PERSPECTIVE

Reporting Performance in Organic Photovoltaic Devices

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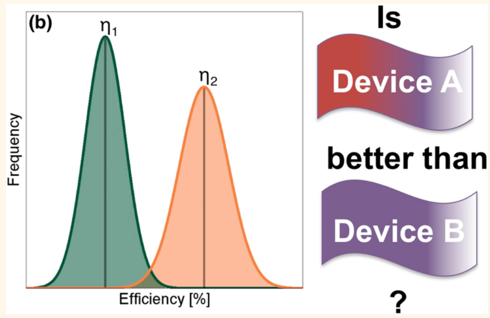

ABSTRACT Research into organic photovoltaics (OPVs) is rapidly growing worldwide because it offers a route to low temperature, inexpensive processing of lightweight, flexible solar cells that can be mass manufactured cheaply. Unlike silicon or other inorganic semiconductors (*e.g.*, CdTe, CIGs), OPVs are complicated by the requirement of having multiple materials and layers that must be integrated to enable the cell to function. The enormous number of research hours required to optimize all aspects of OPVs and to integrate them successfully is typically boiled down to one number—the power conversion efficiency (PCE) of the device. The PCE is the value by which comparisons are routinely made when modifications are made to devices; new bulk heterojunction materials, electron- and hole-transport layers, electrodes, plasmonic additives, and many other new advances are incorporated into OPV devices and compared with one, or a series of, control device(s). The concern relates to the statistical significance of this all-important efficiency/PCE value: is the observed change or improvement in performance truly greater than experimental error? If it is not, then the field can and will be misled by improper reporting of efficiencies, and future research in OPVs could be frustrated and, ultimately, irreversibly damaged. In this Perspective, the dangers of, for instance, cherry-picking of data and poor descriptions of experimental procedures, are outlined, followed by a discussion of a real data set of OPV devices, and how a simple and easy statistical treatment can help to distinguish between results that are indistinguishable experimentally, and those that do appear to be different.

The past decade has seen an enormous surge of publications in the area of organic, nanoparticle, and dye-sensitized photovoltaics.[1−3] From the design and synthesis of low-band gap polymers[4,5] and small molecules[6,7] that capture larger swaths of the solar spectrum, to solution-processed nanoparticle-based photovoltaic cells,[8,9] to implementation of flexible plastic substrates,[10] to addressing stability,[11] among many other interesting and exciting directions,[12] the field is growing rapidly. These relatively new directions in photovoltaics are attractive for a number of reasons: (i) they are soundly based in nanoscience, materials science, and surface chemistry, which is a natural and compelling application of the enormous body of research built up over the last few decades, and (ii) there is a great deal of personal satisfaction directing one's research to an enormous challenge that many see as central to humanity's very survival.[13] There is, however, a steep price to be paid for working in photovoltaics that is familiar to anyone working in the area, which is having to continually respond to the following question:

"What is the efficiency of your solar cell?"

The entire area of photovoltaics research is under enormous pressure to condense many person-years of research effort into a single metric of performance, the power conversion efficiency, which is a number that provides no indication of reliability, reproducibility, yield, lifetime, or related statistics. Incremental improvements in device performance may, as a result of this pressure, be more highly valued than important findings related to understanding fundamental properties, mechanisms, and exploration of creative new ideas in photovoltaics. Even more worryingly, we have become very concerned that within the area of photovoltaics research, poor (or no) statistical treatment is the norm. Given the unavoidable and significant variability in device performance, despite best efforts at achieving identical processing conditions, it is impossible to assess the validity of declared improvements in device performance without some basic statistical treatment. As such,

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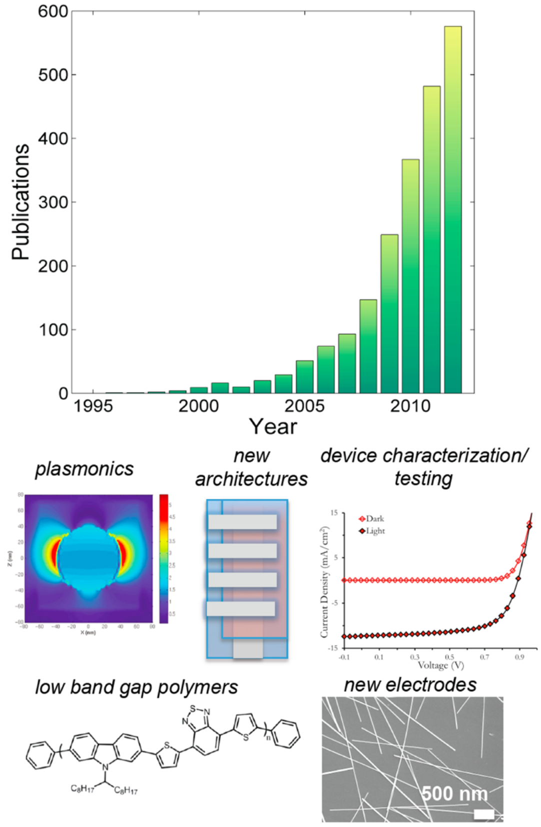

