UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

LAM RESEARCH CORP.,

Petitioner,

v.

INPRIA CORP.,

Patent Owner.

Case No. IPR2024-00033

Patent No. 9,823,564

REBUTTAL DECLARATION OF KAREY HOLLAND, PH.D. IN SUPPORT OF PETITIONER'S REPLY TO PATENT OWNER'S RESPONSE AND OPPOSITION TO MOTION TO AMEND

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EXHIBIT LIST

Exhibit	Description	Previously Submitted
1001	U.S. Patent No. 9,823,564 ("'564 patent")	X
1002	Expert Declaration of Karey Holland, Ph.D. ("Holland Declaration")	X
1003	U.S. Patent Publication No. 2011/0293888 ("'888 publication")	X
1004	U.S. Application No. 12/850,867 Prosecution History ("'867 application PH")	X
1005	U.S. Application No. 61/350,103 ("'103 provisional")	X
1006	Declaration Sylvia Hall-Ellis, Ph.D. Regarding Authenticity and Public Availability ("Hall-Ellis Declaration")	X
1007	'564 Patent Prosecution History ("'564 patent PH")	X
1008	Stowers, Jason K. *Direct patterning of solution deposited metal oxides*. Oregon State University, 2008 ("Stowers 2008")	X
1009	U.S. Patent No. 10,782,610 Prosecution History ("'610 patent PH")	X
1010	Stowers, Jason, and Douglas A. Keszler. *High resolution, high sensitivity inorganic resists*. Microelectronic Engineering 86.4-6 (2009): 730-733 ("Stowers 2009")	X
1011	Complaint for *Inpria Corp. v. Lam Research Corp.*, 1-22-cv-01359	X
1012	Jouve, A., et al. *Overcoming pattern collapse of ultra high resolution dense lines obtained with EUV resists*. Advances in Resist Technology and Processing XXII. Vol. 5753. SPIE, 2005 ("Jouve")	X
1013	Stowers, Jason, Alan Telecky, and Andrew Grenville. *Imageable oxides: new solutions for resist*. EUVL Symposium. 2010, available at https://euvlsymposium.lbl.gov/pdf/2010/pres/RE-08.pdf	X
1014	Telecky, Alan, et al. *Photopatternable inorganic hardmask*. Journal of Vacuum Science & Technology B 28.6 (2010): C6S19-C6S22 ("Telecky 2010")	X
1015	Summons for *Inpria Corp. v. Lam Research Corp.*, 1-22-cv-01359	X
1016	Amador et al. *Patterning chemistry of HafSOx resist*. Advances in Patterning Materials and Processes XXXI. Vol. 9051. SPIE, 2014	X
1017	Reserved	X

Exhibit	Description	Previously Submitted
1018	Decker, Shawn R. *On the Properties and Characteristics of Solution Processed Hafnium Based Thin-Films from Aqueous Precursors*. Oregon State University, 2017	X
1019	Oleksak et al. *Evaluation of Thermal and Radiation Induced Chemistries of Metal Oxo–Hydroxo Clusters for Next-Generation Nanoscale Inorganic Resists*. ACS Applied Nano Materials 1.9 (2018): 4548-4556	X
1020	Reserved	X
1021	U.S. Patent No. 7,773,365	X
1022	Stowers et al. *Directly patterned inorganic hardmask for EUV lithography*. Extreme Ultraviolet (EUV) Lithography II. Vol. 7969. SPIE, 2011 ("Stowers 2011")	X
1023	Reserved	X
1024	Trikeriotis et al. *Development of an inorganic photoresist for DUV, EUV, and electron beam imaging*. Advances in Resist Materials and Processing Technology XXVII. Vol. 7639. SPIE, 2010 ("Trikeriotis") (Library Version)	X
1025	Matsuzawa et al. *Theoretical estimation of absorption coefficients of various polymers at 13nm*. Journal of Photopolymer Science and Technology 12.4 (1999): 571-576.	X
1026	Puurunen et al. *Hafnium oxide films by atomic layer deposition for high-κ gate dielectric applications: Analysis of the density of nanometer-thin films*. Applied Physics Letters 86.7 (2005).	X
1027	Ober and Giannelis. *New oxide nanoparticle extreme-UV photoresists achieve high sensitivity*. SPIE Newsroom (2014): 2-4.	X
1028	*Curriculum Vitae* of Sylvia D. Hall-Ellis, Ph.D.	X
1029	*Curriculum Vitae* of Karey L. Holland, Ph.D.	X
1030	Anderson, Jeremy T., et al. *Solution-processed HafSOx and ZircSOx inorganic thin-film dielectrics and nanolaminates*. Advanced Functional Materials 17.13 (2007): 2117-2124 ("Anderson")	X
1031	Trikeriotis et al. *Development of an inorganic photoresist for DUV, EUV, and electron beam imaging*. Advances in Resist Materials and Processing Technology XXVII. Vol. 7639. SPIE, 2010 ("Trikeriotis") (Electronic Version)	X
1032	Deposition Transcript of Jason Stowers, Ph.D. (October 11, 2024)	X

