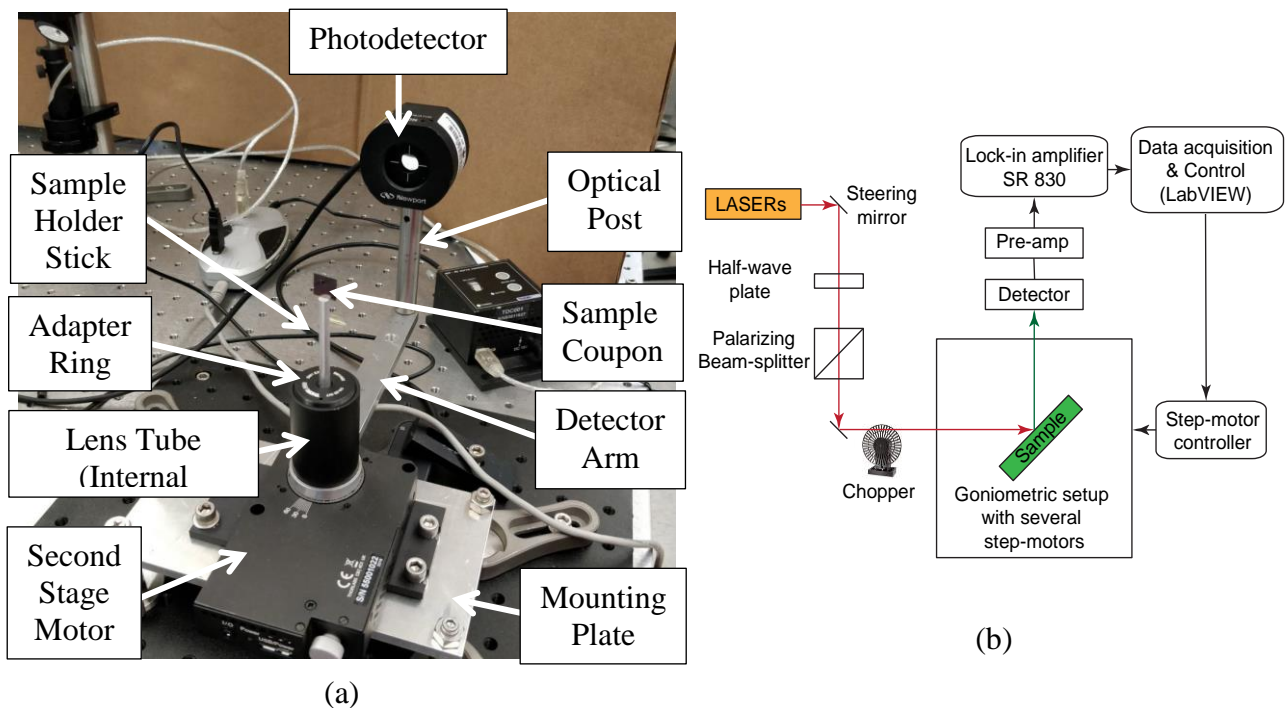


**Fig. 3** Samples created by dipping a silicon wafer and a glass microscope slide into the can of Z303.

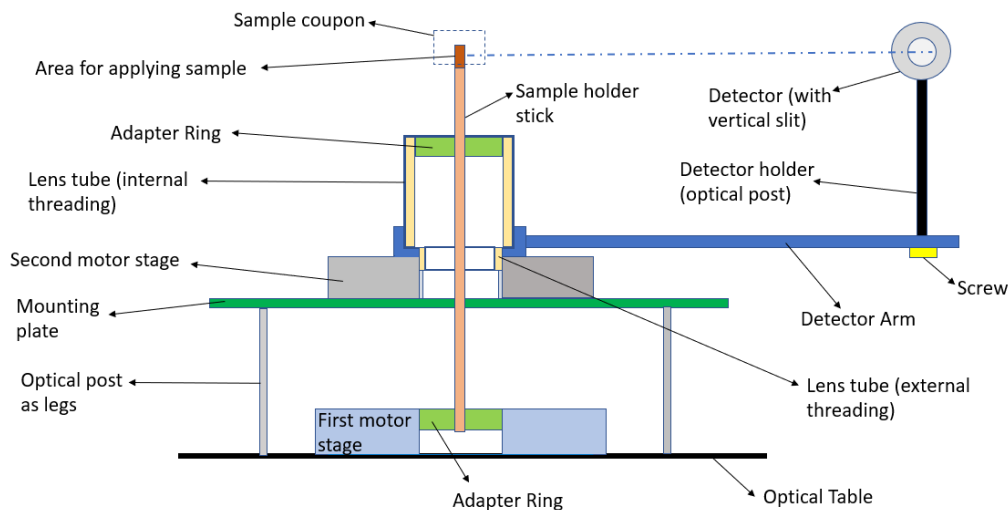
### 3. THE AUTOMATED GONIOMETER RELECTOMETER

We measured the BRDF of our Z302 samples using a two-stage automated goniometer reflectometer of our own conception and fabrication [6]. Figure 4(a) is a photograph of the apparatus, Fig. 4(b) is the optical and

control layout, and Fig. 5 displays the mechanical layout. The rotational angles of both precision stage motors are controlled by a LabVIEW program which also logs their angular position and the output of the Newport 918-UV photodetector. The first stage motor rotates the sample to a series of pre-programmed incidence angles, and the second stage motor rotates the photodetector to a set of pre-programmed viewing angles. A narrow beam from one of three lasers covering the UV, visible, and IR ranges is directed to the sample through two steering mirrors, a half-wave plate, a polarizing beam splitter, and a chopper. The half-wave plate and beam splitter together control both the intensity and polarization of the beam, while the chopper further attenuates the beam power and, along with the lock-in amplifier, removes the ambient light background from the photodetector signal. This latter feature allows measurements to be taken in ambient lighting conditions. A 200- $\mu\text{m}$  slit is positioned as a field stop in front of the photodetector. Calibration is assured by first observing the known-power laser source with