

Dr. Radislav A. Potyrailo

- MS Optoelectronics (Kiev Polytechnic Institute), PhD Analytical Chemistry (Indiana Univ.)
- Principal Scientist at GE Research, leads sensors programs for industrial/consumer markets
- Principal Investigator on programs funded by GE, industrial partners, US Government
- 125+ granted US Patents on sensing concepts/implementations to solve monitoring needs
- Examples of commercialized sensors for GEbusinesses: GEPlastics, GEWater, GEOil & Gas
- 150+ publications, 10+ keynote/plenary/lectures
- Examples of contributions to scientific community:
 - Editor of the Springer-Nature book series *Integrated Analytical Systems*
 - Chair of the MEMS and Sensors Industry Group (MSIG) Device Working Group
 - North America Regional Chair of International Society for Olfaction and Chemical Sensing
 - Initiator of the First Gordon Research Conference on Combinatorial Materials Science
- Recent recognitions include SPIE Fellow, Prism Award by Photonics Media/SPIE

GE Research Electronics & Sensing

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>> Stacey Standridge: Good afternoon, and thanks for tuning in for today's webinar, "Nanotechnology For A New Generation of Gas Sensors: And Industrial Perspective on Fundamental, Applied, and Commercialization Aspects." My name is Stacey Standridge. I am Deputy Director of the National Nanotechnology Coordination Office, and I will be the moderator today. This webinar is the first in our quarterly webinar series in support of the "Nanotechnology for Sensors and Sensors for Nanotechnology" Signature Initiative, and I'm happy to introduce our guest today, Dr. Radislav Potyrailo. Dr. Potyrailo is a principal scientist at GE Global Research, leading the sensors programs for industrial and consumer markets with commercialized sensors for businesses, including GE Plastics, GE Water, and GE Oil and Gas. Dr. Potyrailo has been a principal investigator on programs funded by National Nanotechnology Initiative participating agencies, including DoD, DOE, DHS, NIH, and NIOSH. Efforts from these and other programs have resulted in excess of 125 granted U.S. patents and 150 publications on transducer technologies, sensing materials, and data analytics describing sensing concepts and their implementations. You can see a bit more about his background on the slide in front of you. I will also say that Dr. Potyrailo's recent recognitions included being named SPIE Fellow and receiving the Prism Award by Photonics Media at SPIE. (Continued...)

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>> Stacey Standridge: In the webinar today, Dr. Potyrailo will first discuss possibilities for new principles of gas sensing based on nanomaterials and nanostructures. Then, he will discuss the development of sensors capable of quantifying individual chemical components in mixtures, rejecting interferences, and enhancing response stability in wearable, stationary, and other formats. If you have questions as the presentation is going along, please feel free to send those to webinar@nnco.nano.gov, or you can use the “Submit Your Questions Here” window in the webinar interface. If you would like more information about the Signature Initiatives and the Federal resources supporting the development of nanosensors, or if you would like information about upcoming events like National Nanotechnology Day, which is taking place next week on October 9, please feel free to visit our website at nano.gov. And with that, I am happy to hand the floor over to Dr. Potyrailo.

>> Radislav Potyrailo: Thank you, Stacey, and hello everybody! Good morning, and good day! I'm glad to discuss today with you this topic. You can see from the first slide that I'm at the GE Global Research Center, an organization that is developing new types of electronic and sensing technologies. So, I will be going through the slides in such a way as to give you the perspective of the state-of-the-art of gas sensors now, and how nanotechnology solutions are advancing this state-of-the-art quite significantly.

Nanotechnology for a new generation of gas sensors: An industrial perspective on fundamental, applied, and commercialization aspects

National Science and Technology Council, National Nanotechnology Initiative
Sensors NSI Webinar Series
OCTOBER 02, 2019

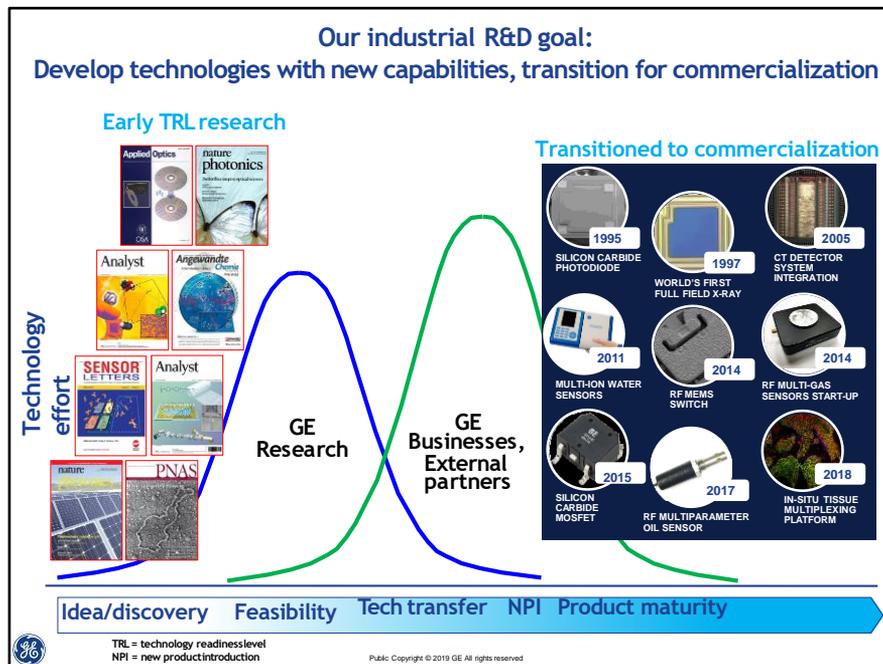
Dr. Radislav A. Potyrailo (GE Research, USA)



Image: GE (R.A. Potyrailo)

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>> Radislav Potyrailo: This is the title of the presentation.



>> Radislav Potyrailo: Because we are an industrial research organization, we are not only focusing on innovative aspects of sensor technologies (you can see on the left of the slide the early technology readiness level (TRL) research that we do), but also working with our GE partners and organizations in the United States and around the world to transition our findings into successful commercial products.

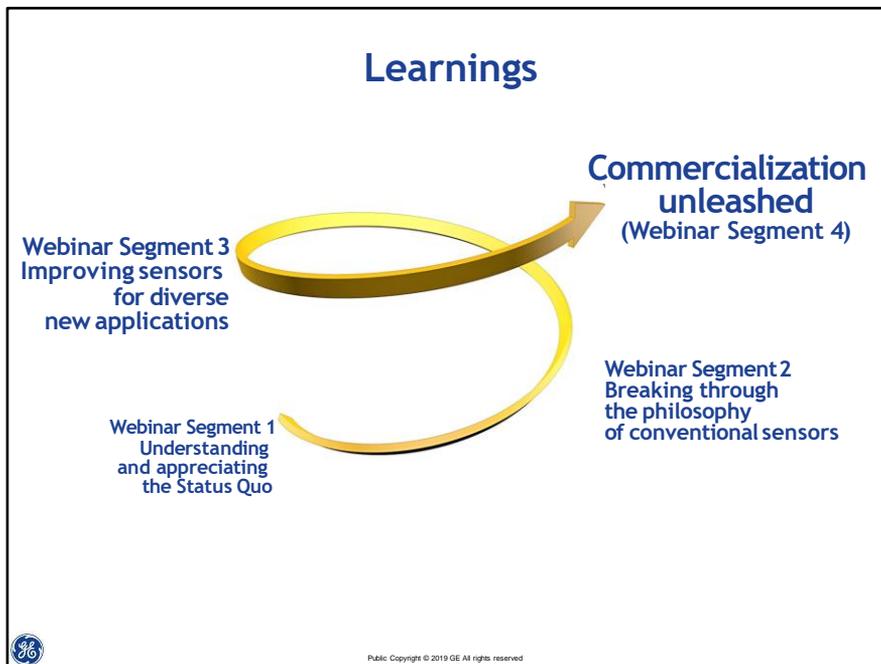
Webinar outline

- Segment 1:** Understanding and appreciating the Status Quo
- Segment 2:** Breaking through the philosophy of conventional sensors
- Segment 3:** Improving sensors for diverse new applications
- Segment 4:** Commercialization unleashed

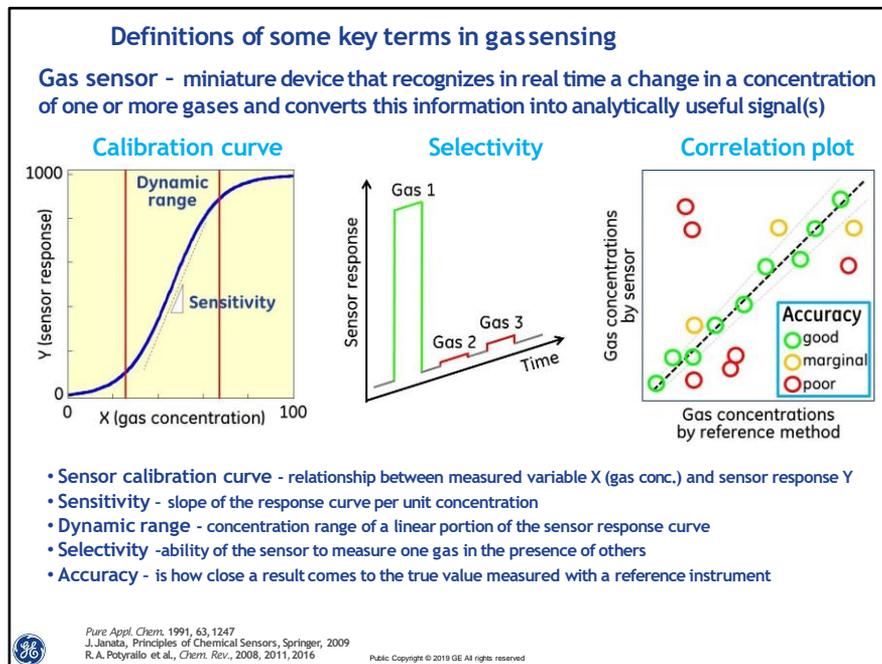


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>> Radislav Potyrailo: The webinar is organized in four segments, outlined here. Segment 1 is giving us the ability to learn the basics, to understand the state-of-the-art now (I call it the “Status Quo”). Segment 2 will demonstrate how to break through the philosophy of conventional sensors. Segment 3 will show us how we can improve the needed capabilities of gas sensors, what types of improvements are still required, and how we can take advantage of all these learnings. Segment 4 will focus on commercialization.



>> Radislav Potyrailo: I view the four segments as a spiral of learning, because the basics that we are learning in Segment 1 are expanded into the other segments. At the end, we have a lot of knowledge, which helps us move the technology forward.

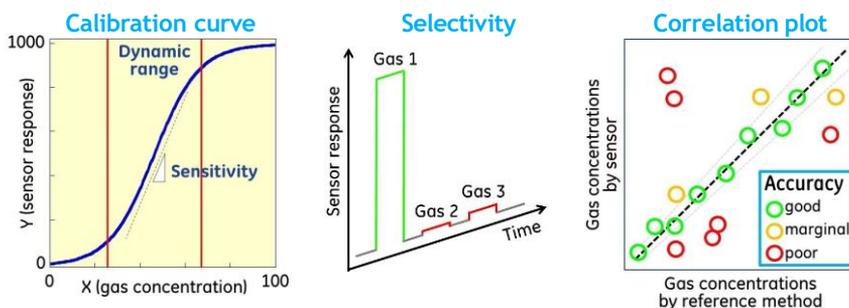


>> Radislav Potyrailo: It's very important to define what gas sensors are early, because that will give us the ability to understand more clearly the examples that I will be showing in this presentation. For this community, a gas sensor is a device, and this device recognizes changes in the concentration of one or more gases in real time by continuously doing measurements and by converting the information that is contained in the device into analytically useful signals.

On this slide, I highlight important aspects of gas sensors. To the left, the calibration curve is the relation between the gas concentration and the sensor output, or sensor response. As an example, I am showing you one output per sensor. The dynamic range is the linear portion of this calibration curve, and the slope of this response is known as the sensitivity. In the middle, I show the selectivity, which is another important aspect of a gas sensor. The selectivity is related to how nicely I can detect the gas of interest, which I am labelling "Gas 1," in the presence of other types of gases. So, suppressing the effects on my sensor response is a very important aspect of being highly selective, or selective enough. (Continued...)

Definitions of some key terms in gas sensing

Gas sensor - miniature device that recognizes in real time a change in a concentration of one or more gases and converts this information into analytically useful signal(s)



- **Sensor calibration curve** - relationship between measured variable X (gas conc.) and sensor response Y
- **Sensitivity** - slope of the response curve per unit concentration
- **Dynamic range** - concentration range of a linear portion of the sensor response curve
- **Selectivity** - ability of the sensor to measure one gas in the presence of others
- **Accuracy** - is how close a result comes to the true value measured with a reference instrument

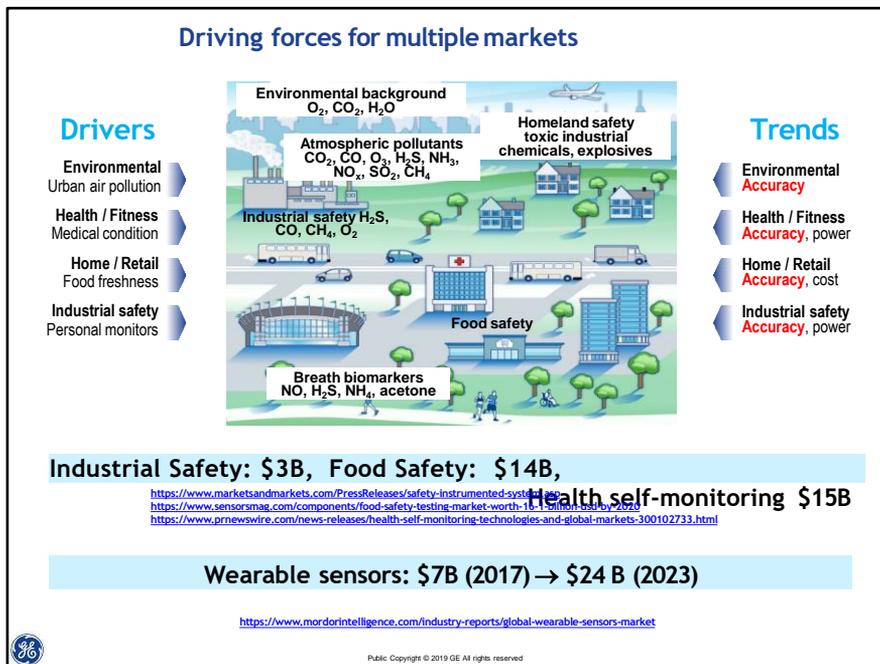


Pure Appl. Chem. 1991, 63, 1247
J. Janáček, Principles of Chemical Sensors, Springer, 2009
R.A. Potyrailo et al., Chem. Rev., 2008, 2011, 2016

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>> Radislav Potyrailo: To the right, I show another very important aspect of gas sensors: the correlation plot. On one axis, you're doing the gas measurement by a reference method – the method that you trust. It may be quite expensive but, very importantly, such a graph will give you an idea of how good the sensor is in relation to a trusted method. The closer the data points are to this line (with a slope of one), such as the green points in this example, the better the correlation is and the more successful the sensor will be in different applications. I am also showing points with other colors: yellow points, with marginal accuracy (or marginal correlation), and red points, with poor correlation.