Exhibit	Description	Previously Submitted
1033	Deposition Transcript of Sanjay K. Banerjee, Ph.D. (September 27, 2024)	
1034	Oleksak RP & Herman GS, *Characterization of High-Resolution HafSOx Inorganic Resists*. Proc. SPIE 9048, Extreme Ultraviolet (EUV) Lithography V, 90483H (2014).	
1035	Trakhetenberg S et al., *Non-Catalytic Photo-Induced Immobilization Process In Polymer Films*, *in* Physico-Chemical Phenomenia in Thin Films & At Solid Surfaces (Trakhetenberg, L, Lin, S; Ilegbusi, eds. 2007)	
1036	Krysak, Marie E., et al., *Investigation of Novel Inorganic Resist Material for EUV Lithography*. Proc. SPIE 9048, Extreme Ultraviolet (EUV) Lithography V, 904805 (2014) ("Krysak")	
1037	Brainard, Robert L, *Photoresists for EUV Lithography*, *in* EUV Litogarphy (Bakshi, V. Ed. 2018)	
1038	International Application Publication No. WO2008/082448 ("Keszler")	
1039	R. Chang & K Goldsby, *Chemistry* (11th ed. 2013) (excerpts).	
1040	Expert Declaration of Karey Holland, Ph.D.	
1041	Declaration of Heather Stewart	
1042	Email from Brad Engelbert, Cataloging & Government Documents Manager of Oregon State University, to Risa L. Murphy, Morgan, Lewis & Bockius (August 13, 2024)	
1043	Declaration of Patrick Franzen	
1044	Fallica et al., *Absorption Coefficient of Metal-Containing Photoresists in the Extreme Ultraviolet*, J Micro/Nanolith. MEMS MOEMS 17(2), 023505 (2018)	
1045	Reserved	
1046	Kirchauer, Heinrich, *Photolithography Simulation*, Vienna University of Technology, 1998 (available at https://www.iue.tuwien.ac.at/phd/kirchauer/diss.html) (excerpt)	
1047	Schnabel, Wolfram, *Polymers & Electromagnetic Radiation: Fundamentals & Practical Applications* (2014).	

I. INTRODUCTION

1. My name is Karey Holland, and I have been retained by counsel for Petitioner Lam Research Corporation ("Petitioner") to provide opinions on certain issues relating to U.S. Patent No. 9,823,564 (EX1001, "the '564 patent"), which I understand is assigned to Inpria Corporation ("Inpria") in the Inter Partes review proceedings with trial number IPR2024-00033.

2. I provide my opinions related to Patent Owner's Response to Petition for Inter Partes Review of the '564 Patent ("Response"), and Patent Owner's Motion to Amend the claims of the '564 patent ("the Proposed Amended Claims").

3. My opinions are based on my years of education, consulting experience, research and teaching experience, as well as my investigation and study of relevant materials.

4. I may rely upon these materials, my knowledge and experience, and/or additional materials to rebut arguments raised by the patent owner. Further, I may also consider additional documents and information in forming any necessary opinions, including documents that may not yet have been provided to me.

5. My analysis of the materials produced in this investigation is ongoing and I will continue to review any new materials as they become readily available. This declaration represents only those opinions I have formed to date. I reserve the right to revise, supplement, and/or amend my opinions stated herein based on new information and based on my continuing analysis of the materials already provided.

6. I hereby incorporate my earlier declaration (EX1002) in support of the Petition for *Inter Partes Review* in its entirety.

II. QUALIFICATIONS AND BACKGROUND

7. I have provided my qualifications, compensation and prior testimony, and background information. My curriculum vitae is attached as EX1017 to the Petition.

8. In addition to my education, knowledge of the relevant published art, training, and experience, in forming the opinions I provide in this declaration, I have also considered the exhibits in the exhibit list at the front of this declaration.

III. LEGAL PRINCIPLES

9. For my opinions in this declaration, I understand that it requires applying various legal principles. As I am not an attorney, I have been informed about various legal principles that govern my analysis. I have used my understanding of those principles in forming my opinions. I summarize my understanding of those legal principles as follows:

10. **Burden of Proof**. I understand that Petitioner bears the burden of proving unpatentability in this proceeding by a preponderance of the evidence. I am informed that this preponderance of the evidence standard means that Petitioner must show that unpatentability is more probable than not.

11. **Claim Construction**. I have also been told that when I review and consider the claims in the challenged patent that the claim term(s) should be given their ordinary and customary meaning as understood from the perspective of one of ordinary skill in the art at the time of the invention, taking into account the claim language itself, specification, and prosecution history pertaining to the patent, as well as relevant extrinsic evidence. I have applied this standard in formulating my opinions.

12. **Written Description.** I have been informed by counsel that a patent may be invalid if the patent specification does not convey to a POSA that the inventors in fact invented what is claimed. I understand that the assessment of compliance with the written description requirement is an objective inquiry into the four corners of the patent from the perspective of a POSA. This

inquiry is often articulated as whether a POSA would recognize that the inventor was in possession of the claimed invention as of the effective filing date.

13. I have been informed by counsel that the written description requirement aims to ensure that the scope of the right to exclude, as set forth in the claims, does not overreach the scope of the inventor's contribution to the field. Compliance with the written description requirement involves both an assessment of the scope of the claims and the adequacy of the disclosure to support claims of such scope.

14. I have been informed by counsel that a written description of an invention involving a genus of chemical compounds, which are compositions of matter, is adequate when the patent specification provides a precise definition of the genus, such as by structure, formula, or chemical name, or physical properties sufficient to distinguish the claimed subject matter from other materials.

15. I understand that when a patent claims a genus using functional language to define a desired result, as in the present case ("wherein exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating"), the specification must demonstrate that the inventor has made a generic invention that achieves the claimed result across its full scope. I have been informed by counsel that this requirement may be met by demonstrating either a structure-function relationship or that the inventor has invented enough species to support a claim to the functionally claimed genus.

16. I understand that what is required is the disclosure of enough species to be representative of the scope of the functionally claimed genus. The number of representative species required varies depending on the facts of the case, but that where the degree or level of the function

recited in the claim is broad or open-ended, disclosure of multiple species representing only a small fraction of the scope of the claimed function is inadequate to meet the written description requirement. I further understand that for inventions in an unpredictable art, adequate written description of a genus which embraces widely variant species cannot be achieved by disclosing only one species within the genus.