a Coherent FieldMate laser power meter in combination with a Coherent PowerMax PM10 detector head. The beam is then observed in front of the sample. Details of the design, fabrication, testing, and use of the reflectometer are reported elsewhere [8].



**Fig. 4** (a) Photograph of the automated goniometer reflectometer, and (b) optical and control layout [6]



**Fig. 5** Schematic layout of the automated goniometer reflectometer [6]

#### 4. EXPERIMENTAL RESULTS AND DISCUSSION

To this point the instrument has been used to obtain BRDF measurements in the visible ( $\lambda = 532$  nm) [6] and in the ultraviolet ( $\lambda = 350$  nm). Measurements in the infrared ( $\lambda = 808$ ) are currently underway. Figure 6 is a plot of BRDF versus viewing angle at 350 nm for both p- and s-polarization at incidence angles of 10, 20, 30, 40, 40, 60, 70, and 80 deg, measured for the Z302-on-glass sample shown in Fig. 3. The observed dip in the BRDF for p-polarized light at the Brewster angle—about 50 deg in this case—is anticipated by classical optics [9, p. 365]. This same trend with approximately the same Brewster angle is reported in Ref. 6 for p-polarized 532-nm light reflected from the grainy sample in Fig. 2(a). However, in the cited reference the peaks are broadened and somewhat attenuated due to scattering by the grain structure. The general trend of increasing peak BRDF with incidence angle for s-polarized light, reported here for the UV, has also been reported in the visible for s-polarized light [6] as well as for naturally polarized light [3-5, 7].