I am spending a lot of time on this because you will see examples from the field, and you will judge for yourself how they look like. These are very important aspects of gas sensors. The line with slope one will give you the quality of the fit. Typically, people call that quality the r^2 value. So, if $r^2 = 1$, then all of my data points line up extremely well into this line.



>> Radislav Potyrailo: This slide shows that in the air, many gases are present. In this example, I am showing ambient air. You can see the environmental background and different types of pollutants. If you are in an industrial setting, then there would be industrial emissions, as well. The drivers behind the development of different types of gas sensors are highlighted on the left, and the trends are highlighted on the right. You can see that I highlighted the word “Accuracy” in red, because the end users and applications are calling for adequate accuracy. I'm not saying 100% accuracy, but “adequate” is a very important aspect of that.

Another important aspect is that the driving forces are different types of markets that are interested in having that new gas-sensing solution. You can see that industrial safety markets are around \$3 billion, food safety \$14 billion, and health self-monitoring \$15 billion. So these are very important, sizable driving forces that allow us to focus on impactful directions and provide technical solutions. Also, at the bottom of this slide, I am showing that wearable sensors (the ones that will have the high-quality performance at low power and form factors) are also quite significant markets, so these are very important driving forces.

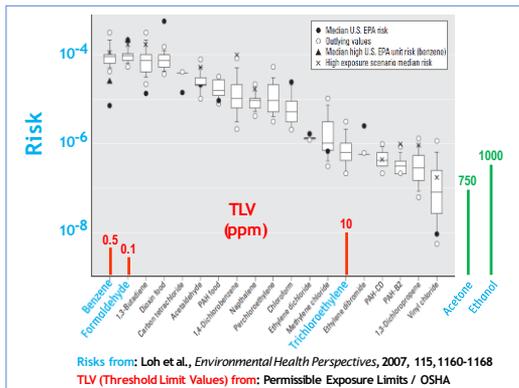
Important health-related volatiles: outdoor and indoor environments

Major outdoor volatile pollutants

Ozone	< 0.1 ppm
Nitrogen dioxide	< 0.2 ppm
Sulfur dioxide	< 0.2 ppm
Carbon monoxide	<30 ppm

Brunekreef, Holgate, *Lancet* 2002, 360, 1233-1242
who.int/ceh/capacity/Outdoor_air_pollution.pdf
euro.who.int/document/e71922.pdf
who.int/airpollution/ambient/pollutants/en/

Indoor volatiles of importance



- Some volatiles are much more toxic than others
- Toxic volatiles often are at much lower levels than benign volatiles
- Need to detect toxic volatiles in complex background of benign volatiles



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>> Radislav Potyrailo: This slide highlights a subset of the volatiles from the previous slide. I am highlighting the volatiles that I (and many people) care about, because of health concerns about these volatiles. On the left, I am showing the top four volatiles from ambient outdoor air, and you can recognize that these are ozone, nitrogen dioxide, sulfur dioxide, and carbon monoxide. You heard about them a lot, and you can see that their allowed levels are still on the borderline of being dangerous to health. Ozone, nitrogen dioxide, and sulfur dioxide are less than one part per million, and carbon monoxide is less than 30 parts per million. These values are important to keep in mind. So, out of all kinds of gases that you saw on the previous slide, these are important outdoor pollutants for health.

On the right, I am showing data from the literature about indoor volatiles. This graph shows on the y-axis the risks of different types of volatiles; the x-axis is highlighting many examples of those volatiles. Those highlighted in blue show the levels of so-called threshold limit values (TLVs) – permissible exposure limits, in parts per million. (Continued...)

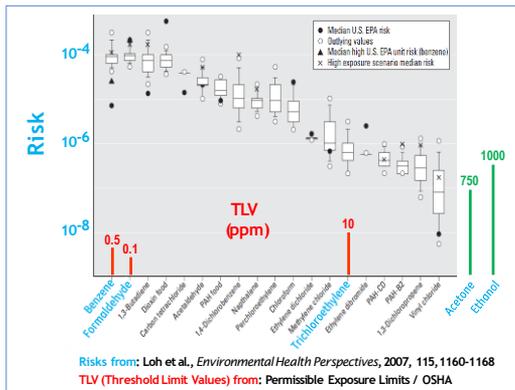
Important health-related volatiles: outdoor and indoor environments

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Brunekreef, Holgate, *Lancet* 2002, 360, 1233-1242
who.int/ceh/capacity/Outdoor_air_pollution.pdf
euro.who.int/document/e71922.pdf
who.int/airpollution/ambient/pollutants/en/

Indoor volatiles of importance



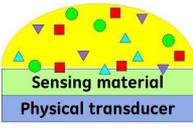
- Some volatiles are much more toxic than others
- Toxic volatiles often are at much lower levels than benign volatiles
- Need to detect toxic volatiles in complex background of benign volatiles



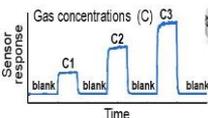
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>> Radislav Potyrailo: How much are different volatiles allowed to be present before they become dangerous to human health? You can see that some volatiles can be dangerous after they reach very small levels (less than one part per million), such as benzene and formaldehyde. Other ones can be dangerous only after they reach higher concentrations, such as ethanol and acetone (from nail polish remover, for example). What this means (the punch line for this graph) is that in the presence of all kinds of volatiles, we need to focus on the ones that are really harmful to our health and, in the presence of other volatiles, we need to focus on the ones that are less harmful but are present in maybe four orders of magnitude higher concentrations. Resolving the low concentrations may be a challenge. The take-home message is that if we would like to reliably detect these types of toxic volatiles, we need to do it in a complex background of volatiles that are much more benign.

Conventional gas sensors: Serving sensing needs in established markets



Sensing material
Physical transducer



Gas concentrations (C) C1 C2 C3
Sensor response
Time



- mature technologies
- widely available
- Interchangeable
- inexpensive

alphasense.com
amphenol-sensors.com

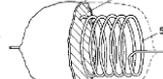
Understanding the roots: single-output devices for detection of expected gases at high concentrations

Polymeric blends for humidity



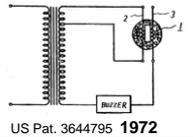
US Patent 2047638, **1936**

Catalysts for combustible gases

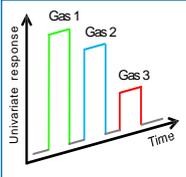


US Patent 3092799, **1963**

Metal oxides for gases



US Pat. 3644795 **1972**



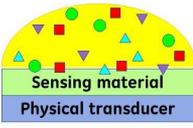
Univariate response
Time

“Simple sensors perform best when pollution levels are high and when the compound of interest swamps others” Lewis, Edwards, *Nature* 2016

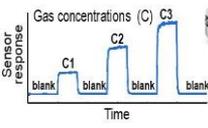
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>> Radislav Potyrailo: This slide shows that the gas sensors from the previous slide come from innovations that have been happening continuously since the 20th century. You can see that gas sensors have been invented in the 1930s, 1960s, and 1970s. They have been very important analytical instruments to measure gases whose presence needs to be avoided in the atmosphere, or when they create explosive or toxic environments. That's why single-output sensors, like the ones shown here, are perfect for those scenarios. Currently, gas sensors have very small form factors and consume less and less power. They are getting into different types of wearable devices and are quite inexpensive. In the middle of the slide, I'm showing that gas sensors often are comprised of a sensing material and a physical transducer, to recognize changes in the sensing material. By design, these sensors are reversible. By definition, a sensor is continuously doing measurements, so the interaction forces in these types of sensing materials are quite weak, in order to release the gas that is being detected. (Continued...)

Conventional gas sensors: Serving sensing needs in established markets



Sensing material
Physical transducer



Gas concentrations (C) C1 C2 C3
Sensor response
Time
blank blank blank blank



- mature technologies
- widely available
- Interchangeable
- inexpensive

alphasense.com
amphenol-sensors.com

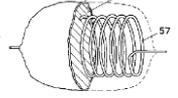
Understanding the roots:
single-output devices for detection of expected gases at high concentrations

Polymeric blends for humidity



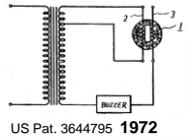
US Patent 2047638, **1936**

Catalysts for combustible gases

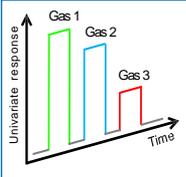


US Patent 3092799. **1963**

Metal oxides for gases



US Pat. 3644795 **1972**

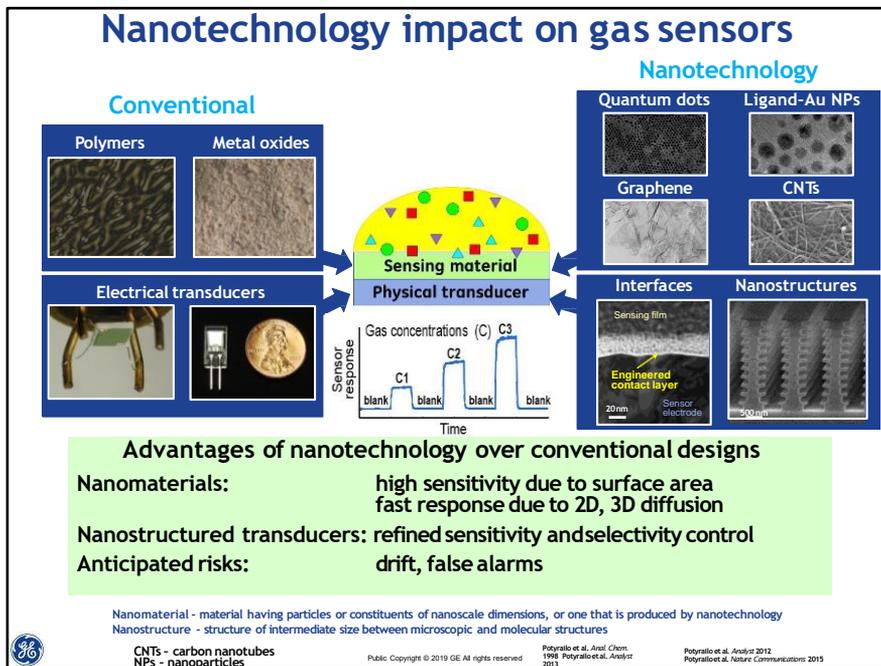


Univariate response
Time

“Simple sensors perform best when pollution levels are high and when the compound of interest swamps others” Lewis, Edwards, *Nature* 2016

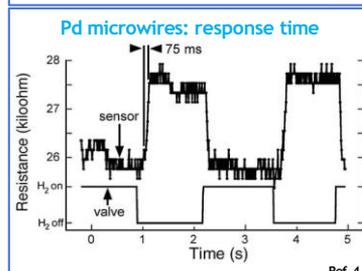
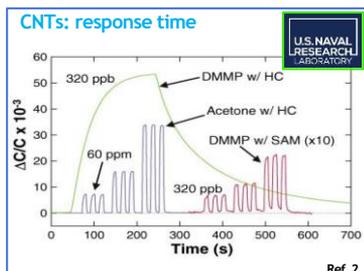
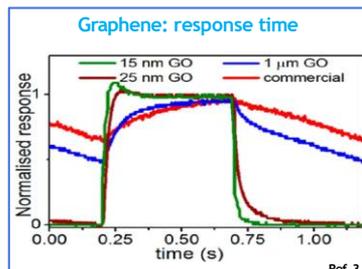
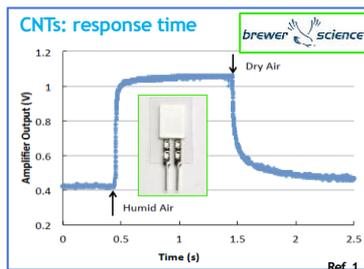
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>> Radislav Potyrailo: On the top of this slide, the second plot from the left shows the response of a sensor in the presence of “blank” (no gas present). You can see that the increase in gas concentration made the signal change more and more proportionately. So schematically, what I'm showing here is that because of the way the sensor was designed, it is okay that it will respond to different gases with slightly or similar intensity, because for those intended applications, there is no other gas that you care about. If you are interested in flammable gases, you know that a flammable gas is the only one that would be present. That's why at the bottom of the slide, I put a quote from a *Nature* article about the fact that these types of sensors are simple by design and perform best when the signal is strong and when nothing else will affect that sensor response. That's a very important aspect of that type of sensors.



>> Radislav Potyrailo: Over the past several years, nanotechnology solutions have been impacting the development of these types of sensors in a very positive way. On this slide, you can see on the left conventional sensing materials (polymers and metal oxide powder) and conventional transducers (you can see a couple of transducer resistive readouts). On the right of the slide are examples of new types of materials and interfaces from nanotechnology. In my personal opinion, the advantages of this nano-approach are quite broad, and they range from improvement in materials to much higher sensitivity, compared to conventional materials (because of their surface area and properties), and much faster response time and recovery time (due to the ability of the gases to have radial or 3D diffusion). If we make different types of transducers that have that improved sensitivity and selectivity, we also need understand design principles for this transducer to behave that way. Anticipated risks are still related to drift and false alarms. From recent literature, we see here impactful examples of these types of technical solutions using nanomaterials.

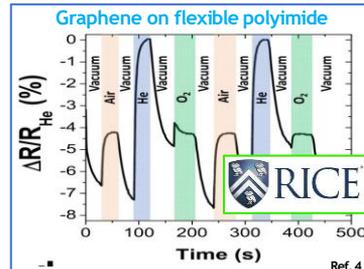
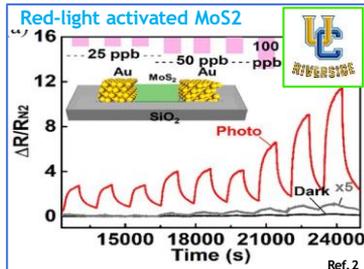
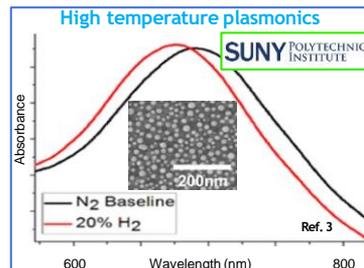
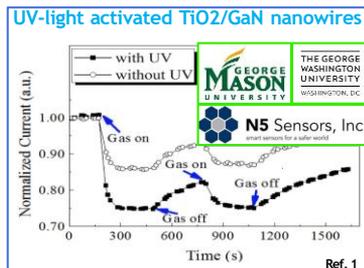
Prominent examples of gas nanosensors:
Fast response time - CNTs, graphene, nanowires



Ref. 1: Kayastha *Sensors EXPO & Conference* 2017
 Ref. 2: Snow et al., *Science* 2005, 307, 1942-1945
 Ref. 3: Borini et al., *ACS Nano* 2013, 7, 11166-11173
 Ref. 4: Favier et al., *Science* 2001, 293, 2227-2231

>> Radislav Potyrailo: This slide shows the response times that have been achieved for sensors based on carbon nanotubes, graphene, and palladium microwires. These response times are very impressive. Brewer Science (plot on the top left) is a company that commercializes gas sensors for community measurements. The Naval Research Laboratory (plot on the bottom left) shows very impressive work, which was published a few years ago in *Science* (on rapid response and selectivity for functionalized carbon nanotubes). You can see other examples from the literature, as well.