Figure 1. Number of publications per year, according to Web of Science, found using the search term "organic photovoltaics", accompanied by examples of the diverse and emerging areas in organic photovoltaics that are based upon developments in nanoscience, materials, and polymer science. The image representing plasmonics is reprinted from ref 17. Copyright 2011 American Chemical Society.

the inclusion or use of a new material may appear to improve device performance; when simply comparing average efficiencies, however, after analysis, it may be found that the measured differences are statistically insignificant from each other. If such proposed improvements in device performance are not subjected to the necessary statistical tests, then future researchers can be misled, and valuable resources wasted. As stated in a piece in *Nature* from April 2013, "Reproducibility separates science from mere anecdote."[14] Anecdotes will not bring photovoltaics to fruition, and worse, could lead to frustration of researchers in the field and the premature demise of not-yet-commercialized technologies such as organic photovoltaics (OPVs). Sloppy reporting of results also results in false claims appearing in the media, which further erode public trust in science when they are shown to be wrong, or simply do not lead to meaningful progress the field.

When reporting improvements in photovoltaic performance, it is critical to provide sufficient information in the form of statistics and experimental conditions to enable the work to be reproduced, and to allow the reader to evaluate the reliability of the data. The first sin of the "The Seven Sins in Academic Behavior in the Natural Sciences" is "A poor or incomplete description of the work," rendering it impossible for future readers to determine its significance accurately or to replicate it.[15] The third related sin is "Insufficient connection between data and hypothesis or message, leading to lack of support for the message or over-interpretation of data"; overinterpretation of data is certainly going to be a problem when statistically insignificant data is thoroughly wrung dry to draw seemingly concrete, but unsubstantiated, conclusions.[15] And last, the fourth sin, "The reporting of only favorable, positive, or desired results,"[15] manifests itself in the photovoltaics world as "cherry-picking"—as objective as scientists need to be, without a neutral, cold, and broadly applied statistical method that is used without fail, reporting only the best, and possibly irreproducible, efficiencies will be simply too tempting for many.

If proposed improvements in device performance are not subjected to the necessary statistical tests, then future researchers can be misled, and valuable resources wasted.

An excitonic OPV device is complex, and is built up from multiple layers that are composed of two electrodes, at least one which must be optically translucent, a morphologically complex nanoscale mixture of a donor and acceptor, various interfacial layers to enable charge extraction, and perhaps metal nanoscale plasmonic materials designed to enhance device performance.[16,17] These structures are by their very nature intricate, requiring a great deal of expertise, time, resources, and effort to produce functional devices. As shown in Figure 1, the number of papers appearing in the ISI citation database, using the representative search term "organic photovoltaics" shows a dramatic increase in publications over the past 5–7 years.[18] The area has become incredibly interdisciplinary and is drawing researchers together from disparate areas of science and engineering, which is extremely

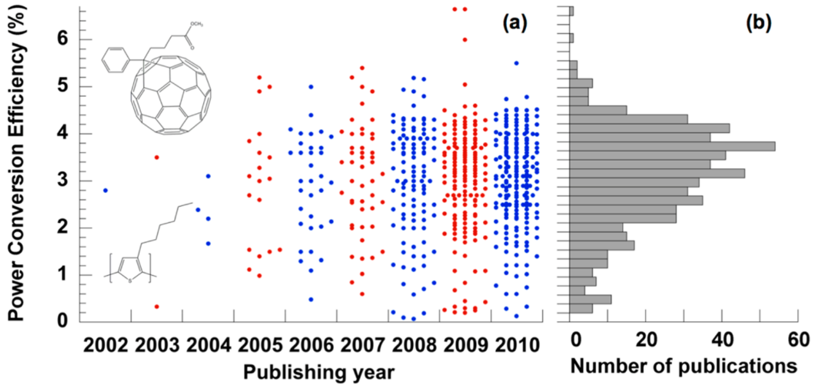

Figure 2. (a) Power conversion efficiencies (PCEs) of the P3HT/PC_{61}BM bulk heterojunction (BHJ) combination from 579 papers up to 2011, and (b) a histogram representation of the number of publications using this BHJ combination versus PCE. Reprinted with permission from ref 22. Copyright 2011 Wiley-VCH.

promising for the future of OPVs. The lack of consensus, however, on how to report data, results, and experimental conditions in the area of OPVs could completely undermine the long-term health of the field.