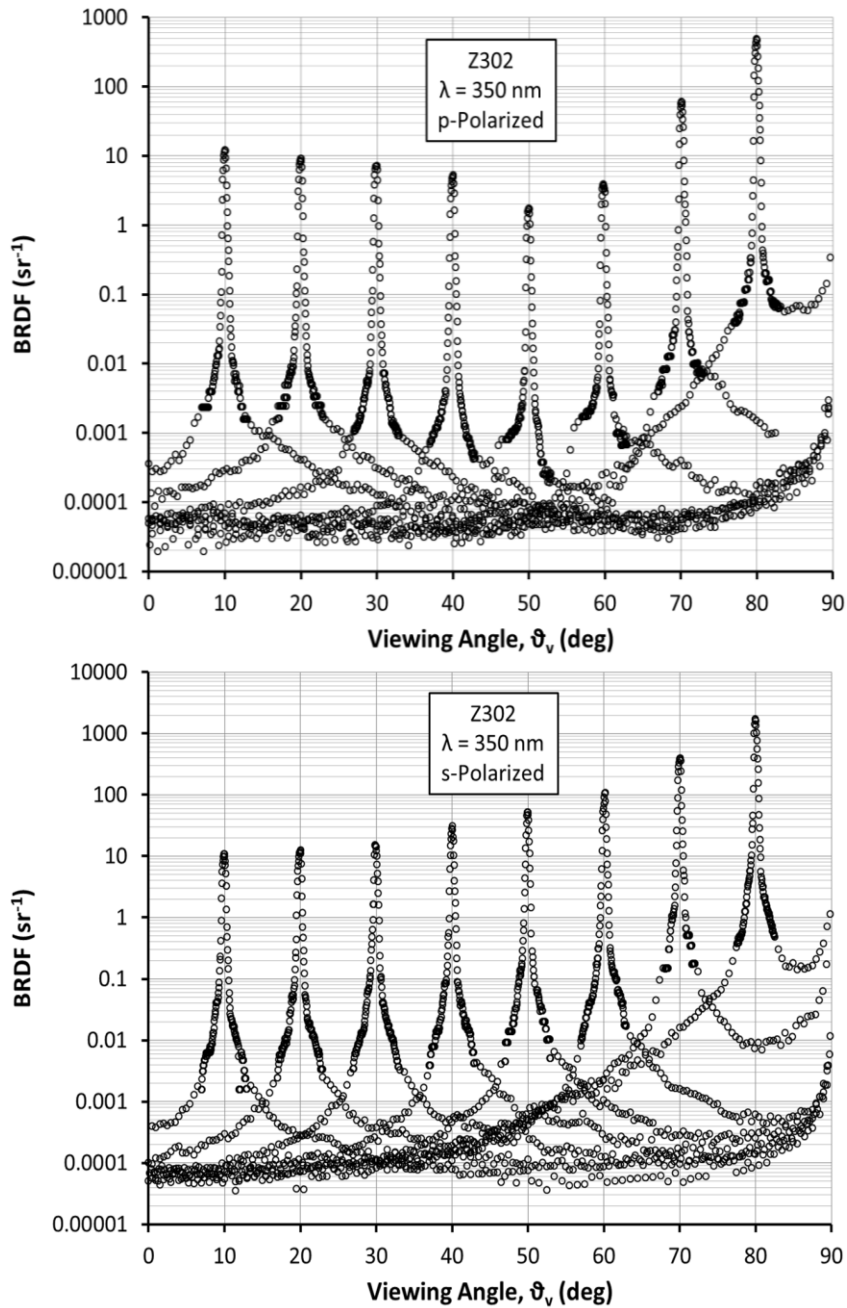
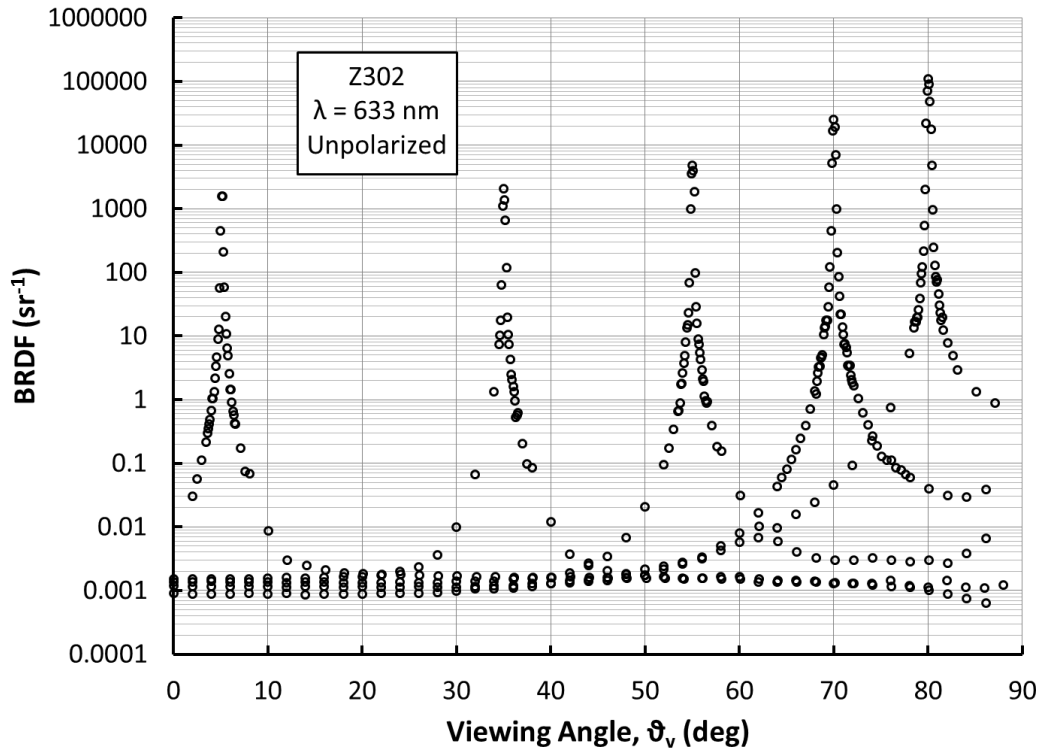