17. I have been informed by counsel that embodiments that post-date the patent's priority date, including an accused infringing product, can be taken into consideration when determining the scope of the functionally claimed genus and whether the species disclosed in the patent are sufficiently representative of the product accused to entitle the inventor to capture such post-priority products within its scope.

18. Finally, I have been further informed that post-priority date references and actions on the part of the inventors may be considered in determining whether patent claims are adequately described. Evidence showing that a claimed genus does not disclose a representative number of species may include evidence of species that fall within the claimed genus that are not disclosed by the patent, but, instead, post-date the priority date of the patent.

19. **Enablement**. I have been informed by counsel that a patent may be invalid if the patent specification does not enable a POSA to make and use the claimed invention without undue experimentation. I understand that the enablement requirement has two aspects to it: a "how to make" aspect and a "how to use" aspect. A POSA must be able to make the claimed invention across its full scope without undue experimentation.

20. Whether the claims of a patent are enabled is assessed based on the teachings of the specification in conjunction with what is generally known to a POSA as of the patent's priority

date. I understand that the relevant question is whether, with the specification and the knowledge available at the time of the invention, a POSA could perform routine experimentation to make and use the full scope of the claimed invention. In answering this question, the following factors (known as the *Wands* factors) are typically considered and balanced:

- the nature of the claimed invention;
- the breadth of the patent claims;
- the relative skill of those in the art;
- the presence or absence of working examples in the patent specification;
- the amount of direction or guidance presented in the patent specification;
- the predictability or unpredictability of the art;
- the state of the prior art; and
- the quantity of experimentation necessary to carry out the full scope of the claimed invention

21. The patent specification must be more than a mere starting point for further research to obtain the claimed compositions. The patent specification is not enabling if it amounts to no more than an invitation to a POSA to engage in research that may or may not result in the claimed compositions.

22. I understand that while the enablement analysis accounts for the knowledge of a POSA, the patent specification, not the knowledge of a POSA, must supply the novel aspects of an invention to constitute adequate enablement; in other words, it is improper to rely on a POSA's knowledge to fill the gaps in the specification. Where a POSA is deemed to have little or no knowledge independent from the instructions in the patent specification (such as with undeveloped or "nascent" technologies), one cannot merely rely on the information known to a POSA; instead,

the specification must provide a specific and useful teaching to a POSA for how to make and use the full scope of the claimed invention.

23. I have been informed by counsel that post-priority date references and actions by the inventors may be considered in determining whether compound claims are enabled, as subsequent failures or subsequent undue experimentation needed to make and use the claimed compounds is evidence that the patent was non-enabling as of its priority date.

24. **Indefiniteness**. I am informed that the patent statute requires that a patent must conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the inventor or a joint inventor regards as the invention. I am also informed that some claims, despite being issued by the Patent Office, can later be found to be invalid if they contain claim terms that are indefinite. I am informed that a patent is invalid for indefiniteness if its claims, when read in light of the specification delineating the patent and the prosecution history, fail to inform, with reasonable certainty, those skilled in the art about the scope of the invention. Thus, I understand that indefiniteness is to be evaluated from the perspective of someone of ordinary skill in the relevant art at the time the patent was filed, reading the claims in light of the patent's specification and prosecution history.

25. **Anticipation**. I have been asked to consider the question of anticipation, namely, whether the claims cover something that is new, or novel. I am told that the concept of anticipation requires that each and every element of a challenged claim is present in or otherwise taught by a single reference. I also understand that an anticipatory reference does not need to explicitly describe each element because anticipation can occur when a claimed limitation is necessarily present, inherent or otherwise implicit in the relevant reference.

26. **Obviousness**. I have been asked to consider the question of obviousness. Again, I am told that this analysis must be from the perspective of the person of ordinary skill in the art, and whether such person would consider any differences between the prior art and what is claimed to have been obvious. To make this assessment, I have been informed that the concept of patent obviousness involves four factual inquiries: the scope and content of the prior art; the differences between the claimed invention and the prior art; the level of ordinary skill in the art; and so-called secondary considerations of non-obviousness.

27. I have further been instructed that one must consider what the person of ordinary skill in the art knew, and what the art taught; suggested; or motivated the person of ordinary skill in the art to further pursue; and to differentiate between steps that were routinely done (such as in response to known problems, steps, or obstacles), and those which, for example, may have represented a different way of solving existing or known problems. Further I have been informed that one cannot use the challenged patent (here, the ’564 patent) as a guide from which to select prior art elements, or otherwise engage in hindsight.

28. I am also informed that when there is some recognized reason to solve a problem, and there are a finite number of identified, predictable, and known solutions, a person of ordinary skill in the art has good reason to pursue the known options within his or her technical grasp. If such an approach leads to the expected success, it is likely not the product of innovation but of ordinary skill and common sense. In addition, when a patent simply arranges old elements, with each performing its known function, and yields no more than what one would expect from such an arrangement, the combination is obvious.

29. I understand that before reaching any final conclusion on obviousness, the obviousness analysis requires consideration of objective indicia of nonobviousness, if offered. These must be considered to ensure that, for example, there were not some unanticipated problems, obstacles, or hurdles that may seem easy to overcome in hindsight, but which were not readily overcome prior to the relevant invention date of the patents/claims at issue here. I understand that these objective indicia are also known as "secondary considerations of non-obviousness," and may include long-felt but unmet need and unexpected results, among others. I also understand, however, that any offered evidence of secondary considerations of non-obviousness must be comparable with the scope of the challenged claims. This means that for any offered evidence of secondary considerations of non-obviousness to be given substantial weight, I understand the proponent of that evidence must establish a "nexus" or a sufficient connection or tie between that evidence and the merits of the claimed invention, which I understand specifically incorporates any novel element(s) of the claimed invention. If the secondary considerations evidence offered actually results from something other than the merits of the claim, then I understand that there is no nexus or tie to the claimed invention. I also understand it is the patentee that has the burden of proving that a nexus exists.