One of the most basic and fundamental approaches in experimental science to improve an existing system is to modify one variable at a time systematically, and to examine the outcome. In OPVs, a similar approach has been successfully followed in the area of low-band gap polymers, where several series of polymers have been designed, synthesized, and characterized, and then integrated into OPV devices, for example.[19] If higher efficiency OPV devices are the goal, with each modification, researchers must decide whether or not polymer A is the same as, better, or worse than polymer B. In the language of statistics, the researchers must decide whether the data support the null hypothesis (that there is no difference in efficiency between polymers A and B) or the alternative hypothesis (that there is real difference in efficiency between polymers A and B). By computing a statistical quantity known as the *p*-value, it is possible to assess whether the data support the null or alternative hypotheses. As a result, statistics are an absolute necessity to determine if a given change provides a real improvement in device performance.

As a starting point in the field of OPVs, we encourage all researchers to carefully read two important synopses that focus on: (i) accurate measurement and calibration of OPV devices,[20] and (ii) the requirements for reporting solar cell efficiencies and external quantum efficiencies (EQEs), particularly for power conversion efficiencies that approach and exceed thermodynamic limits, and those that are on the low end for a particular photovoltaic technology.[21] These papers provide detailed explanations regarding the instrumentation setup for characterization and testing, and details regarding light source and calibration, EQE measurements, and efficiency calculations, data, and information that need to be provided to readers:

(i) Accurate Measurement and Characterization of Organic Solar Cells[20]
(ii) Reporting solar cell efficiencies in *Solar Energy Materials and Solar Cells*[21]

The concerns expressed in this Perspective relates to the handling of device statistics, and reliability of these data—are the changes or improvements statistically relevant, and thus "real"? In OPVs, the most widely studied acceptor/donor combination is P3HT/PC_{61}BM, as shown in Figure 2. In 2011, Wantz and coworkers compiled the results of 579 P3HT/PC_{61}BM bulk heterojunction (BHJ) OPV devices, and noted that the reported power conversion efficiencies (PCE) varied from just over 0% to 6.5%, with an average of 3−4%.[22] Interestingly, P3HT/PC_{61}BM cells prepared by Konarka, Plextronics, and Sharp gave certified PCEs on the order of 3.5−4.0%.[22] The scatter of efficiencies, however, is remarkable and leads to questions of how one can make comparisons between different cells, how much of an improvement is real, and how one determines "how much" is significant?

Many reports in the OPV field are focused on the improvement of solar cell efficiencies *via* modification of processing conditions (solvent/thermal annealing), interfacial layers, or thickness optimization. For these types of investigations, it is critically important that the authors report statistically significant results if they wish to make any relevant conclusions about the efficacy of a given modification to the solar cell. As an example, the following hypothetical experiment illustrates a commonly used analytical approach in the OPV literature, which can lead to unsupported conclusions. Suppose we wish to investigate a new interfacial layer to be used instead of the workhorse, PEDOT:PSS. In order to determine if the new interfacial layer improves solar cell efficiency, a reference cell using PEDOT:PSS and one where the PEDOT:PSS is replaced with the new interfacial layer are fabricated and tested. It is found that the cell with PEDOT:PSS has an efficiency of 3.4%, while the cell