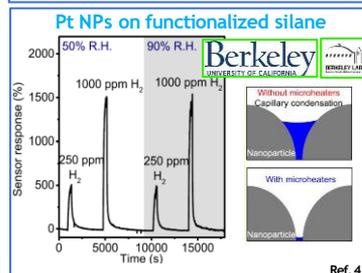
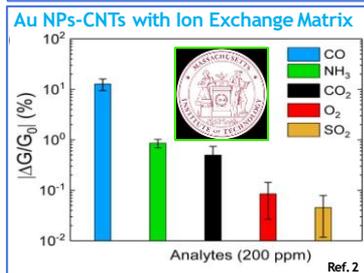
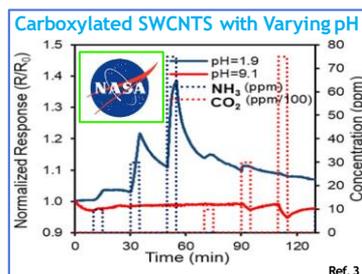
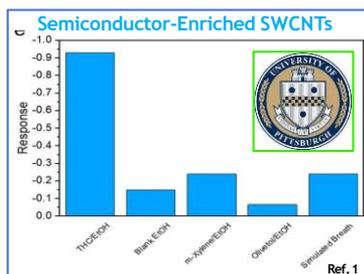
Prominent examples of gas nanosensors:
 Room- and high-temperature operation, light activation, flexible



Ref. 1: Shi et al., *Appl. Phys. Lett.* 2019, 115, 121602
 Ref. 2: Mulchandani & co-workers, *ACS Nano* 2019, 13, 3196–3205
 Ref. 3: Houllihan et al., *ACS Sens.* 2018, 3, 2684–2692
 Ref. 4: Stanford et al., *ACS Nano* 2019, 13, 3474–3482 slide 13

>> Radislav Potyrailo: Our next examples from the literature are about other types of sensors with nanomaterials at room temperature and high temperature and also light activation (plot on the bottom left), to improve the response recovery times of the nanomaterials and also for wearable devices and other applications. Technical solutions on flexible substrates (plot on the bottom right) is an important aspect of that development, as well. The example of new UV light-activated nanowires (plot on the top left) is something that I'm very happy to share with you. It comes from George Mason University, George Washington University, and N5 Sensors, Inc. Also, as you can see, most of these examples are from this year or from last year.

Prominent examples of gas nanosensors:
 Functionalization, elimination of humidity effects for selectivity



Ref. 1: Star & co-workers, *ACS Sens.* 2019, 4, 2084-2093
 Ref. 2: Swager & co-workers, *Chem. Mater.* 2019, 31, 5413-5420
 Ref. 3: Meyyappan & co-workers, *ACS Appl. Nano Mater.* 2019, DOI: 10.1021/acsnam.9b01401
 Ref. 4: Javey & co-workers, *ACS Sens.* 2019, 4, 1857-1863
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>> Radislav Potyrailo: My last example on nanomaterials is related to the functionalization of different types of nanomaterials – mostly carbon nanotubes, as you can see, and also different types of nanoparticles. Why? Because we need to get a more selective response and also to reject the humidity effect. Because humidity is present in ambient air at very high levels, at 50% humidity, there is 15,000 parts per million of water vapor. At much higher humidity, that number increases to up to 20,000 parts per million. So, the ability to reject such levels of humidity is a very important aspect of practical sensors.

Requirements and opportunities for modern sensors

**Top 10
General
sensor
requirements**

- Dynamic range
- Initial cost
- Long-term stability
- Operation cost
- Power consumption
- Response speed
- Robustness
- Selectivity
- Sensitivity
- Size

**Top 3
Focused
sensor
requirements**

- High reliability,
accuracy
- Low cost
- Low power

Markets for Sensors in Industrial Internet, 2014
Potyrallo, IDTech Internet of Things 2014
Potyrallo, TSensors Summit 2015

Potyrallo, Mirsky, Chem. Rev. 2008
Potyrallo et al., Chem. Rev. 2011
Potyrallo, Chem. Rev. 2016

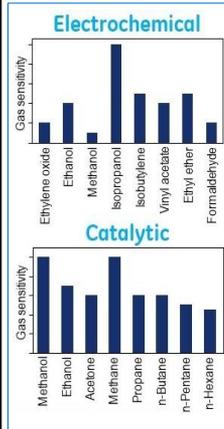
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>> Radislav Potyrallo: What we hear from end users is that while there are many technical requirements for sensors (you can see a list of those requirements in alphabetical order on the left of this slide), basically, the technical requirements boil down to the top three, which are driven by those different markets that I showed on slide 2. Those top three requirements are related to how accurate a sensor is (reliability is related to accuracy), low cost to use it in ubiquitous applications, and low power for different types of wearables and mobile solutions. These are the requirements that are coming from end user and system integrators.

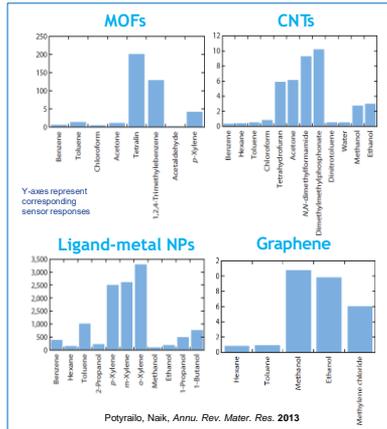
A gas sensor is a type of analytical instrument that was designed in the 20th century to have a single output. Why? Because you don't need more outputs, you need to detect only one gas (and you know what it is). That's why these days, if we're trying to use these types of sensors in applications that there were not intended for, then we need to pay attention to the fact that they will respond to other gases, as well.

Gas cross-sensitivity of sensors: what does it mean?

Conventional

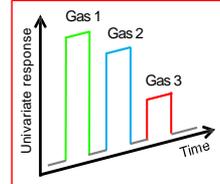


Nanomaterials



MOFs - metal organic frameworks
CNTs - carbon nanotubes
NPs - nanoparticles

Common theme for conventional and nanomaterials



Gas cross-sensitivity in conventional and nanomaterials due to conflicting requirements for sensor selectivity vs reversibility



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>> Radislav Potyrailo: When we buy conventional sensors (left side of the slide) from different manufacturers, they have tables or bar graphs (or other type of summaries) that say “I am calibrated for this gas or that gas, but I will respond to many other gases, so please pay attention to that fact.” Why? Because, as we were mentioning, the response from these sensors needs to be reversible, and therefore the interaction forces between the sensing material and the gas should be weak enough to release the gas, once the concentration gets smaller and smaller. So that's why these nonselective responses are happening, because of the way these sensors are designed.

For gas sensors made with nanomaterials, here are examples from the literature (right side of the slide) showing that different types of nanomaterials have these types of responses to different gases, or vapors. (I call them “volatiles,” in general.)

My schematic (“Common theme for conventional and nanomaterials”) shows that, indeed, both types of sensors will respond to different gases.

Exploring new applications with traditional gas sensors

“Simple sensors perform best when pollution levels are high and when the compound of interest swamps others” Lewis, Edwards, *Nature* 2016



<https://www.treehugger.com/clean-technology/air-quality-sensor-makes-a-fashion-statement.html>



<https://techcrunch.com/2017/01/03/plume-labs-flow-is-an-air-quality-tracker-to-avoid-pollution/>

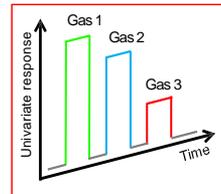


<https://plumelabs.com/en/>



<https://www.itri.org.tw/eng/>

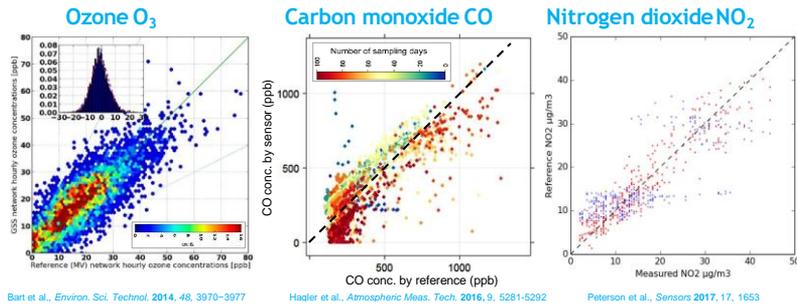
Common theme for conventional and nanomaterials



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>> Radislav Potyrailo: So, if single-output sensors respond with different types of designs, then their nonselective responses will be observed, regardless of whether a big battery is needed to carry it or whether it is miniaturized. As you can see on this slide, single-output sensors can have variable formats. This challenge of not recognizing different types of gases still remains, due to the design of these types of sensors.

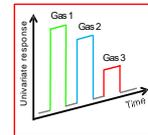
Gas cross-sensitivity of sensors: what does it mean?



The biggest headaches are caused by interfering chemicals
 Lewis, Edwards, *Nature* 2016

Air monitoring is more involved than manufacturers of cheap sensors suggest
 Austen, *Nature* 2015

Common theme for conventional and nanomaterials



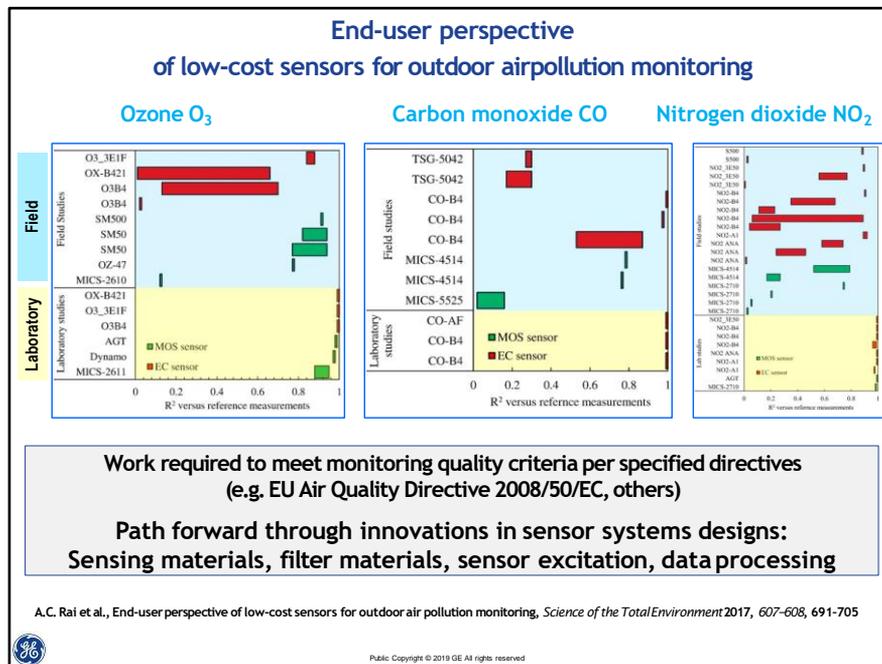
For gas sensors revolution to take off, accuracy must improve



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>> Radislav Potyrailo: For end-use applications, people are really interested to see how accurately, or how reliably, they can detect pollutants in ambient air and whether they can use different types of sensors for those reliable measurements. Ozone, carbon monoxide, and nitrogen dioxide are examples of pollutants in ambient outdoor air. You can see here the correlation between the reference instrument and the sensors, which were recently reported in the literature. You can see that if these data points line up perfectly on the line with the slope of one, then that will be a perfect correlation. If they don't line up as well, then it will be a less perfect correlation. So depending on the end-use scenario, some of the correlations, even if they're not perfect, may or may not be allowed. It really depends on the end-use scenario.

You can see from these two quotes from *Nature* that the end users are saying that interfering chemicals are really adding to uncertainties in our measurements, something that one of them calls the “biggest headaches.” What it means for us is that we need to pay attention to how we deal with this performance in the field.



>> Radislav Potyrailo: This slide shows a summary from European work, where researchers were doing measurements of ozone, carbon monoxide, and nitrogen dioxide. But the researchers were doing calibrations in the lab (yellow regions) and in the field (blue regions). In the lab, when they were calibrating, then the linear fit quality (r^2) was always almost one: $r^2 = 1$ is the best quality. You see that no matter what the gas was, the quality of that correlation in the lab was perfect. When those researchers were in the field, then the challenges started to show up very clearly. The correlation dropped quite significantly, and depending on the type of sensor, some of them were more affected than others. What this means is that that we, as a community of sensor developers, need to think of how to improve this type of sensor performance so that it is as close to the required correlation as appropriate.

Inspiration from physiological wearable sensors

For The Wearable Revolution To Take Off, Accuracy Must Improve

CVC, *Inside Activity Tracking*, 2013



<http://www.insideactivitytracking.com/tracking-accurately-how-it-d-one-why-it-difficult/>

Fitness Wearables Lack Accuracy

C.Van Hoof, *EE Times*, 2014



http://www.eetimes.com/author.asp?section_id=365&doc_id=1321978

More accurate readings without the goop

Y. Zhu, *NC State Engineering FALL/WINTER 2015*



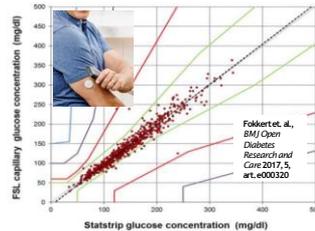
<https://news.engr.ncsu.edu/2015/10/more-accurate-readings-without-the-goop/>

Apple Watch 4 Is Now An FDA Class 2 Medical Device: Detects Falls, Irregular Heart Rhythm

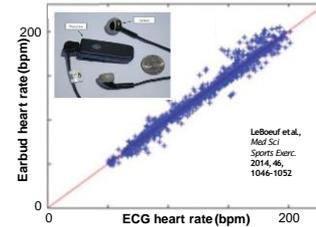


<http://www.fda.gov/oc/2018/08/2018-08-20-apple-watch-4-is-now-an-fda-class-2-medical-device-detects-falls-irregular-heart-rhythm-082120180211>

FreeStyle Libre Flash



Earbud HR VSECG



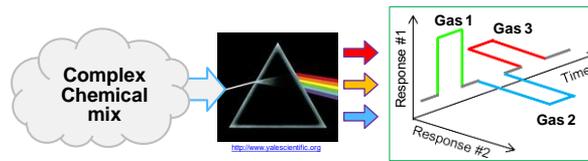
The wearable revolution did take off once accuracy has been improved



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>> Radislav Potyrailo: Many inspirations come from adjacent fields. For researchers developing different types of sensors, part of that inspiration has come from physiological wearable sensors. Several years ago, when the first wearable sensors were introduced, there was also a fashion statement. But then, people started to confess (in their reports) that the expected “revolution” from these wearable sensors couldn’t take off because their accuracy wasn’t good enough. This means that the accuracy, or the correlation plot between the reference measurements and the sensor measurements, was “sprayed.” So, researchers showed that if the accuracy, or the performance, of these sensors needed to become as good as hospital equipment or FDA-approved equipment. Then, they developed wearable sensors with nice correlation plots, and more and more of those wearables were produced. That’s when the “revolution” took off because the accuracy improved significantly, and the end users were satisfied.