30. However, I understand that secondary considerations will not overcome a strong showing of obviousness.

IV. PERSON OF ORDINARY SKILL IN THE ART ("POSA")

31. As explained in my previous declaration (EX1002, § IV), I have been informed by counsel that my analysis is to be conducted from the perspective of a person of ordinary skill in the art at the time of the invention. I also understand that the person of ordinary skill in the art is

assumed to know, understand, and be familiar with all of the relevant prior art, and that such person is a person of ordinary creativity.

32. It is my opinion that a POSA would have been an individual with at least a graduate degree, such as a masters or Ph.D., in a relevant discipline such as in chemistry, materials science, chemical engineering, physics, or an equivalent field. A POSA would also have 1-3 years of experience in the research, design, development and/or testing of resist materials. I am clearly a POSA and have applied this standard throughout my declaration. I understand Dr. Banerjee adopted my definition of a POSA.

33. I am a POSA and have applied this standard throughout my declaration.

V. OPINIONS IN SUPPORT OF PETITIONER'S REPLY TO PATENT OWNER'S RESPONSE

A. The '564 Patent Is Not Entitled To Claim Priority to '867 Application

34. It remains my opinion that the Challenged Claims should not be given a priority date earlier than September 18, 2015. Specifically, for the reasons stated in my First Declaration, *i.e.*, a POSA would not understand the '564 Patent inventors to have been in possession of a purported invention with a coating having a thickness from "5 nm to 30 nm" (claim 1) or "5 nm to 25 nm" (claim 6) based on the disclosure of application no. 12/850,867 ("'867 application") to which the '564 patent claims priority. EX1002, ¶¶63-68. I understand that Dr. Banerjee has opined that a POSA "would have immediately discerned that the inventors possessed the claimed range of 5 nm to 30 nm." EX2009, ¶¶23-29. I disagree.

35. First, Dr. Banerjee cites portions of the '867 application disclosing *how* to deposit coatings on a substrate—e.g., via spin-coating—and *how* to achieve various coating material

thicknesses—*e.g.*, varying the viscosity, precursor concentration, spin speed, or other coating parameters. EX2009, ¶¶24-26. However, even if a POSA knows *how* to achieve a particular coating thickness, the POSA would still not know *which* thicknesses are appropriate or inventive. Thus, these examples do not provide a POSA with any guidance what thicknesses are within the scope of the described "invention."

36. Second, Dr. Banerjee points to the exemplary ranges in the '867 application specification:

> In some embodiments, the coating materials can have an average thickness of no more than about 1 micron, in further embodiments no more than about 250 nanometers (nm), in additional embodiments from about 1 nanometers (nm) to about 50 nm, in other embodiments from about 1 nm to about 40 nm and in some embodiments from about 1 nm to about 25 nm.

EX1004, 23:18-24. Dr. Banerjee opines that because the ranges of 5 nm to 30 nm and 5 nm to 25 nm are encompassed by the ranges disclosed in the '867 Application, a POSA "would have immediately discerned that the inventors possessed the claimed range[s]." EX2009, ¶¶28-29. Dr. Banerjee also points to the '867 application's statement that a POSA "will recognize that additional ranges of thicknesses within the explicit ranges above are contemplated and are within the present disclosure." EX1004, 23:18-24.

37. Dr. Banerjee does not explain *why* a POSA would have understood the inventors to have possessed the thicknesses of 5 nm to 30 nm and 5 nm to 25 nm. There are numerous other ranges within those disclosed in the '867 application and it is not clear *how* a POSA would identify 5 nm to 30 nm or 5 nm to 25 nm as being the invention. The '867 application never even mentions

thicknesses of 5 nm or 30 nm. Similarly, a POSA would not be able to identify what *other* ranges of thicknesses the inventors possessed as their alleged invention. Dr. Banerjee agreed, testifying that he had not considered whether a POSA would have understood the inventors to have possessed any other ranges based on the '867 application, and that he would need "more context" than just the paragraph providing the ranges of no more than about 1 μm, no more than about 250 nm, about 1 nm to about 50 nm, about 1 nm to about 40 nm or about 1 nm to about 25 nm. EX1033, 18:-43:14 ("In my report I was asked to opine on two specific ranges: 5 to 30 and 5 to 25. For the other ranges you have to, you know, give me more context.").

38. Dr. Banerjee ignores the inventors' statement during the prosecution of the '564 patent that the prior art thickness of 35 nm and the claimed range of 5 to 30 nm "do not overlap and they are not close enough that one would expect them to have the same properties." EX1007 , 508; EX1033, 89:14-91:3 (admitting he did not discuss the '564 patent prosecution history in his declaration). In view of these statements, a POSA could not determine which thicknesses within the ranges disclosed in the '867 application would "have the same properties" as those of the claimed invention. Accordingly, a POSA would not have recognized that the inventors possessed the claimed invention based on the disclosure of the '867 application.

B. The Materials in Stowers 2008 Are Materially The Same as Those Disclosed In The '564 Patient

39. It remains my opinion that the HafSOx and ZircSOx coating materials of Stowers 2008 and '564 patent are the same in all material respects. EX1002 ¶¶101-104. I understand that Dr. Banerjee opines that a POSA "would have understood that differences in the ratios and concentrations of the materials used to form the coatings, as well as the differences in the manner

in which those materials are combined, can result in coating films with different characteristics." EX2009, ¶30. Dr. Banerjee generalized statement is not supported, however, by the evidence.