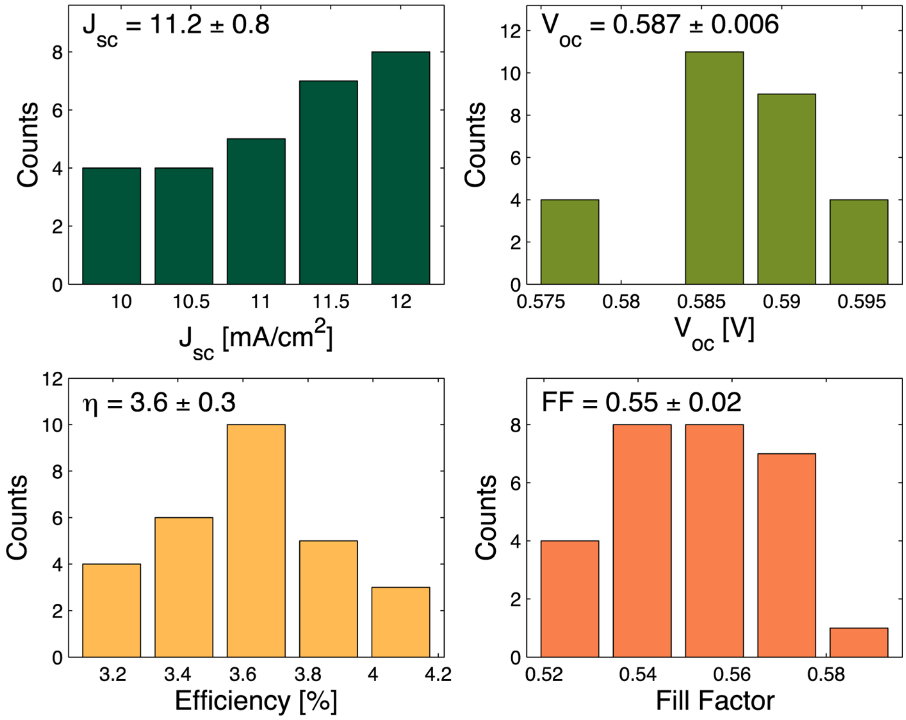

Figure 3. Histograms of device parameters (J_{sc}, V_{oc}, η, and FF) for a set of ITO/PEDOT:PSS/P3HT:PC_{61}BM/LiF/Al cells, composed of 28 separate devices.

using the new interfacial layer has an improved efficiency of 3.7%. In addition to solar cell testing, the PEDOT:PSS and new interfacial layer are characterized using a variety of analytical techniques. In particular, the work functions of each interfacial layer are measured using ultraviolet photoelectron spectroscopy (UPS), where it is found that the work function of the new interfacial layer is 0.2 eV greater than that of PEDOT:PSS. From these data it is concluded that the new interfacial layer provides superior power conversion efficiency as a result of a reduced hole injection barrier. The paper is then written, submitted, reviewed, and published.

Despite seemingly convincing results, these conclusions are not supported by the data. Shown in Figure 3 are histograms displaying the distribution of device parameters (J_{sc}, V_{oc}, η and FF) for a set of ITO/PEDOT:PSS/P3HT:PC_{61}BM/LiF/Al standard cells, composed of 28 separate devices prepared at the same time. Despite best attempts to achieve identical processing conditions for each cell, we see that the histogram of device efficiencies resembles a normal distribution, and has standard deviation of 0.3%, which is typical of a batch of cells produced in our lab, and most laboratories. Subtle variations in the thickness of the various layers, particularly the BHJ, can have profound effects on device performance.[23] From these data it is clear that one cannot make meaningful conclusions about improved device efficiency by comparing *individual* devices. In our previous example, it was found that the PEDOT:PSS device had a power conversion efficiency of 3.4%; if, however, device efficiencies are assumed to be similarly distributed to those in Figure 3 and we fabricated another PEDOT:PSS device, it is just as likely to have an efficiency of 3.8%, which would dramatically change the previous conclusions. The conclusions would be the opposite—that the new interfacial layer was worse than PEDOT:PSS! Figure 4 attempts to summarize this conundrum: if there is significant overlap between the intrinsic distributions of the power conversion efficiency for two different types of devices, then determining if an improvement in device performance is "real" becomes challenging.

> Statistics are an absolute necessity to determine if a given change provides a real improvement in device performance.

In general, if we are to conclude that solar cell efficiency is improved (or worsened) by changing an aspect of the device architecture or processing, it is necessary to measure the PCE of multiple devices; from these data the *p*-value can be computed, which is then used to evaluate the statistical significance of the results. The use of *p*-values is best explained by continuing with our previous example of changing interfacial layers. Suppose that we have measured the PCE of a large number of ITO/PEDOT:PSS/P3HT:PC_{61}BM/LiF/Al standard cells and, as such, we find that it is normally