Traditional analytical instruments: Eliminated gas cross-sensitivity problem by design



Systems with multiple analytical detectors



<https://danvillesanramon.com>



<http://ms-me.com>



<https://uk-air.defra.gov.uk>

Gas chromatography Mass spectrometry Laser spectroscopy



Qin, Gianchandani, *Microsyst. Nanoeing.* 2016



May 28, 2018



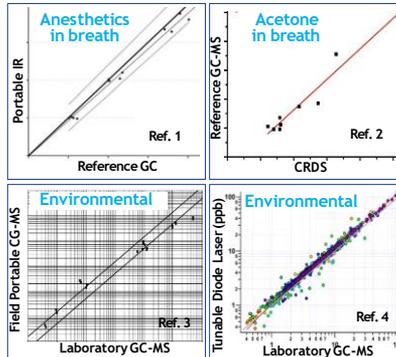
<https://alliedscienceflcpro.com>

Excellent progress in miniaturization, accuracy

- Ref. 1: Hendrickx et al., *BMC Anesthesiol.* 2008, 8, 2
- Ref. 2: Sun et al., *Rev. Sci. Instrum.* 2015, 86, 095003
- Ref. 3: EPA *Environm. Techn. Verification Report* 1997
- Ref. 4: Lerner et al., *Atmos. Meas. Tech.*, 2017, 10, 291-313



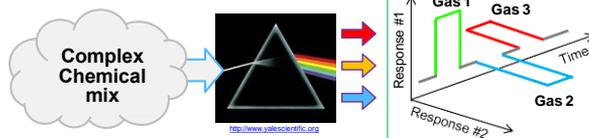
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CRDS - cavity ringdown spectroscopy
GC-MS - chromatography-mass

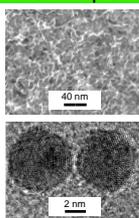
>> Radislav Potyrailo: This slide shows that by design, there are also other types of analytical instruments that have more than one output (on purpose). The reason for that is that when these instruments are used for analysis of complex mixtures in gas phase, you would like to detect and quantify multiple components in that mixture. Then, by design, you have more than one output. On the left side of the slide are examples of different instruments. They can be big, but many of them are getting smaller and smaller. Some examples are gas chromatography (where the independent variable is retention time), mass spectrometry (where the independent variable is the mass-to-charge ratio), or laser spectroscopy (where the independent variable is the wavelength of light). On the right side of the slide are correlation plots, which are quite impressive to me. Why? Because they are much tighter, which helps to line up those data points along this slope of one. So that's a very impressive progress in miniaturization and keeping the accuracy quite high, even if you use a small instrument.

2019 advances in micro-gas chromatography (GC):
Eliminated gas cross-sensitivity problem by design

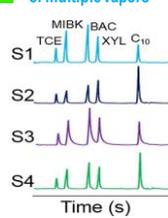


Zellers et al., Anal. Chem. 2019, 91, 4747-4754

NEW GC detector based on nanoparticles



Resolution of multiple vapors



omniscent.com/wp-content/uploads/2019/05/Solution-Brief-2100-v2.pdf

2019 achievements in miniaturization and power reduction



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>> Radislav Potyrailo: This slide is one of my favorites. This is an example of a wearable, portable, belt-attached gas chromatography system. This very impressive work is from the University of Michigan. You can see that the detector is placed on gold nanoparticles to detect different types of volatiles (Gas 1, Gas 2, and Gas 3, top right). The sensor can be in a form factor where it can be a part of a wireless network, and you resolve the different types of volatiles in a very nice way from a complex mixture. That's where we currently are in gas chromatography, and that's inspiring for the whole community of analytical science and sensors community.

Top three learnings from Segment 1

Segment 1:

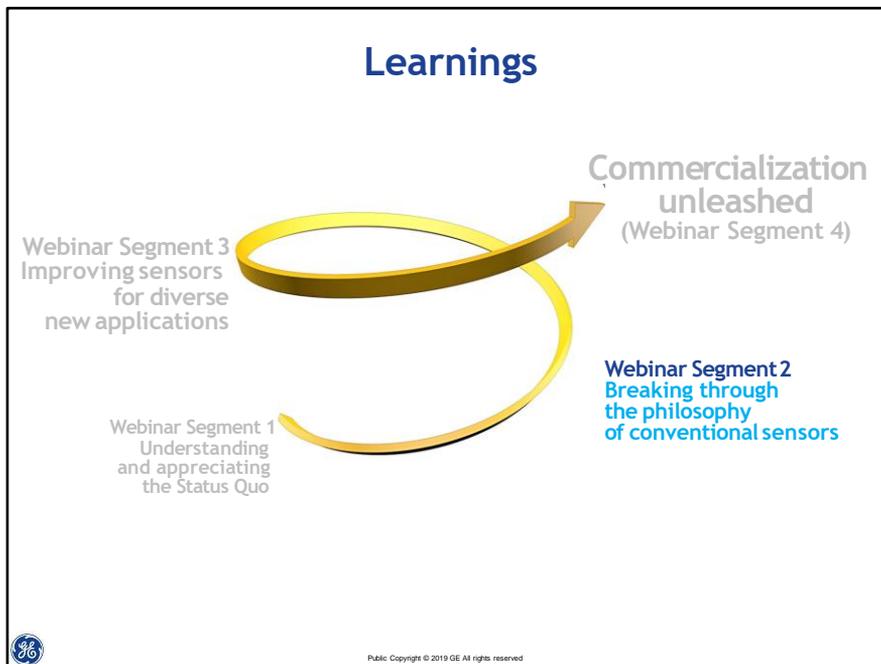
Understanding and appreciating the Status Quo

1. Gas sensors were invented as single-output devices to detect expected gases at relatively high concentrations
2. Sensing nanomaterials boost sensor sensitivity and enhance response speed
3. Mature analytical instruments reject interferences by their design principles; some contemporary fieldable deployments utilize nanomaterial-based detectors

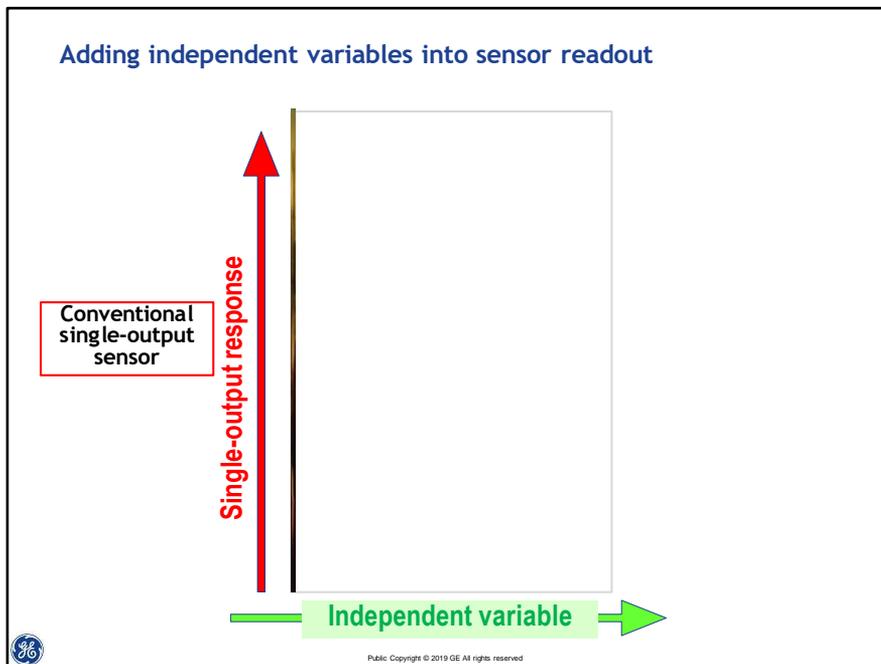


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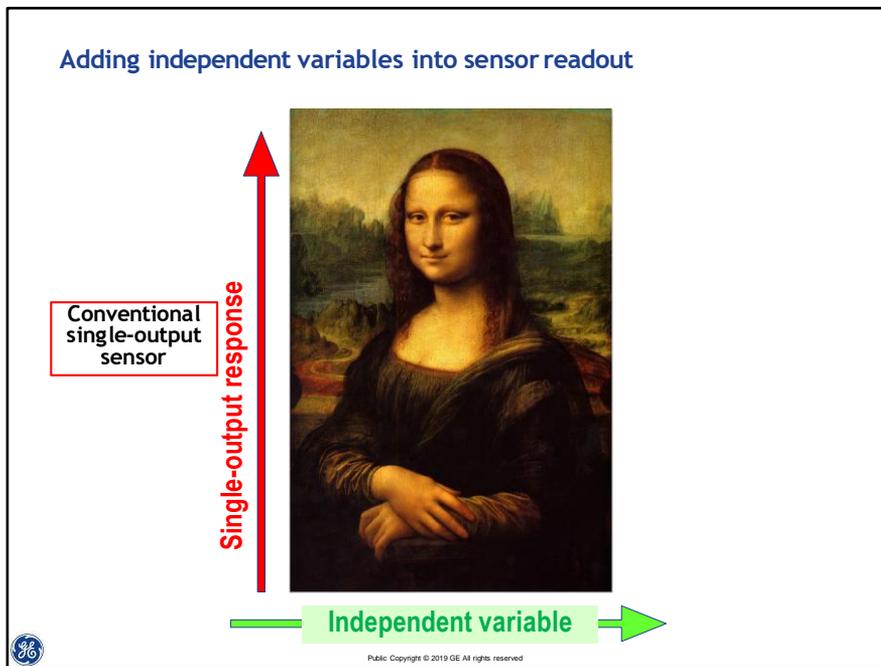
>> Radislav Potyrailo: This is what we learned from Segment 1: First, we understand that gas sensors were, by design, invented as single-output devices (they detect expected gases). Second, we also realized, from examples that I showed you, that sensing nanomaterials improve sensor sensitivity and improve response speed. Third, we recognized that there are other types of analytical instruments that are based on design principle, where you have more than one output. Those were examples that I showed you on the last two slides; I also showed you an example of a sensor from this year, indicating that sensors are getting smaller and smaller, and that's a very encouraging direction.



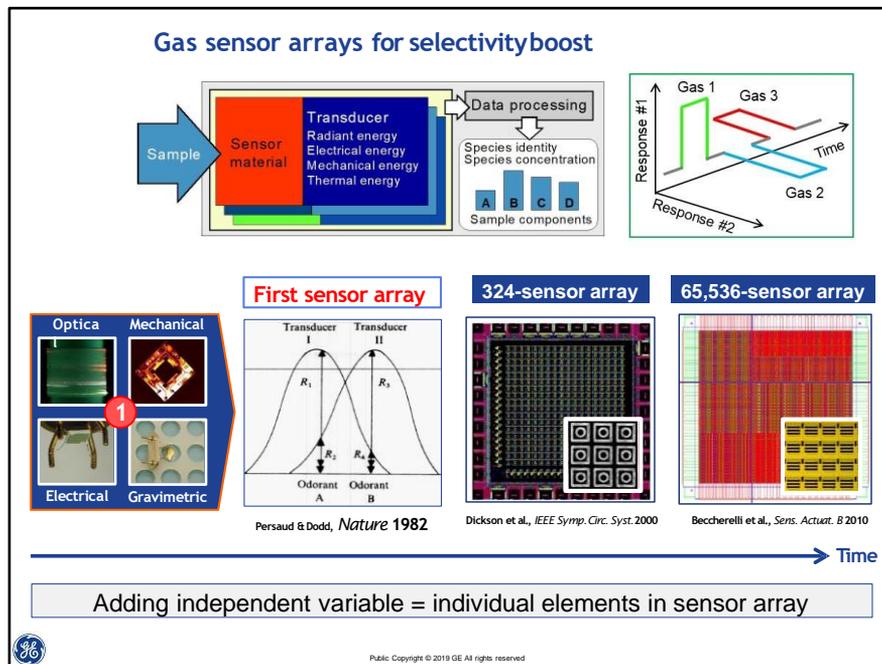
[There is no text for this slide].



>> Radislav Potyrailo: Now, let's look at how can we improve the state of the art of existing sensors. We need to recap that we have individual sensors that have single output, and if we add independent variables, then we may be able to get more information from the sensor designs.



>> Radislav Potyrailo: If I stretch this line slightly this way, then it opens up a new dimension – literally a new x-axis. Now, you can see what you have been missing!

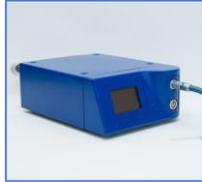


>> Radislav Potyrailo: Adding this independent variable, or dimension, is an approach that was introduced by Krishna Persaud in 1982. This approach consists of adding different types of sensing elements, and they are indeed giving you this independent variable. These sensing elements are individual elements in the sensor array. Then, you can do your data analytics and get the response that, schematically, is the same as from more sophisticated instruments that we discussed in Segment 1.

From examples from the literature, you can see sensor arrays with 65,000 elements. We are part of the sensor array community. We have been doing a lot of work in sensor arrays. A synonym for that is “electronic nose,” because when you are doing data processing and are mimicking an electronic nose’s performance.