40. For example, Dr. Stowers (the author of Stowers 2008 and an inventor of the '564 Patent) in an email to Inpria's then-CEO, Dr. Grenville, stated that the "[r]elevant magic sauce recipe is on page 68" of his dissertation (i.e., of Stowers 2008). EX2008, 6. It is unclear whether "page 68" refers to the PDF numbering system (which is identical to the numbering of EX1008) or to the page numbers at the top of each page (which corresponds to page 84 of EX1008). However, upon review, the "magic sauce recipe" could only refer to the page 68 of the PDF, which provides the method used for forming the HafSOx and ZircSOx photoresists. EX1008, 68. In contrast, page 84 is part of the introduction to Chapter 4 (which also relates to HafSOx and ZircSOx), but describes his findings that HafSOx and ZircSOx are "high-sensitivity inorganic resist[s]." *Id.*, 84. Indeed, Dr. Stowers admitted that a "recipe" is only found on page 68 of EX1008—and not on page 84 of EX1008. EX1032, 62:20-65:5. Dr. Grenville replied that the magic sauce recipe is "covered by the patent application." EX2008, 6. Dr. Stowers testified that although he could not be sure that the "patent application" was a draft of an application that led to '564 patent, at the time, Inpria's other patent applications related to "solar cell material options" or "chalcogenide materials," which were not described in his dissertation (Stowers 2008). EX1032, 69:20-71:7. Thus, it is clear that the precursor solutions and coatings of Stowers 2008 and '564 patent are the same in all material aspects.

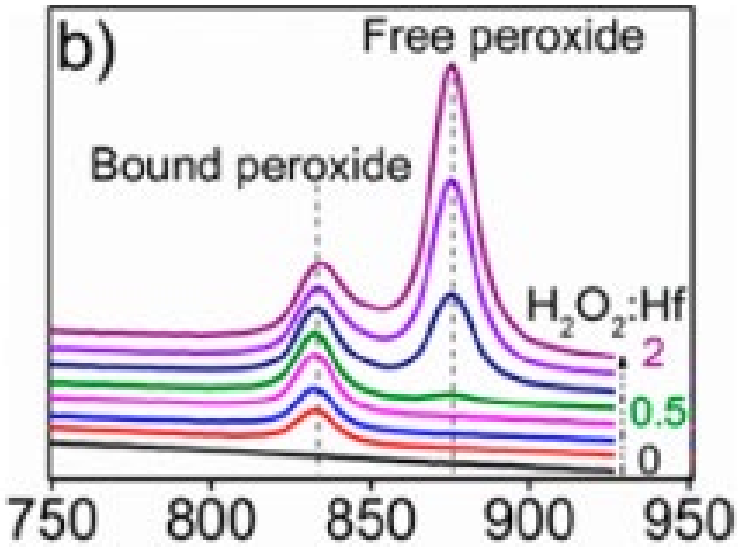
41. Moreover, as I explain below any differences in the concentration of the precursor solutions and the processing conditions between the coating materials of Stowers 2008 and '564 patent do not materially change the properties of the coating.

42. The precursor solution of Stowers 2008 comprises a final metal concentration of 0.5 M. EX1008, 68. The precursor solution of Example 1 of the ’564 patent comprises a final metal concentration of 0.15 M (for Hf-based resists) or 0.16 M (for Zr-based resists). EX1001, 21:57-67. Dr. Banerejee states that the difference in the metal concentrations “could impact the ability to deposit the resulting coating to the desired thickness.” EX2009, ¶38. Yet, publications making and testing HafSOx have shown that films can be formed using metal concentrations of either 0.5M or 0.15M. EX1034, 2 (“Final solutions . . . were 0.15 M or 0.5 M in concentration on the basis of hafnium.”). Moreover, as Dr. Banerjee explained during his deposition, in addition to viscosity (i.e., metal concentration), spin-coating speed also controls the film thickness. EX1033, 75:12-76:3. Accordingly, the difference in metal concentration between precursor solutions Stowers 2008 and the ’564 patent do not change the resulting film.

43. The precursor solution of Stowers 2008 contains a metal to peroxide ratio of 1:0.75. EX1008, 68. The precursor solution of Example 1 of the ’564 patent contains a metal to peroxide ratio of 1:6.62-8.81 (for Hf-based resists) or 1:0.66 to 0.88 (for Zr-based resists). EX1001, 21:10-67). Notably, the metal to peroxide ratio used in Stowers 2008 is within the range of those used for Zr-based resists in the ’564 patent, and thus would impart the same properties. Dr. Banerjee opines that higher peroxide content “improves the stability of the precursor solution,” but fails to explain how improved stability **changes the resulting film**. EX2009, ¶31. At best, Dr. Banerjee opines that a higher peroxide content could improve contrast. *Id.* ¶¶ 32-33.

44. Dr. Banerjee, however, ignores publications by Dr. Stowers explaining the role of peroxide in the HafSOx and ZircSOx. EX1019. The authors of EX1019 tested “$HfOCl_2$ solutions containing peroxide to meal ratios from 0 to 2.” *Id.*, 4. As shown in Figure 3 (reproduced below),

they found that for ratios less than 0.5 "all of the peroxide coordinates to the [hafnium] clusters." *Id.* However, when the ratio "exceeds 0.5, the free peroxide stretching mode . . . appears while the bound peroxide saturates in intensity" which "suggests that approximately two peroxide groups coordinate to each hafnium tetramer." *Id.*



Id. at Figure 3.