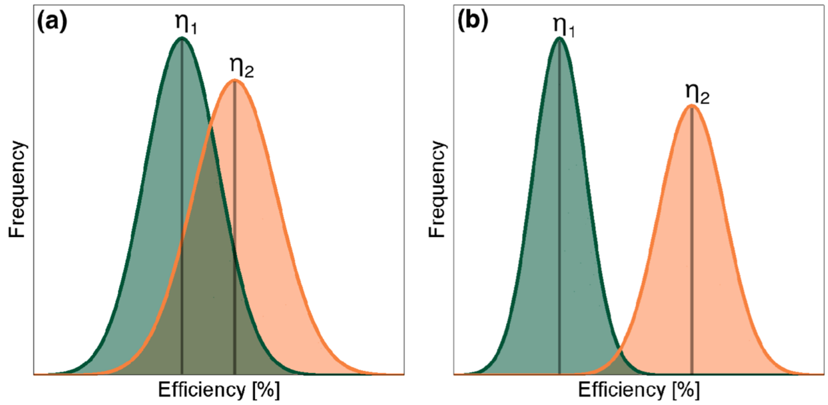

Figure 4. (a) Distribution of device power conversion efficiencies (PCEs) for two different processing methods of an organic photovoltaic device. Process 1 (green) has an average PCE of η_1 with a standard deviation of σ_1, while process 2 (orange) has average PCE of η_2 and a standard deviation of σ_2. There is significant overlap between the two distributions, making it difficult to determine if process 2 will reliably provide improved PCEs. (b) As the standard deviation of both processes are reduced there is minimal overlap between the distributions, and process 2 clearly is the cause of improved PCEs.

distributed and the "true" value of the average PCE is $\eta_0 = 3.5\%$. Now $N = 5$ devices are fabricated using the new interfacial layer, where it is found that the average PCE is $\eta_1 = 3.7\%$, with a standard deviation of $\sigma_1 = 0.3\%$. We now wish to determine the probability that a set of 5 standard cells will have an average PCE of 3.7% or greater—this probability is called the p-value. If the p-value is lower than some level of desired statistical significance, α (typically 5% or $\alpha = 0.05$), then we can conclude with 95% confidence that the average PCE of cells fabricated using the new interfacial layer is greater than 3.5%. To calculate the p-value, we must first determine the Z-score, which is the number of standard deviations η_1 is from η_0, given by

$$Z = \frac{\eta_1 - \eta_0}{\sigma_1/\sqrt{N}}$$

In this case, a value of $Z = 1.49$ is obtained. Using a simple look-up table or an online calculator, the probability of being 1.49 standard deviations (or greater) away from the norm is $p = 0.136$. Therefore, these results do not reach our desired level of statistical significance and we cannot conclude that the use of the new interfacial layer will provide a higher average PCE than the standard cells. Conversely, if 20 cells are fabricated and the average PCE is found to be $\eta_1 = 3.7\%$, with a standard deviation of $\sigma_1 = 0.4\%$, the p-value will be $p = 0.025$. This would allow the authors to conclude that cells made with the new interfacial layer will have a higher average PCE than the standard cells.

Recommendations. The goal of clearly providing this information on device performance is not to test the manufacturing capabilities of a particular laboratory, but to enable others to judge for themselves the statistical significance of a reported advance.

How Many Devices? Drawing from the previously discussed example, two important recommendations can be made with regard to the number of devices that are necessary to make useful comparisons between different device processing/architectures. First, it is critical to have excellent statistics for the control devices, *i.e.*, your *standard cells*. Since this group of standard cells forms the statistical foundation for all other comparative improvements, ideally this would be upward of $N = 100$ standard cell devices in order to ensure that the true distribution of device PCEs for the control devices is known and understood. It is noted that this is a large number of experiments, but it would only need to be done periodically, and can be used as the basis for several different ongoing projects in a lab. In order to determine the number of non–standard cells (*new* cells) that need to be fabricated for statistically relevant results, it necessary to have preliminary results to provide some indication of the magnitude of the change (and improvement or decrease) in device performance. One would need to have a working/expected efficiency and standard deviation of the new process/architecture. For instance, if a 0.2% increase in efficiency with a standard deviation of 0.4% appears to be the case, then $N = 16$ devices would produce a p-value less than 0.05. Moreover, histograms like those shown in Figure 3 are useful to supplement tabular data since the reader can easily grasp the spread and scatter within the data. If one is limited, however, to a small number of devices due to scarce materials or limited instrumentation access or the like, the number of devices should be stated clearly, and readers should be allowed to arrive at their own conclusions. In tables of device characteristics (efficiencies, fill factors, *etc.*), indicate the standard deviation, the number of cells measured, and what statistical approach was used (*e.g.*, the average with highest and lowest values, the standard deviation, *etc.*).