Examples of modern gas sensor arrays: 2019

Metal oxides



<https://airsense.com/en/products/portable-electronic-nose>

Nanofiber



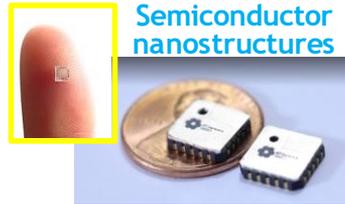
<http://www.vaporsens.com/products/>

Metal oxides



<https://dooclip.me/UN6sncfybic.html>
OMNI SENTIR ceo@sid4d.com

Semiconductor nanostructures



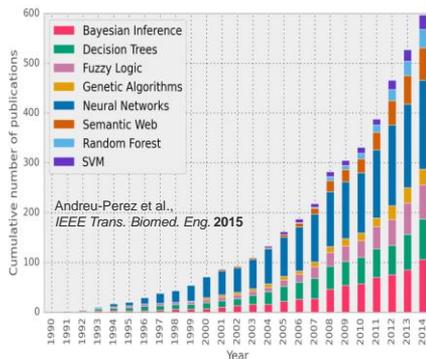
<http://n5sensors.com/>



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>> Radislav Potyrailo: Here are a few examples of modern gas sensor arrays (which I took from the internet) from different companies.

Tools for data analysis of multivariable sensors: Machine learning (ML), data analytics, multivariate statistics



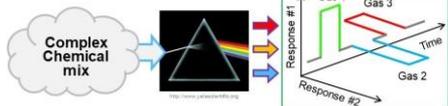
Examples of GE ML tools

- Support Vector Machines (SVM)
- Principal component analysis (PCA)
- Hierarchical cluster analysis (HCA)
- Discriminant Analysis (DA)
- Artificial Neural Network (ANN)
- Independent Component Analysis (ICA)
- Partial least squares (PLS) regression
- Principal Component Regression (PCR)

• New tools for boosting sensor stability

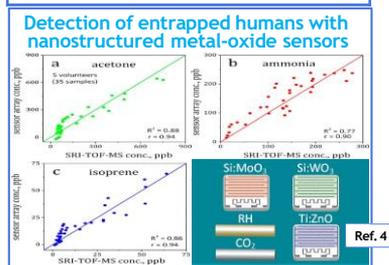
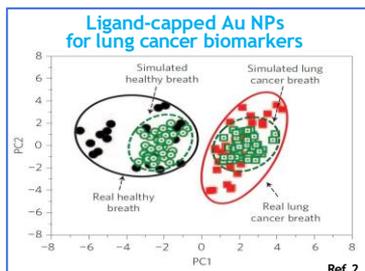
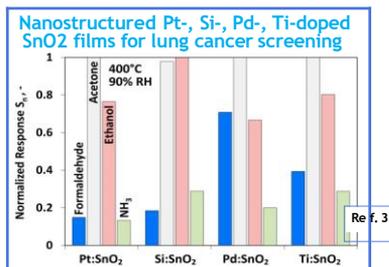
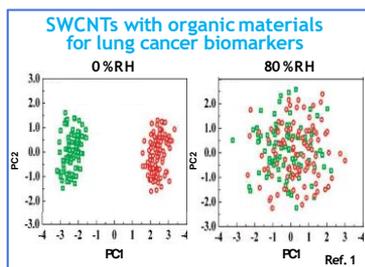
R. A. Potyrailo, Multivariable sensors for ubiquitous monitoring of gases in the era of Internet of Things and IndustrialInternet, *Chem. Rev.* 2016, 116, 11877–11923

Importance of response dispersion in a sensor array for reliable performance



>> Radislav Potyrailo: Data analysis is a very important aspect of getting that information from individual elements. On the left of the slide, you can see a very nice graph from the literature that summarizes different types of analytical tools that are being used. There are synonyms for these analytical tools: chemometrics, data analytics, multivariate statistics, and machine learning (the most recent synonym). On the right of the slide, I show an almost complete list of the types of tools that we are using. We are developing new tools, as well. The bottom line here (bottom of the slide) is that to have this dimensionality as big as appropriate, or as big as you can, in order to get that selectivity line, you need response dispersion in a sensor array.

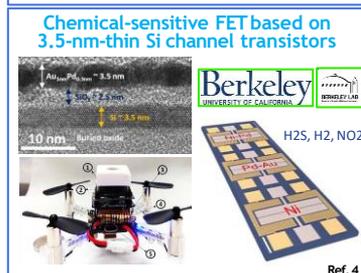
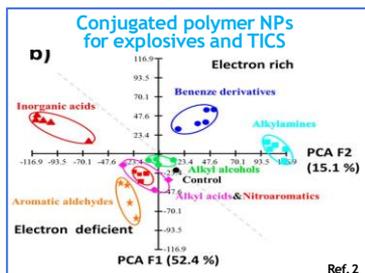
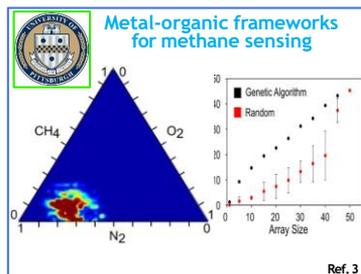
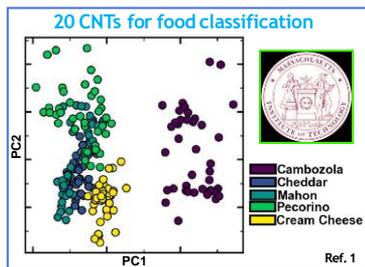
Prominent examples of sensor arrays:
Breath analysis, humans detection



Ref. 1: Haick & co-workers, *Nano Lett.* 2008, 8, 3631-3635
 Ref. 2: Haick & co-workers, *Nature Nanotechnol.* 2009, 4, 669-673
 Ref. 3: Güntner et al., *ACS Sens.* 2016, 1, 528-535
 Ref. 4: Güntner et al., *Anal. Chem.* 2018, 90, 4940-4945

>> Radislav Potyrailo: From the literature, you can see very impressive examples of analyses of volatiles in complex mixtures and in the presence of high levels of humidity with carbon nanotubes, ligand-functionalized gold nanoparticles, and nano-structured materials of different kinds. These are very impressive recent results.

Prominent examples of sensor arrays:
Room temperature applications for low power



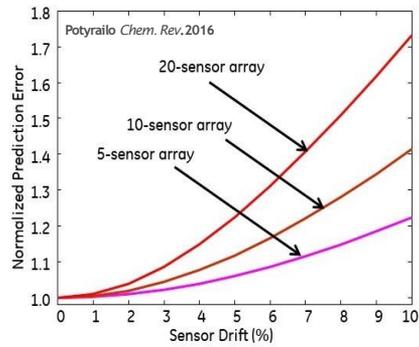
Ref. 1: Swager & co-workers, *ACS Sens.* 2019, 4, 2101–2108
 Ref. 2: Zhao et al., *Anal. Chem.* 2018, 90, 4815–4822

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Ref. 3: Gustafson and Wilmer, *ACS Sens.* 2019, 4, 1586–1593
 Ref. 4: Javey & co-workers, *Sci. Adv.* 2017, 3, e1602557

>> Radislav Potyrailo: Here, I am showing examples from recent results with carbon nanotubes, conjugated polymer nanoparticles, metal-organic frameworks, and other types of structures that are showing progress in this area, which is quite significant.

Long term instabilities can increase prediction error of sensor array



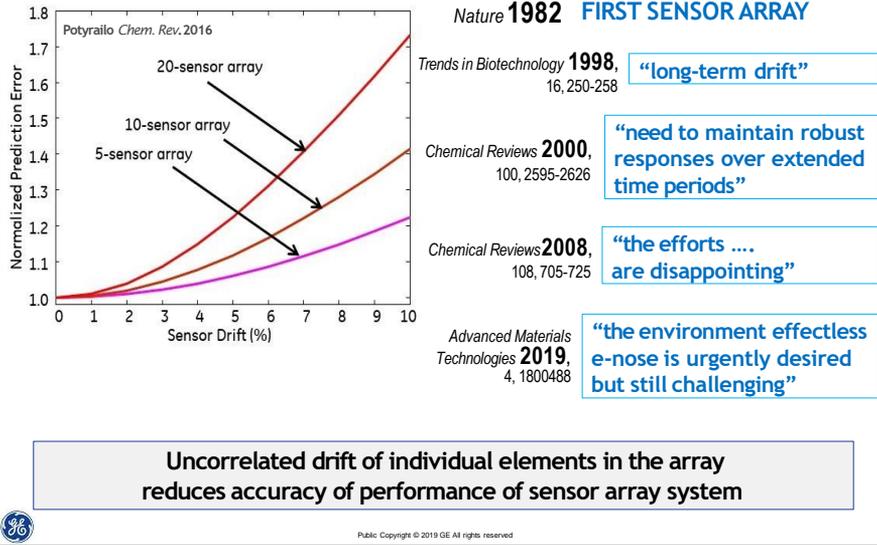
Uncorrelated drift of individual elements in the array
reduces accuracy of performance of sensor array system



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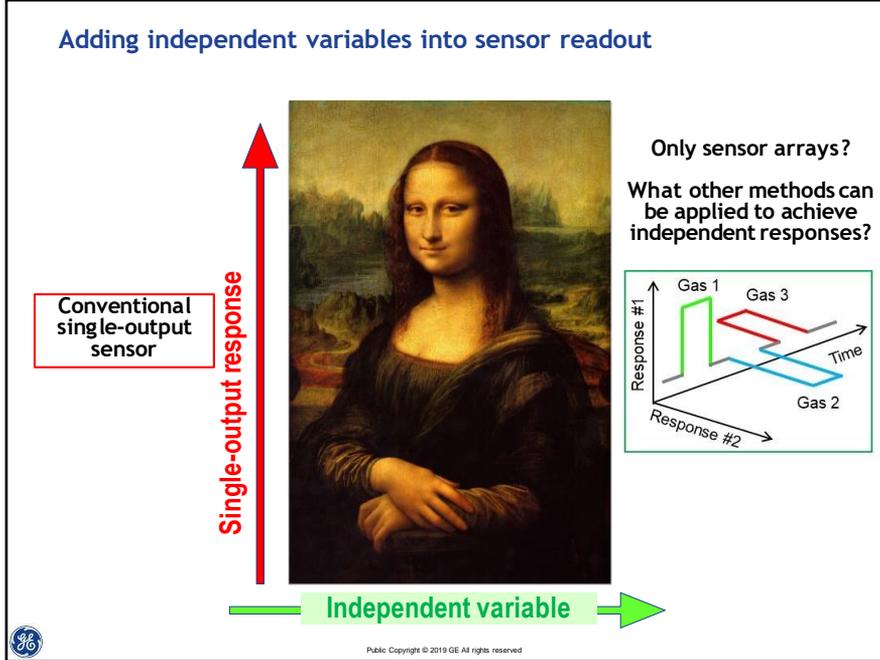
>> Radislav Potyrailo: As sensors were developed in the 1930s as individual sensors, and as sensor arrays were introduced in 1982, then, little by little, we, as a community, started to learn that these individual sensing elements gracefully age, independently from each other, because, by design, they are slightly different. That's why, over time, we lose the prediction accuracy using a sensor array. And we, as the community, are working hard on this challenging problem.

**Long term instabilities
can increase prediction error of sensor array**

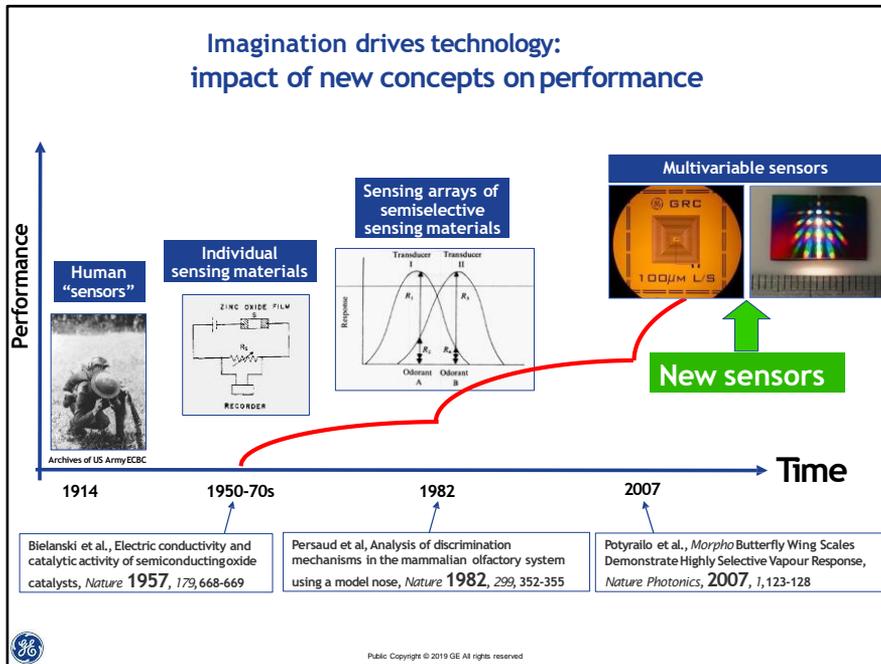


>> Radislav Potyrallo: This slide recaps that since the development of the first sensor array in 1982, we started to work on removing the effects of long-term drift. You can see quotes from different review articles from 1998 to 2019. All of this is work in progress, and we need to refocus, perhaps, and use less explored methods in reducing the drift. The reason I'm saying "less explored" is because, we, as a community, have explored these analytical tools quite well. So, my personal opinion (and that of others) is that we need to think about this situation even further.

Adding independent variables into sensor readout



>> Radislav Potyrailo: If I look at slide 34 again (Mona Lisa) (and I'm paying attention to the x-axis because it says "independent variable"), if I move a little bit outside of my comfort zone – such as individual elements in a sensor array – then I will think, "What other types of changes can I induce?" That's that punch line. What we see is that since the development of the first sensors and sensor arrays, perhaps we need to switch to new concepts in getting that data out (or data acquisition) through excitation of the sensors, in order to get more stability.



>> Radislav Potyrailo: This slide summarizes the timeline. You will note that before sensors were invented, people were doing measurements with humans, mammals, and birds. So we are advancing this knowledge.