45. The authors performed further testing "to determine the peroxide to hafnium ratio in films." *Id.* They found that the molar ratio of peroxide to hafnium was 0.67, which "suggest[ed] peroxide binds to clusters at a rate of ~2 peroxides per Hf tetramer, and the bound peroxide is completely retained in the HafSOx thin film after spin-coating the precursor solution." *Id.* These studies show that HafSOx films of Stowers 2008 (formed from precursor solutions having a ligand : metal ratio of 1:0.75) and the HafSOx films of the '564 patent examples (made from precursor solutions having a ligand : metal ratio of 1:6.62-8.81) would have the same properties.

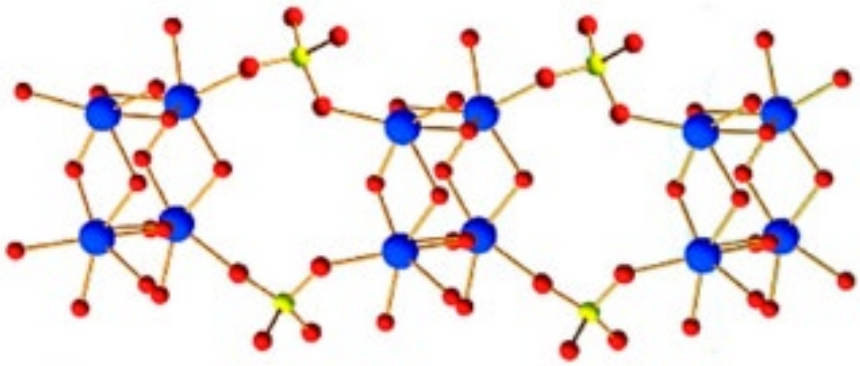
46. Dr. Banerjee also opines that the differences in amount of rest time in the methods of forming precursor solutions (15 minutes in Stowers 2008 and 5 minutes in the '564 patent)

could result in different coating films. EX2009, ¶35. I disagree. As I explained in my first declaration, both Stowers 2008 and the '564 patent disclose the same sequential addition of materials: (1) $HfOCl_2$ or $ZrOCl_2$ is mixed with H_2O_2 and allowed to rest, (2) H_2SO_4 was added to the solution and again allowed to rest, and (3) the solutions are diluted with ultrapure water. EX1002, ¶¶102-103. Stowers explains that the rest times "allow for ligand exchange" EX1008, 68. Accordingly, the 15 minutes rest period in Stowers 2008 merely allows more time for ligand exchange, and does not "result in different coating films" as Dr. Banerjee suggests.

47. The importance in the sequential addition during forming the precursor solutions is made clear by Dr. Stowers' later publications. For example, Dr. Stowers later explained that a "HafSOx solution is formed by the **sequential addition** of aqueous $HfOCl_2$, H_2O_2, and H_2SO_4." EX1019, 1. As Dr. Stowers explains, the peroxide *prevents* hafnium tetramers from polymerizing and then the sulfate links the tetramers:

> Peroxide acts as a capping ligand, stabilizing the Hf tetramer and preventing significant polymerization of clusters in solution. The addition of sulfate in the absence of peroxide results in the perception of the solution species in less than 1 h. In the presence of Hf tetramers stabilized by peroxide, sulfate acts as a bridge, linking tetramers into short chains as shown in Figure 2b.

Id., 3. Figure 2b depicts the "Hafnium tetramers linked by bridging sulfate groups (yellow spheres are S)."



Id.

48. Dr. Banerjee ignores this evidence and instead assumes that "the rest time after the mixing steps . . . limit the formation of larger, undesirable particles in the solution." EX2009, ¶36-37. Rather, it is the **order** of addition that prevents the "the formation of larger, undesirable particles in the solution," which are identical in Stowers 2008 and the '564 patent. Accordingly, the difference in rest times between formation of the precursor solutions in Stowers 2008 and the '564 patent are inconsequential to the composition of the final film.

49. I understand that Inpria has argued that the spin coating conditions could also affect the resulting films. POR, 33-34. Yet, the only difference between the spin coating conditions in Stowers 2008 and the '564 patent, is that the '564 patent includes an initial 100 RPM spin for five seconds. *Compare* EX1008, 68 *with* EX1001, 22:7. As the '564 patent explains, this "spreads the resist across the wafer." EX1001, 22:17-20. Accordingly, this will not change the composition of the resulting films.

50. Finally, I understand that Inpria has argued that the baking conditions could also affect the resulting films. POR, 33-34. Both Stowers 2008 and the '564 patent employ a pre-exposure bake and a post-exposure bake:

	Stowers 2008	**'564 Patent**
Pre-Exposure Bake	100° C (or lower) for 2 minutes (EX1008, 70)	40-200° C for 0.1 – 5 minutes *or* 50° C (EX1001, 22:20-21, 23:36-37)
Post-Exposure Bake	50° C for 30 seconds (EX1008, 70)	40-200° C for 0.1 – 5 minutes *or* 75° C (EX1001, 22:20-21, 23:36-37)

Thus, it is clear that the pre- and post-exposure bakes of Stowers 2008 are within the ranges used in the '564 patent. Accordingly, any differences in the baking conditions in Stowers 2008 and the '564 patent will not change the composition of the resulting films.

51. As shown above, each of the minor differences in precursor solutions and processing conditions is immaterial—both the '564 patent and Stowers 2008 form HafSOx and ZircSOx photoresists. Accordingly, based on my review of the solutions and methods used in the '564 patent and Stowers 2008, the HafSOx and ZircSOx coating materials of Stowers 2008 and '564 patent are the same in all material respects.