How Many Significant Digits? Significant digits are directly related to the precision of the measurement, where precision is the stabililty of the measurement when repeated many times. A precise measurement may not necessarily be an accurate measurement (*i.e.*, the closeness to the *actual* value), but provides information on the variance of the measurement. While a galvanostat/potentiostat may provide a large number of significant digits, one is, of course, limited by the least precise measurement. Device area has been singled out repeatedly as a large source of error, due to shadowing and edge effects, edge roughness, and other related factors. While each experimental setup will be different, reporting efficiencies, short circuit currents, fill factors, and open circuit potentials with precisions better than 1/10 of the unit provide little useful information. An efficiency of 7.1% is most likely sufficient; 7.12% is wishful thinking, at best.

Having a "Bad Cell Day"? Every lab will produce "bad" devices. Bad cells are those that have shorted, malfunctioned due to an experimental error, suffered from contamination, or did not work for no obvious reason. In another common scenario, one may have 10 "good" cells and two "bad" ones; can the two "bad" ones simply be ignored, and under what circumstances? Every laboratory analyzing large amounts of data needs to adopt a neutral and systematic method of analyzing data to enable discarding spurious results. Discarding data needs to be considered very carefully, and should be performed with hesitation and a great deal of caution. A systematic method, however, eliminates the need for guessing and data cherry picking that can make a data set appear to be stronger/better than it actually is. A simple method that can be easily applied is Chauvenet's Criterion, which states that for a sample data set of N measurements, if the probability, $P(x_i)$, of obtaining a given data point, x_i, is less than is $0.5/N$, then it can be rejected from the data set. Chauvenet's Criterion is one of many possible methods for data rejection, and regardless of which method is used, it should be clearly stated how and if data rejection was performed. Take, for example, the following set of efficiencies for a series of six cells:

$$\eta = 6.1\%, 4.9\%, 5.6\%, 5.2\%, 5.4\%, 3.9\%$$

At first glance, the 3.9% efficiency looks low, but can it be discounted? Using the same logic, the same question could be asked of the 6.1% efficiency cell, but human tendencies would probably not be biased toward eliminating the higher value. As a result, a systematic approach is required to data analysis that will provide the basis for elimination of a piece of data. Below we apply Chauvenet's Criterion to the above data set.

The average of this data set is

$$\eta_{\mathrm{AVE}} = 5.183\%$$

The standard deviation of this sample data set is $\sigma = 0.7468\%$

If we assume that the data are normally distributed, then the probability of obtaining a given efficiency can be estimated by computing how many standard deviations from the mean it is. For example, the measured efficiency of 3.9% is 1.719 standard deviations below the mean. Therefore, the probability of obtaining a value that is 1.719 standard deviations from the mean for a normally distributed variable is 0.0857 (easily determined from online software or a standard Z-score table), which is greater than $0.5/N = 0.5/6 = 0.0833$. As such, the 3.9% efficiency value must be kept. Note that more rigorous methods, such as Pierce's Criterion, are recommended;[24] however, Chauvenet's Criterion is presented as an example here due to its simplicity.

Experimental Details. When writing an experimental section, be sure to include all necessary information to enable future readers to reproduce your results. The problem of reproducibility in the scientific literature may be more systemic than most realize.[15] Have an experienced colleague walk through your experimental section before submission for publication—are there any points of confusion or lack of clarity? Would someone not familiar with your work, but nonetheless skilled in the art, be able to achieve the same results?

CONCLUSIONS

For synthetically minded groups designing and producing new materials for OPVs, the statistical analyses outlined here may seem onerous. Applying these simple calculations to an existing data set will demonstrate that they are, in fact, simple and quick to carry out. Many other areas of research, particularly medicine, must consistently use rigorous and systematic analyses in their studies, due in part to the diversity within humans and other living subjects.[25] The experimental challenges in actually (re)producing the architectures required for a functional OPV device in these exploratory systems are significant, and thus, careful data analysis is also required. Other closely related fields of science are also grappling with comparative significance, such as the electrochemical storage community.[26] The danger of leading the field astray by reporting experimentally insignificant data is too great not to take the time to repeat one's experiments, with full experimental disclosure. Perhaps a useful ending is a reminder from the wisdom of Richard Feynman:[27]

> *The first principle is that you must not fool yourself—and you are the easiest person to fool. So you have to be very careful about that. After you have not fooled yourself, it is easy not to fool other scientists. You just have to be honest in a conventional way after that.*

Conflict of Interest: The authors declare no competing financial interest.

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