Fig. 6 BRDF for the glass substrate sample shown in Fig. 3.

We have been unable to find BRDF data, either polarized or unpolarized, for Z302 in the ultraviolet in the literature for comparison with our results. However, NASA has recently released previously unpublished unpolarized BRDF data at 633 nm. These data, corresponding to incidence angles of 5, 35, 55, 70, and 80 deg, are plotted in Fig. 7. The measurements were made by Photon Engineering for NASA under subcontract to Harris Space and Intelligence Systems and are used here with permission [5].



**Fig. 7** BRDF of Z302 at 633 nm (unpolarized) for incidence angles of 5, 35, 55, 70, and 80 deg [5]

The polarized BRDF data in Fig. 6 can be converted to equivalent unpolarized BRDF data by averaging the values at corresponding viewing angles. The curves appearing in Figs. 6 and 7 are very similar in shape and trend. For example, the peak values vary with incidence angle by about two orders of magnitude in both datasets. However, the peak and background values differ significantly between the two datasets, with the BRDF values in Fig. 6 being uniformly lower than the corresponding values in Fig. 7 by more than an order of magnitude. On the other hand, the results reported in Fig. 6 are in reasonable agreement with the visible (532 nm) results reported in Ref. 6, and with the infrared (10.6  $\mu\text{m}$ ) results reported in Ref. 3. The differences among results in Fig. 6 and those reported in Refs. 3 and 6 fall within a range attributable to wavelength dependence and/or experimental uncertainty.