Top three learnings from Segment 2

Segment 2:

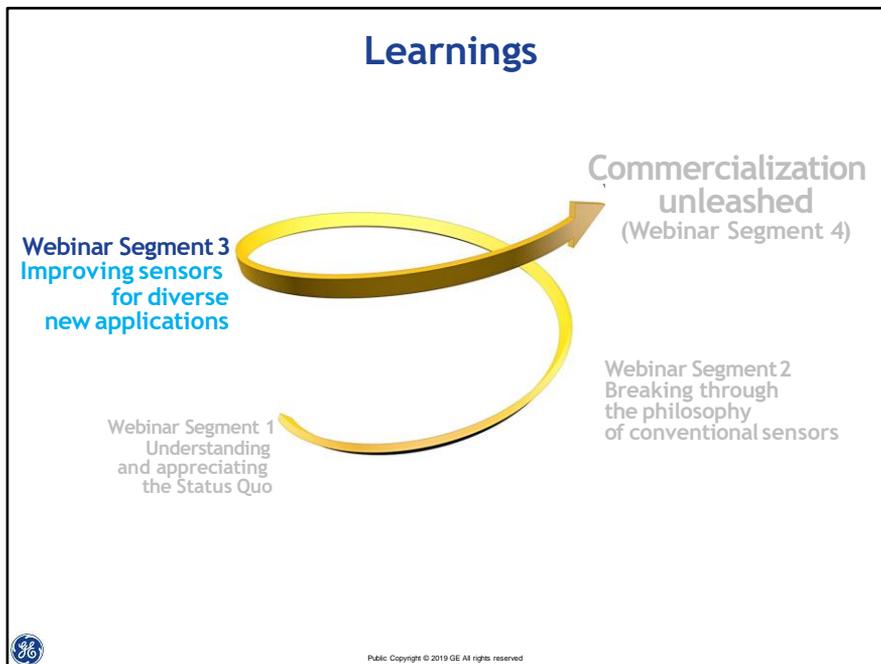
Breaking through the philosophy of conventional sensors

1. Sensor arrays have one independent variable such as elements in the array
2. Sensing nanomaterials have tunable selectivity for determinations of different gases
3. Users request to improve stability of sensor arrays for their broad acceptance

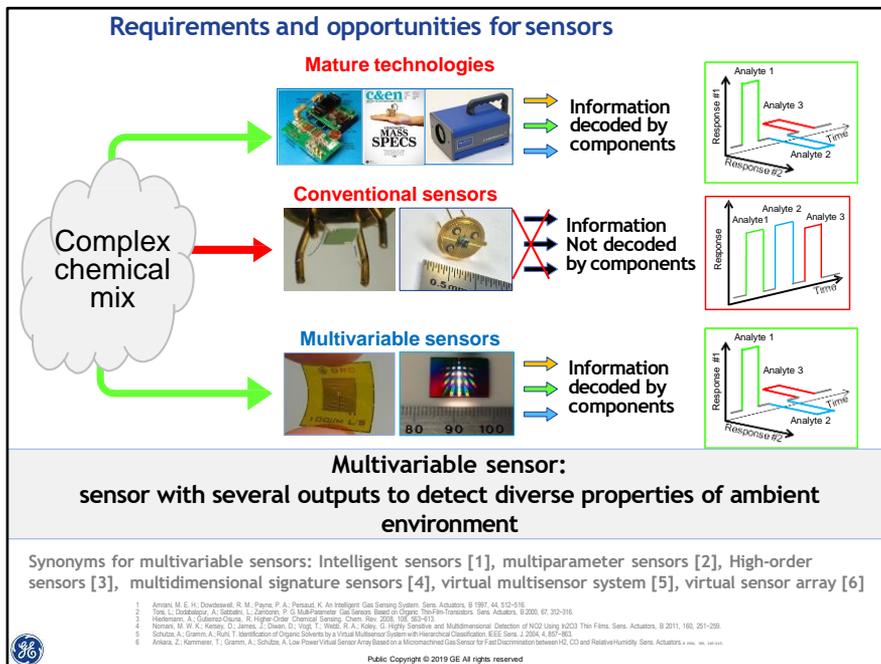


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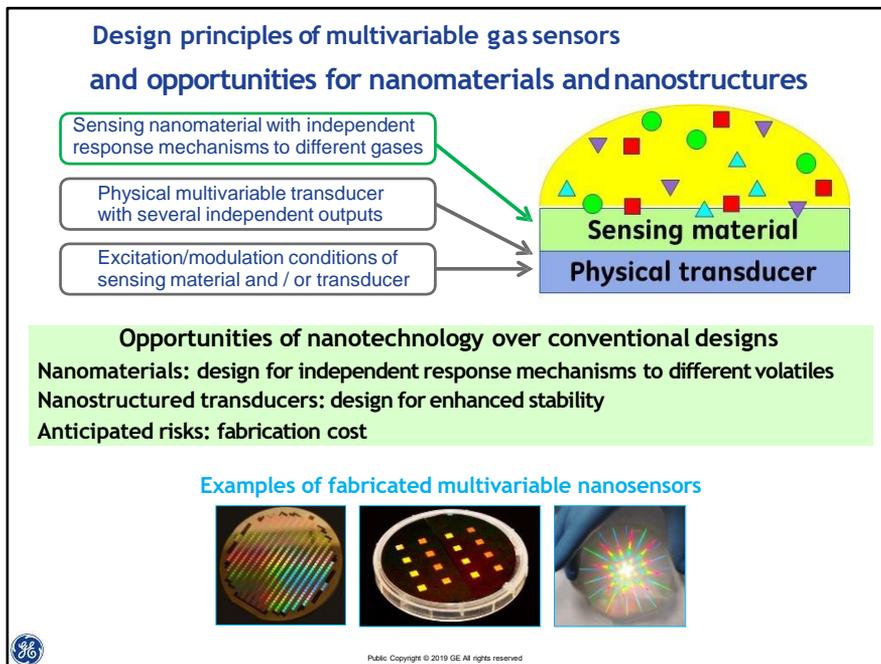
>> Radislav Potyrailo: Segment 2 is showing the following: First, sensor arrays have one independent variable, such as elements in the array, which really helps in discriminating between different types of volatiles and rejecting interferences; second, sensing nanomaterials have a lot of useful additions to the capabilities of the sensor arrays; and third, what we saw from those quotes from different review articles is that users request stability improvements. So, we, as a sensor community, are focusing on that.



>> Radislav Potyrailo: Segment 3 is: How can we improve this situation? We recognize the capabilities of mature analytical technologies, which we discussed in Segment 1. Also, we recognize how arrays and individual sensors operate.



>> Radislav Potyrailo: Now, we are able to develop single sensors but with multiple outputs, also called multivariable sensors. These are sensors with several outputs to detect diverse properties of ambient environment. Different synonyms have been developed over the years for these types of sensors, and I am highlighting them on this slide, as well (bottom of the slide).



>> Radislav Potyrailo: I would like to emphasize that there is nothing magical about this sensor design. We need to learn, or need to apply our learning, on how to develop these sensors to correct remaining things that need to be corrected before they would be accepted by users. First, nanomaterials should have independent response mechanisms to different gases. That's relatively easy to do, and that will help us big time. Then, our physical transducer should be excited or measured in such a way as to detect these changes in the materials. Also, we should pay more attention, perhaps, to how we will excite and modulate our physical transducer. Overall, that will give us the opportunity to improve the performance of our sensors. Here, I'm highlighting opportunities of nanomaterials and nanostructures over conventional designs to solve remaining challenges, as well.

Vapor-response mechanisms: conventional and nanomaterials

Dielectric polymers:

dispersion, polarizability, dipolarity, basicity, acidity, and hydrogen bonding

Conjugated polymers:

density and charge carrier mobility, swelling, conformation transitions of chains

Carbon nanoparticle-filled polymers:

Swelling

Fullerenes:

dispersion, dipolarity/polarizability, and hydrogen-bond acidity

Carbon nanotubes & graphene:

charge transfer and polarization of molecular adsorbates, gas-induced Schottky barrier modulation at contacts

Semiconducting metal oxides:

adsorption/desorption, reduction/reoxidation, bulk effects

Monolayer-protected metal nanoparticles:

electron tunneling between metal cores and chargehopping along the atoms of ligand shell

Zeolites:

molecular discrimination by size and shape

Supramolecular materials:

molecular recognition based on complexation interactions of gases in due to the presence of organic hosts with enforced cavities

Metalloporphyrins and related macrocycles:

hydrogen bonding, polarization, polarity interactions, metal center coordination interactions and molecular arrangements

Metal-organic frameworks:

van der Waals interactions of the framework surface, coordination to the central metal ion, and hydrogen bonding of the framework surface

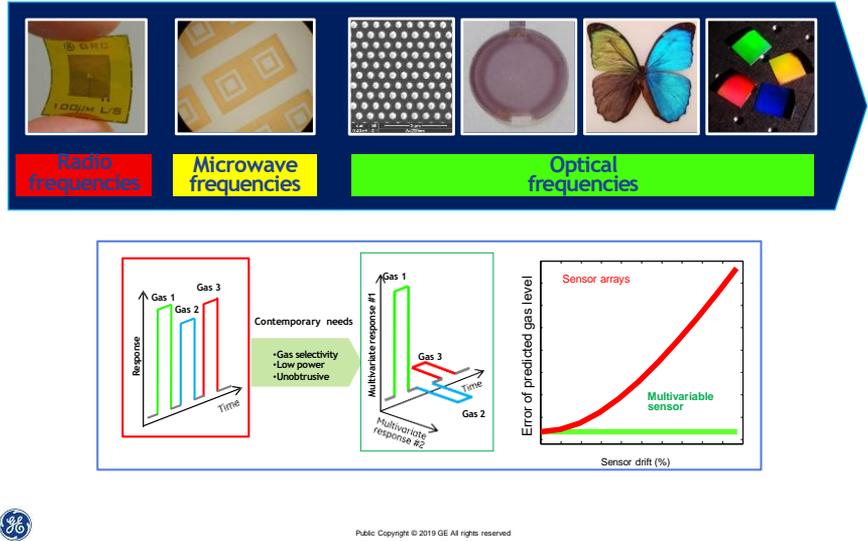
Potyrailo et al., *Chem. Rev.* 2011
Potyrailo, Naik, *Annu. Rev. Mater. Res.* 2013



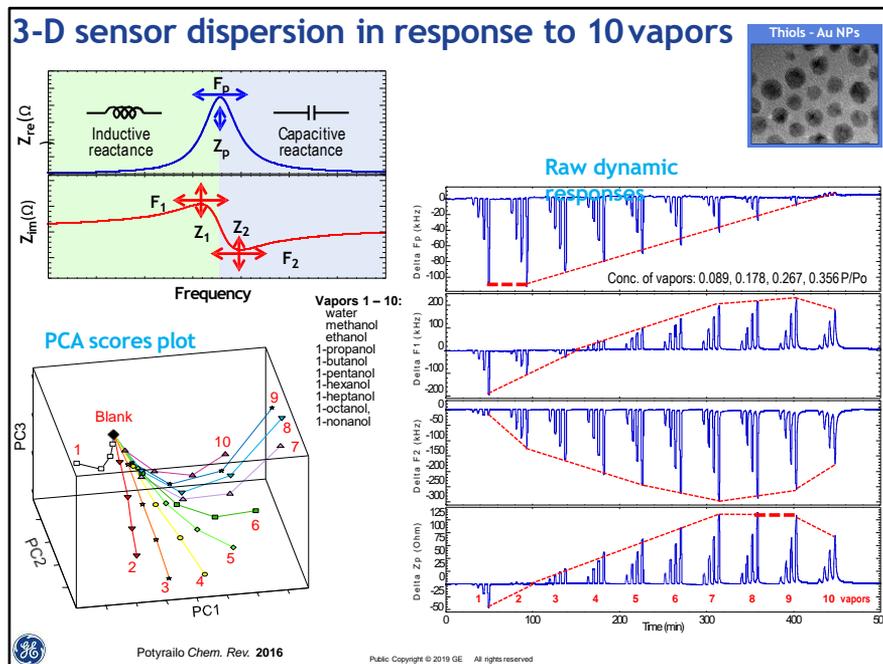
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>> Radislav Potyrailo: The reason I was saying it is relatively easy to think about these different response mechanisms is because we already have a lot of sensing materials that we have been using for some time – conventional ones and nanomaterials. Now, with our new learning, we are seeing what types of response mechanisms sensing materials may have and how to take advantage of these diverse response mechanisms. Then, once we know what to expect, we can have a transducer that will pick up those different responses.

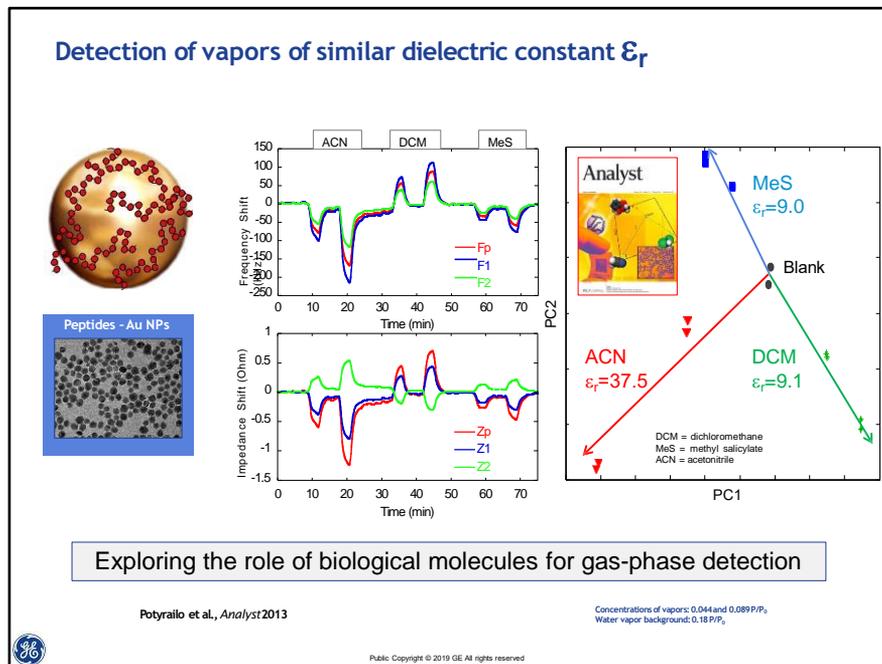
Electromagnetic multivariable nanosensors: New philosophy for selective gas sensing



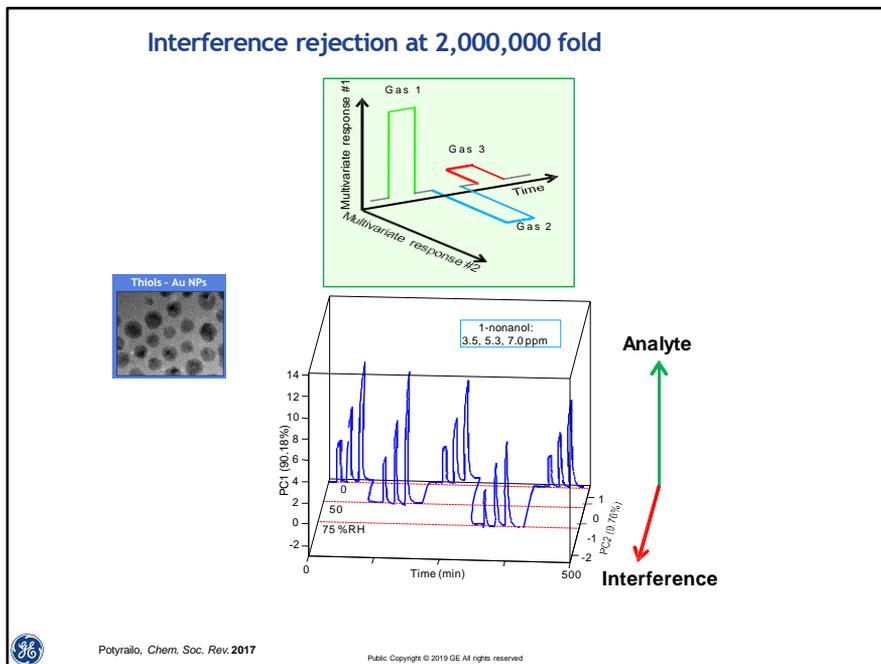
>> Radislav Potyrailo: As an example, on this slide, I'm showing that we can visualize the different types of multivariable sensors on the electromagnetic spectrum. We can have sensors that are operating at radio frequencies (resonant impedance sensors or normal impedance sensors), microwave frequencies, and optical frequencies (different types of resonant and non-resonant optical structures). The goal is the same as for sensor arrays: to put the responses of different gases into different directions after data analysis. Also, what we do more and more is to have our individual sensors as stable (as fresh) as possible when they are slightly aged and when there is a significant sensor drift (which we are inducing on purpose in our labs).