C. The Prior Art Metal Oxide Coatings Comprise Radiation Sensitive Ligands

1. Stowers 2008 Peroxide Ligands are Radiation Sensitive

52. It remains my opinion that the peroxide ligands of Stowers 2008 are "radiation sensitive ligands." Dr. Banerjee opines that a POSA "would not have understood Stowers 2008's disclosure that it is 'expected' that the extinction coefficient to change indicate[s] Stowers 2008's films included a radiation sensitive ligand" and suggests that the peroxide ligands of Stowers 2008 are not "radiation sensitive." EX2009, ¶¶44-46. Dr. Banerjee ignores the teachings of Stowers 2008, the teachings of the '564 patent, and Dr. Stowers' other publications describing the peroxide of HafSOx and ZircSOx as radiation sensitive. Indeed, Dr. Banerjee admitted during his deposition that "we now know that [peroxide] is radiation sensitive." EX1033, 165:21-166:11.

53. First, I disagree that the change in extinction coefficient described by Stowers 2008 fails to indicate that the films are "radiation sensitive." Dr. Banerjee cites no authority for his opinion. Rather, the extinction coefficient is an indication of the sensitivity of the material to radiation:

> Photoresists contain photoactive components, which absorb light of a specific wavelength and upon absorption undergo chemical transformation. As a result of such transformations, the extinction coefficient of the active ingredient changes. In addition, photoresists typically contain other components which may or may not absorb light, but do not undergo chemical reactions as result of exposure and whose extinction coefficients are therefore constant.

EX1035, 679. Accordingly, where the photoresists do not undergo chemical reactions (i.e. are not "radiation sensitive), one would expect the extinction coefficients to be constant. In Stowers 2008, however, the extinction coefficients change upon the addition of peroxide, indicating that peroxide is radiation sensitive.

54. Second, Dr. Banerjee ignores the disclosures of Stowers 2008 which describe peroxide as being radiation sensitive. For example, in a peroxopolytungstate resist "the mechanism of photoactivity has been shown to be the **decomposition of peroxide groups**." EX1008, 30. Moreover, Stowers 2008 explains that the HafSOx and ZircSOx photoresist materials were adapted from materials "designed as high-k dielectrics." *Id.*, 65. Stowers 2008 adapted those materials by **adding peroxide** to create a radiation sensitive photoresist. *Id.*, 143 (the dielectric materials were adapted to be photoresists by "changing the chemistries to a peroxide based system.") *Compare Id.*, 68 *with* EX1030, 2. It therefore follows that peroxide is the radiation sensitive element of HafSOx and ZircSOx because the peroxide imparts the ability for radiation to act as a photoresist.

55. Third, Dr. Banerjee ignores the teachings of the ’564, which as I explain above, describe HafSOx and ZircSOx coating materials which are the same in all material aspects to the HafSOx and ZircSOx coating materials of Stowers 2008. *Supra* §V.B. For example, the ’564 patent provides that “[h]ydrogen peroxide provides a desirable radiation sensitive ligand.” EX1001, 6:9-10. It explains that “upon absorption of the radiation, the peroxide functional groups are fragmented” and that “energy absorbed is transferred to the peroxide ligand which can result in the rupturing of the peroxide bond.” *Id.* 7:62-66; 11:39-42. This corresponds to the Stowers 2008 teaching that exposure to radiation causes “decomposition of peroxide groups.” EX1008, 30. Accordingly, the peroxide group of Stowers 2008 is a “radiation sensitive ligand.”

56. Fourth, Dr. Banerjee ignores Dr. Stowers’ other publications describing the peroxide of HafSOx and ZircSOx as radiation sensitive. For example, in 2010, Dr. Stowers explained that “the mechanism that leads to negative tone patterning is dependent on the presence of peroxide” which is based on “beam-induced decomposition and elimination of peroxide promotes condensation reactions, cross-linking of metal-oxo bridges, and the production of three-dimensional framework structures.” EX 1010, 21. Similarly, in a 2018 publication, Dr. Stowers explained that in HafSOx “[p]eroxide serves as the **radiation sensitive** species” which “decompose[s] via radiative . . . energy.” EX1019, 1.

57. It is therefore my opinion that it the peroxide of the Stowers 2008 coatings are “radiation sensitive ligands”—an inherent property of the peroxide.

2. Trikeriotis Organic Ligands are Radiation Sensitive

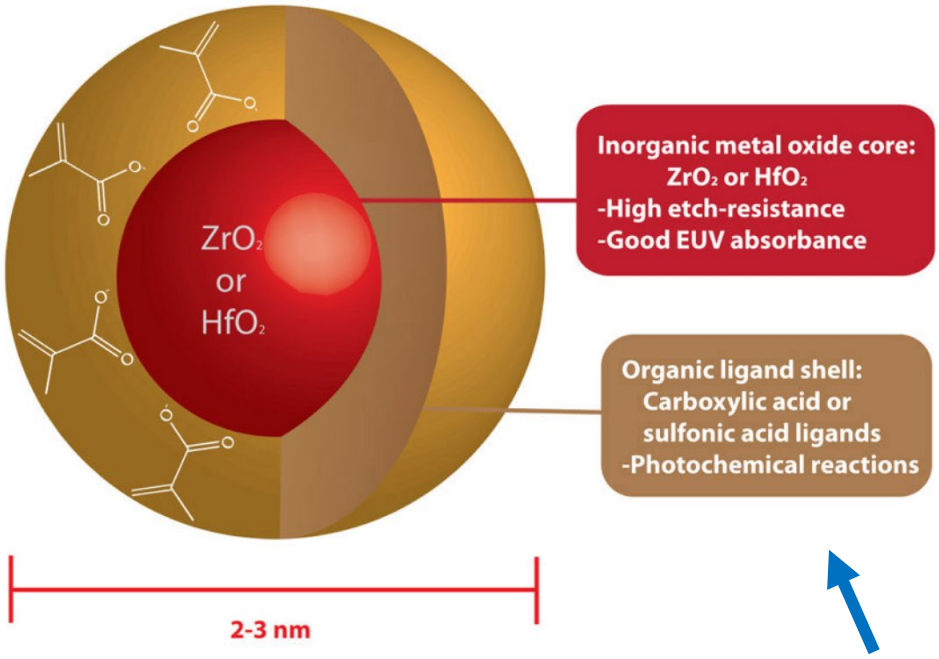
58. It remains my opinion that the organic ligands of Trikeriotis are “radiation sensitive ligands.” I understand that Dr. Banerjee has opined that those ligands are not “radiation sensitive,”