If we reject the hypothesis of a fixed error in either our measurements in Fig. 6 or those of Photon Engineering in Fig. 7, with the latter eventuality deemed less likely than the former, there remain several possible explanations for the observed difference in level. First, a general decrease in reflectivity with wavelength is consistent with an increase in relative roughness,  $\sigma_0/\lambda$ , where  $\sigma_0$  is the optical roughness. Therefore, for a fixed optical roughness, the reflectivity can be expected to decrease with decreasing wavelength. However, extant theory [9] indicates that for a “smooth” surface ( $\sigma_0/\lambda \ll 1$ ) this effect could not by itself account for the observed difference. A second possibility is that ultraviolet radiation penetrates more deeply into the Z302 layer than infrared radiation, in which case UV radiation would be more efficiently absorbed. A third possibility is that the wavelength dependence of the refractive index of Z302, whose variation with wavelength is currently unknown, is sufficient to produce this difference. Finally, there remains the possibility that observed differences in reflectivity are related to preparation of the samples (thickness, surface topography, etc.). Until these details are routinely reported along with BRDF data, this will always remain as an unsampled source of uncertainty. Clearly, this question merits further investigation.

## 6. CONCLUSION

Presented is the design and implementation of an automated goniometer reflectometer conceived to measure the BRDF of glossy, low-reflectivity coatings. The instrument is used to measure the BRDF of Z302, a commercial coating widely used in aerospace and optical applications to reduce scattered light and enhance absorptivity. Measurements are reported for both the p- and s-polarizations at 350 nm. We believe that this is the first report of BRDF measurements for Z302 in the UV and for polarized light at any wavelength. For comparison, we provide previously unpublished unpolarized BRDF measurements of Z302 at 633 nm, and note that these values exceed our 350-nm values by more than an order of magnitude. We conclude that additional investigation is needed to explain this difference.

## ACKNOWLEDGMENT

The authors acknowledge the Climate Sciences Branch of the Science Directorate of NASA's Langley Research Center for its financial and technical support of the effort reported here. We are especially grateful for permission to use the previously unpublished data in Fig. 7.

## REFERENCES

- [1] Mahan, J. Robert, *Radiation Heat Transfer: A Statistical Approach*, Wiley (2002)
- [2] Persky, M. J., Review of black surfaces for space-borne infrared systems, *Review of Scientific Instruments*, **70**(5), pp. 2193-2217 (May 1999)
- [3] Prokhorov, A., and N. I. Prokhorova, Application of the three-component bidirectional reflectance distribution function model to Monte Carlo calculation of the spectral effective emissivities of nonisothermal blackbody cavities, *Applied Optics*, **51**(33), pp. 8003-8012 (November 20, 2012)
- [4] Mahan, J. R., N. Q. Vinh, and K. J. Priestley, An application of the Monte Carlo ray-trace method with bidirectional reflection, *Paper TFEC-2018-22038, 3<sup>rd</sup> Thermal and Fluids Engineering Conference*,
- [5] Shure, Mark, RBI Z302 BRDF Test Data with Unlimited Rights, Harris Space and Intelligence Systems Memo RBI-18-182 to Dr. Kory J. Priestley, NASA LaRC (October 3, 2018)
- [6] Shirsekar, Deepali, Yifei Wang, J. Robert Mahan, Kory J. Priestley, and Vinh Q. Nguyen, Bidirectional reflectance measurement of black absorber layers for use in optical instrument design, *Proceedings of SPIE, Optical Modeling and Performance Predictions X*, **10743**(03), pp. 1-7 (September 17, 2018)
- [7] Mahan, J. Robert, *The Monte Carlo Ray-Trace Method in Radiation Heat Transfer and Applied Optics*, Wiley (2018)
- [8] Shirsekar, Deepali, *Measurement and Modeling of Z302 BRDF*, Master of Science Thesis, Virginia Tech (December 2018)
- [9] Shiffer, Ralf, Reflectivity of a slightly rough surface, *Applied Optics*, **26**(4), pp. 704-712 (February 1987)