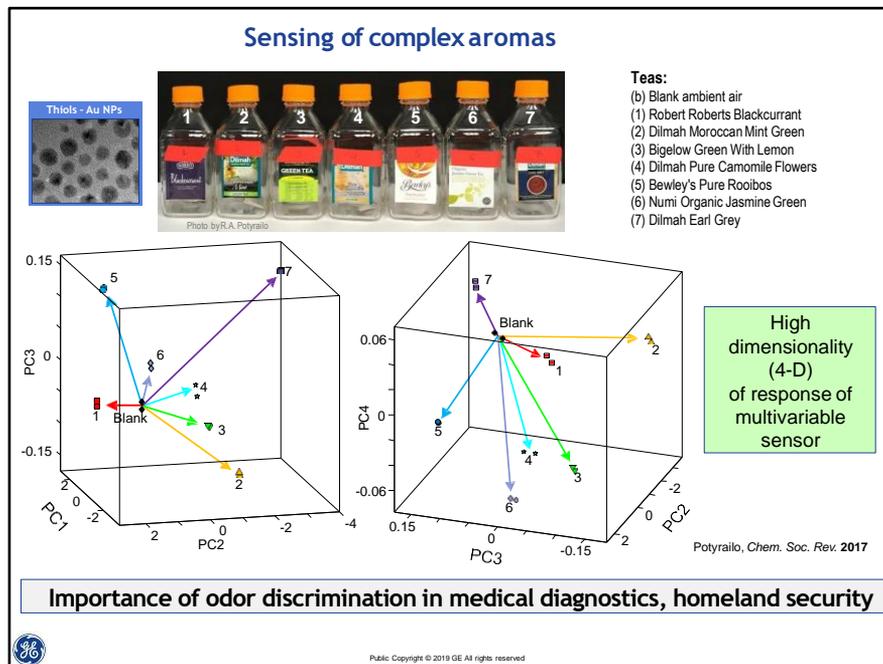
>> Radislav Potyrailo: As an example, here, I am showing that we can have three-dimensional sensor responses after multivariate analysis when we are looking at the resonant properties of a sensor that is coated with gold nanoparticles and when we are exposing this type of sensor to different types of volatiles (here, alcohols from the same family of linear alcohols, and water vapor as interference). You can see that when we are exposing the sensor to full concentrations of each of these volatiles, the response of the peak shift is having the profile shown on the top graph, the F1 and F2 responses (second and third plot from the top, right side of the slide). These are the resonant and anti-resonant frequencies of the imaginary part. They have a different response profile. And then, the big height change of the real part has even further different profiles. So this pattern gives us the ability to discriminate between these 10 vapors quite nicely. These are the individual vapors in the experiment.



>> Radislav Potyrailo: The next example shows that if we use peptide-coated gold nanoparticles (shown on the left of the slide), then we can recognize volatiles of very similar dielectric constants. Very often, the plots that are showing this recognition are the scores plots of the principal component analysis (PCA), which is the unsupervised tool of machine learning that is very often used for analysis of sensor arrays and individual sensors (which you will see more and more).

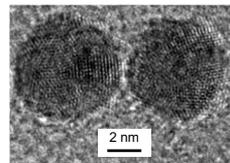


>> Radislav Potyrailo: We are showing here an example of rejection of interference. We have learned how to develop a transducer that will push the response to interference in one plane and the response to our analyte in another plane. So, the detection limit for this model analyte, in relation to a relatively high humidity of 75%, gave us that rejection ratio of two billionfold.



>> Radislav Potyrailo: When we have such a prominent sensor, then we can use this type of system to recognize different complex aromas, as I am showing you in this graph. You can see that there are four dimensions for the response of this individual sensor. From an optical perspective, there are nice solutions for developing such multivariable sensors with optical-detection principles.

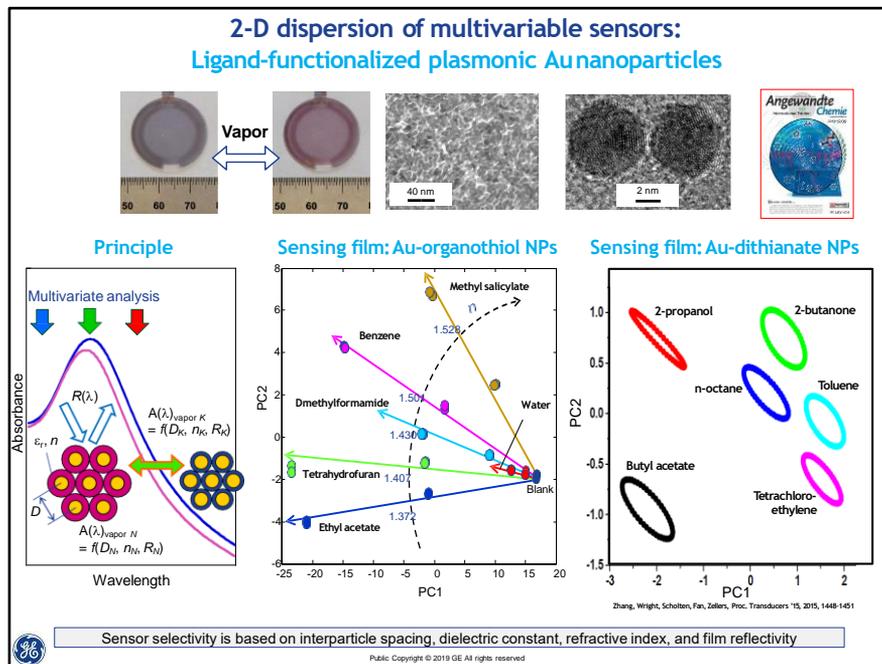
Chemistry and physics of color formation: Applications for multivariable photonic sensors



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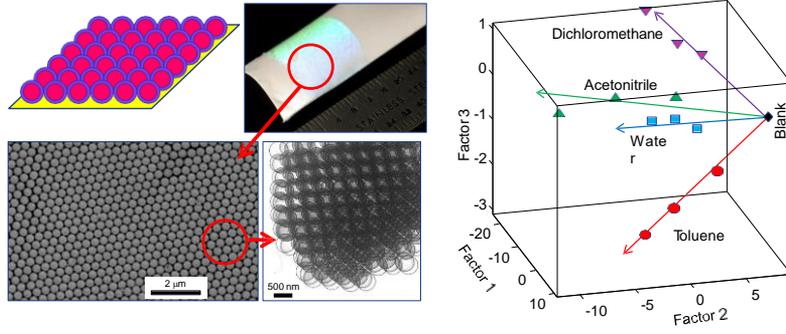
All photos by GE Research

>> Radislav Potyrailo: On this slide, I am showing on the left an example of a chemical response: This is an organic dye that one of our students was breathing on during Science Day. On the right, I am showing examples of physical responses, where iridescence of nanostructures – either natural or nanofabricated – give color changes or plasmonic changes in nanoparticles.



>> Radislav Potyrailo: Talking about plasmonic changes, this slide shows examples of not only color changes (which have been noticed a long time ago), but also discrimination between different types of volatiles (when we look at the full spectrum), which we have been showing in our labs. The University of Michigan showed very nice work of these nanoparticles, as well.

Photonic colloidal crystal gas sensors for 3-D sensor dispersion



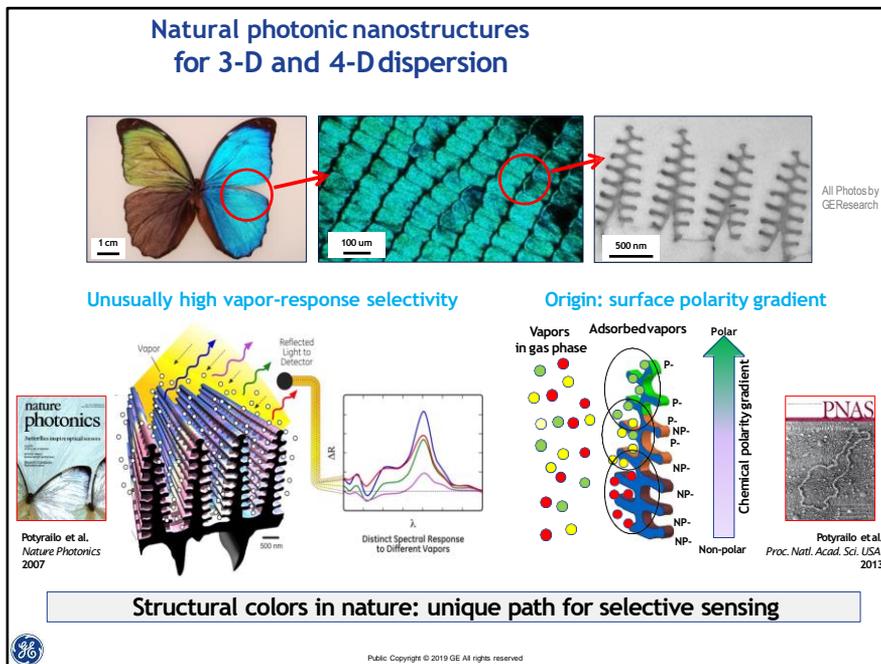
Sensor selectivity is based on optical lattice constant of colloidal crystal with cores and shells of nanospheres responding to diverse vapors

R. A. Potyrailo, Z. Ding, M. D. Butts, S. E. Genovese, T. Deng, *IEEE Sens. J.*, 2008, 8, 815-822.

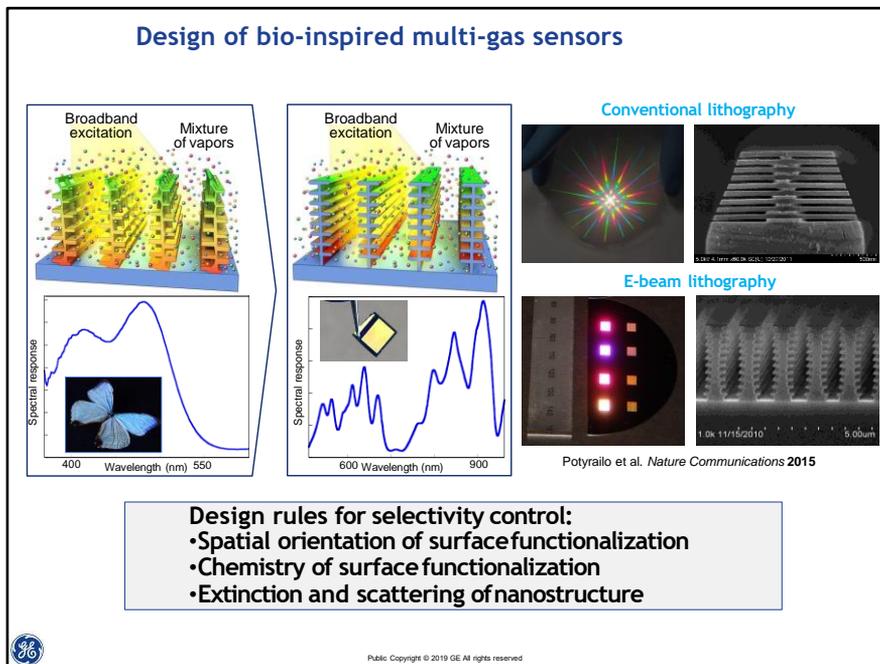


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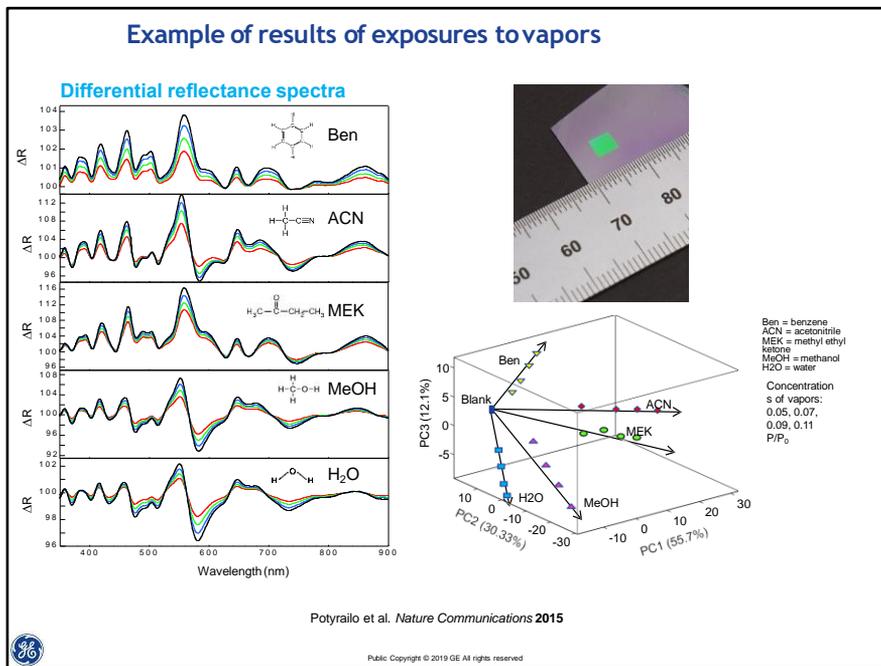
>> Radislav Potyrailo: You can have more significant dispersion, such as three-dimensional dispersion, if you have colloidal crystals arranged for gas sensing. On this slide, I show an example of three-dimensional dispersion using a colloidal crystal cell.



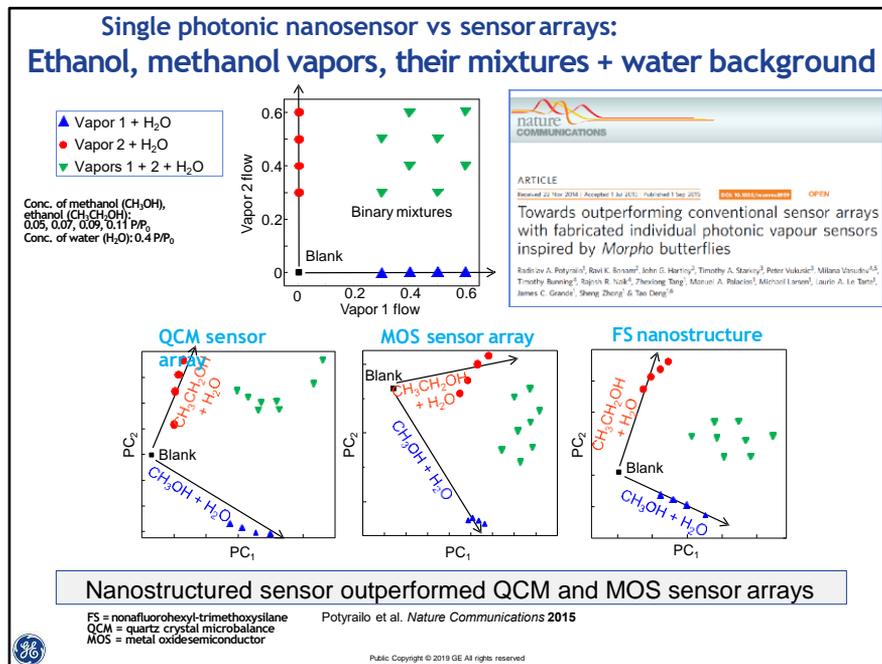
>> Radislav Potyrallo: The biggest success that we have obtained, in terms of dispersion, was when we were using photonic structures from iridescent butterflies. What we saw initially was that under an optical or electronic microscope, these structures were nanostructures of periodic lamellae. These nanostructures are arranged on ridges, which helps in getting the iridescence. When we expose these structures to different types of vapors, they selectively respond to different volatiles in different optical domains.



>> Radislav Potyrailo: We have learned that the selectivity comes from the chemical gradient, or polarity, along the height of this nanostructure, which helped us big time to develop design rules for our engineered sensors. On the left side of the slide, I am showing the spectrum of a natural butterfly (left) and the spectrum of our nanofabricated structures (right), which we are making with different tools (right side of the slide), such as conventional lithography (top) and electron-beam lithography (bottom). At the bottom of the slide, I outline the design rules that we are applying for the detection of different types of gases.



>> Radislav Potyrailo: This slide shows the detection of different types of volatiles with a single sensor. This is done at room temperature.



>> Radislav Potyrailo: Because we are part of the electronic nose and sensor array community, on this slide, we were comparing the behavior of a nanostructured single sensor with our sensor arrays, such as quartz-crystal microbalance (QCM) and metal-oxide semiconductor (MOS) sensor arrays. We saw that our individual sensor was outperforming those two popular sensor arrays.

Advancing design rules of nanostructures for high temperature gas-sensing applications

Selectivity control for vapors at room temp.



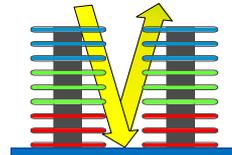
- Polymeric nanostructure
- Absorption and adsorption of vapors

Selectivity control for gases at high temp.

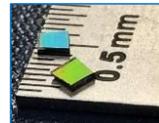
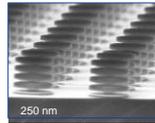


- Inorganic nanostructure
- Catalytic reactions of gases

Interference rejection control



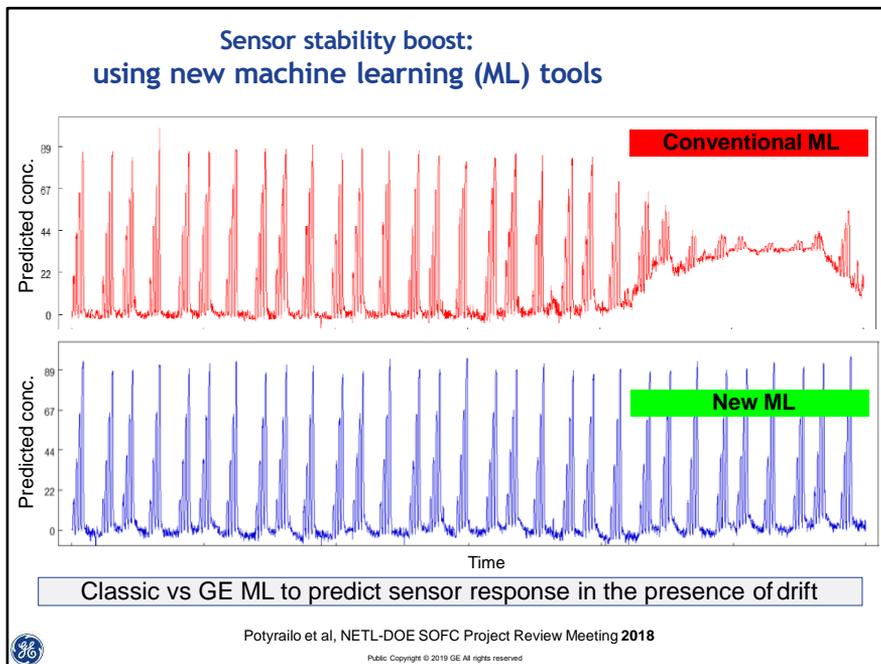
- Multi-material inorganic nanostructure
- Catalytic reactions of gases



Potyrailo et al. *Nature Photonics* 2007
Potyrailo *Chem. Soc. Rev.* 2017

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>> Radislav Potyrailo: We also moved to high-temperature applications, because for fuel cells and other types of applications, it is important to have sensors operating at high temperature. We are doing this work with our collaborators from SUNY Polytechnic Institute.



>> Radislav Potyrailo: The important aspect that I would like to show is that when we are developing our sensors and calibrating them using conventional machine-learning (ML) tools, in the presence of drift, we lose our quality of prediction. But when we use the sensor design in combination with new ML tools, then we keep the quality.

Top three learnings from Segment 3

Segment 3:

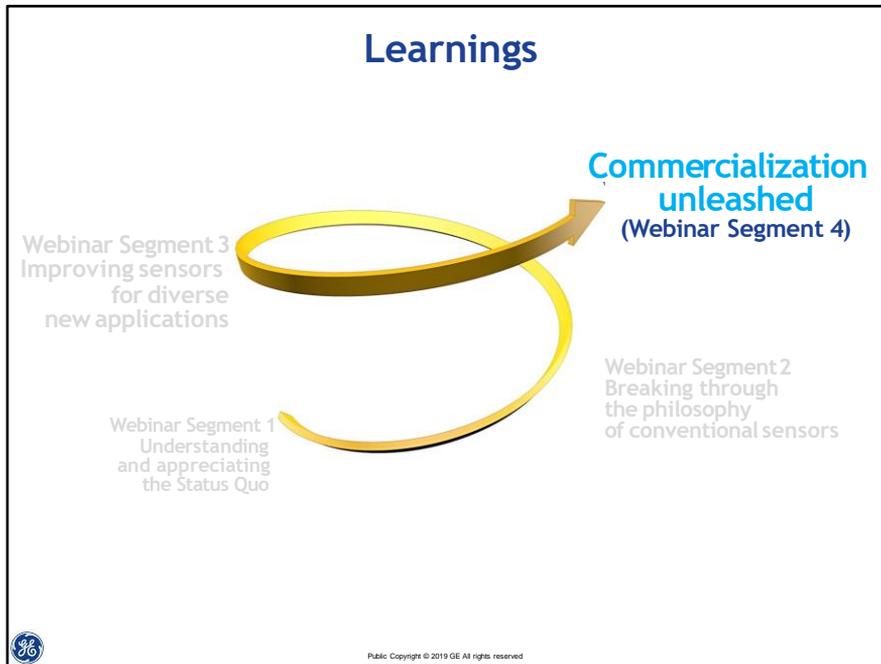
Breaking through the philosophy of conventional sensors

1. Multivariable sensors is the natural next phase driven by progress in sensors science and technology
2. Demonstrated multivariable sensors have 2-D, 3-D, 4-D dispersion
3. Reduction/elimination of drift in multivariable sensors by focused sensor design and simple data analytics



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>> Radislav Potyrailo: So, this is what we learned from Segment 3: First, there is another way of getting the signals out from individual sensors when you have more than one output, and we're calling this type of sensors "multivariable sensors." Second, we are developing multivariable sensors that have two-dimensional, three-dimensional, and four-dimensional dispersion. Third, by focused sensor design and simple data analytics, we can reduce or eliminate the drift.



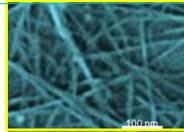
[There is no text for this slide.]

Gas nanosensors: Examples of today's commercial offerings



Rolla, MO

**Carbon
nanotubes**

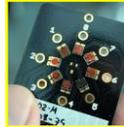


<https://www.brewerscience.com>



Salt Lake City, UT

Nanofibers



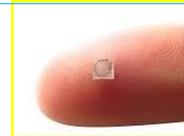
<http://www.vaporsens.com/products/>



N5 Sensors, Inc.

Rockville, MD

**Semiconductor
nanostructures**

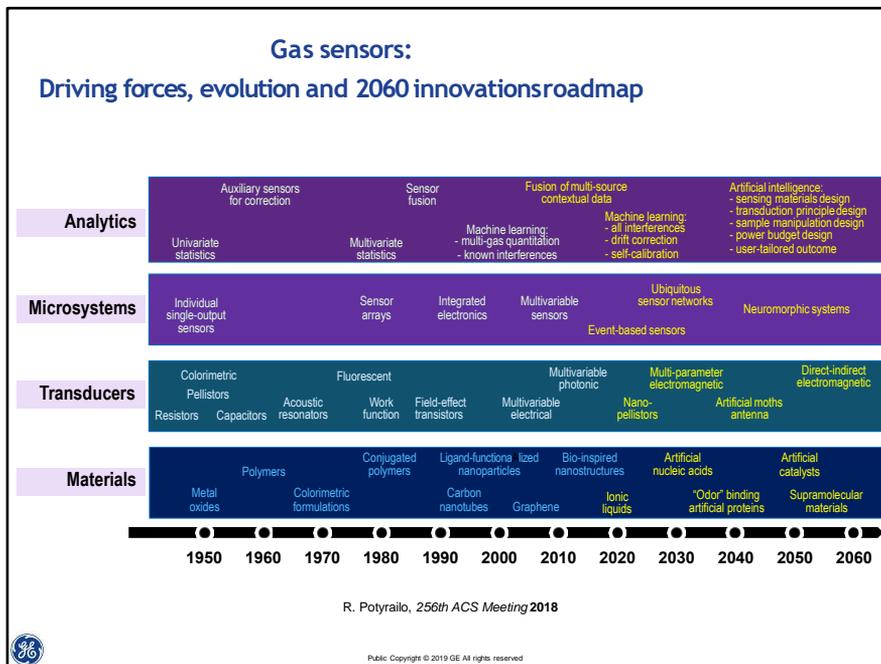


<http://n5sensors.com/>



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>> Radislav Potyrailo: To conclude, I would like to show you that there are successful companies out there, in the U.S., for example. These companies are commercializing different types of gas sensors with nanomaterials quite successfully, which is very inspiring for us, as a research community. If we are looking forward into the future, what type of roadmap is in front of us, and what type of things are we focusing on?



>> Radislav Potyrailo: You can see this roadmap, which includes advances in materials, transducers, microsystems, and data analytics, and, overall, bringing this type of analytical devices to new levels. This is an innovation roadmap for 2060.

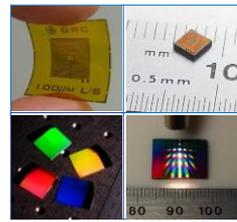
Summary: Multivariable sensor solutions for demanding applications

Instruments based on mature analytical concepts

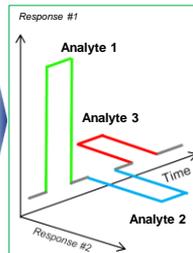


<http://dx.doi.org/10.1016/j.procs.2016.05.018>
 Proceedings of the 12th International Conference on Environmental and Analytical Chemistry (ICEAC-2016)
 Ch. Ghazvini, M. Ghazvini
 © 2016, May 18-20, 2016

Sensors and systems based on multivariable sensing concepts



Photos by R. A. Potyrailo



- Value of multivariable nanosensors over existing traditional analytical instruments: multi-gas detection and rejection of interferences using individual sensors
- Detection sensitivity: part-per-million → part-per-billion → part-per-trillion levels
- High sensor stability due to reduced number of noise sources
- Simplified device packaging



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>> Radislav Potyrailo: In summary, we are not competing, but, with our designs, we are complementing the performance of mature analytical instruments. You see that these complementary efforts pay off with the capabilities that we discussed today.

Acknowledgments

GE Global Research

B. Amm

J. Ashe

J. Brewer

C. Collazo-Davila

T. Deng

S. Go

C. Henderson

J. Iannotti

A. Minnick

M. Nayeri

Y. Lee

A. Obi

M. Palacios

M. Pietrzykowski

B. Scherer

A. Shapiro

V. Srivastava

N. Stoffel

R. St-pierre

A. Tang

S. Zhong

University at Albany

H. Ghiradella

J. Hartley

R. Bonam

M. Carpenter

N. Karker

N. Houlihan

V. Vulcano Rossi

University of Exeter

P. Vukusic

T. Starkey

Air Force Research Lab

R. Naik

J. Slocik

M. Vasudev

N. Kelley-Loughnane

Avery Dennison

DJ Lee

E. McGinniss



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>> Radislav Potyrailo: These are my collaborators and funding agencies. Thank you for your attention.

Thank you

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>> Stacey Standridge: Thank you. That was a really interesting and informative presentation. We had a couple of questions come in, but given the time, I think I will just pose one question to you before we wrap up for the day. Near the end of your presentation, you have a 2060 innovation roadmap, and I was wondering: From your perspective, what are the top priorities for that roadmap?

>> Radislav Potyrailo: I think the top priorities are basically the ability to think ahead what the application scenarios should be for a particular sensor that we're developing and then understand what type of interferences or other types of challenges this sensor will experience. The priority will be to make sure that you are rejecting the interferences that are important to reject in that application. Another priority is that you are focusing on the lifetime of that particular sensor and understanding how to keep that lifetime as long as appropriate because, depending on the application, it can be two weeks, but if it is bedside monitoring, it can be five or ten years. So, knowing those values or numbers up front will give us the priorities. (Continued)

Thank you

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>> Stacey Standridge: That makes perfect sense, and I think that's something we hear very much: It all depends on the case. With that, I want to thank you, Dr. Potyrailo for your time today, and I want to thank everybody who participated in the webinar. Again, a friendly reminder that we have National Nanotechnology Day on October 9th (for 10⁻⁹) next week, and we hope everyone can participate. And please check nano.gov for more updates.

>> Radislav Potyrailo: Thank you, everybody.