but that the mechanism of photoactivity relates to a photoacid compound within the Trikeriotis coatings. I disagree. Dr. Banerjee ignores the teachings of Trikeriotis and takes statements of later publications out of context. Dr. Banerjee also ignores later publications explaining that exposure to radiation cleaves the carboxylic group of the organic ligands.

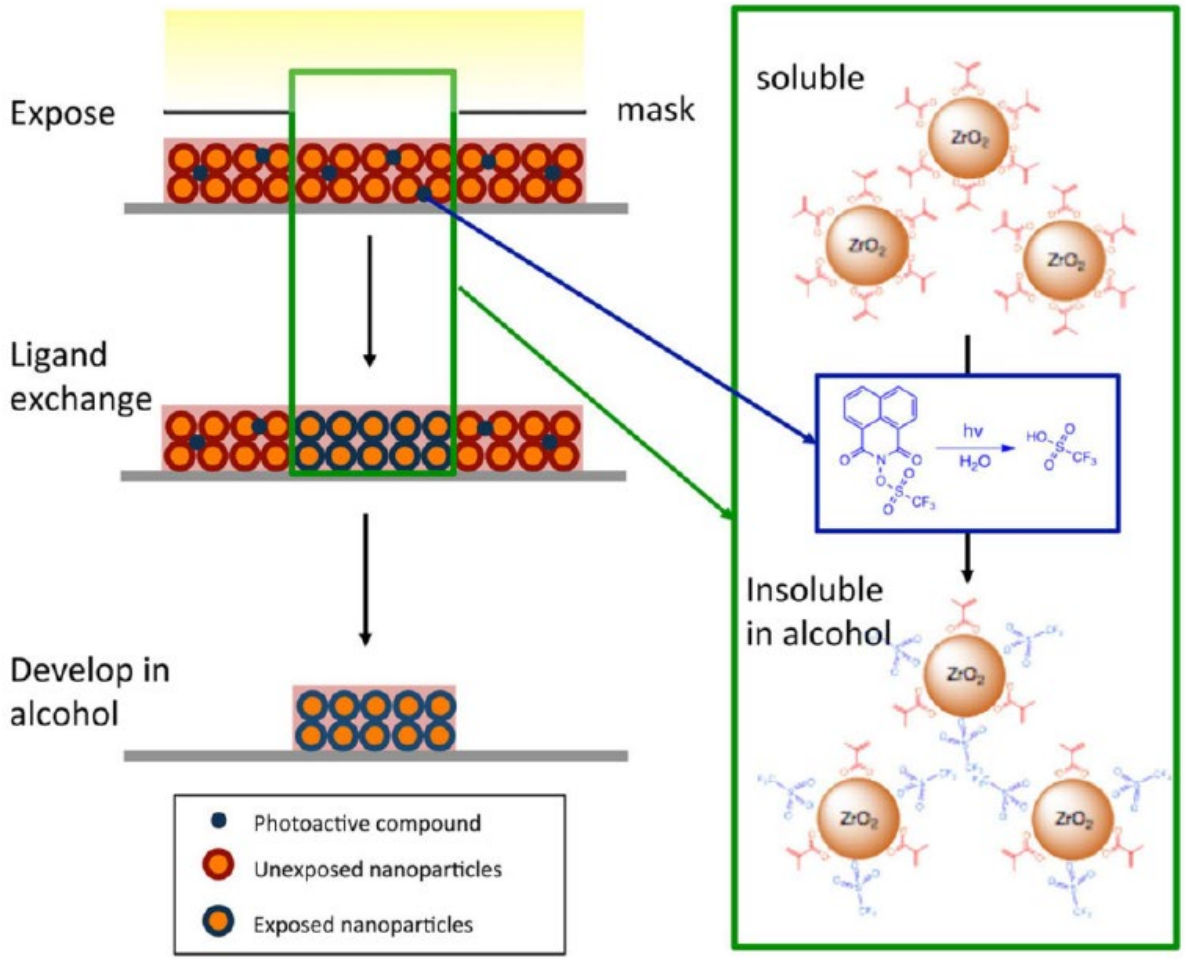
59. First, as I explained in my first declaration, Trikeriotis discloses that the "absorption coefficient of the films depends on the ligands." EX1024, 18. As I described above, the absorption coefficient (also called the extinction coefficient) is an indication of the radiation sensitivity of the material. *Supra,* ¶ 53; EX1035, 679. Accordingly, because the absorption coefficients is dependent upon the ligand, a POSA would understand that the ligand is radiation sensitive.

60. Second, Trikeriotis disclosed that "different ligands on the surface of the nanoparticles give them unique properties, allowing these films to act as positive or negative tone photoresists." EX1024, 15. This indicates that the ligands are radiation sensitive because if the mechanism of photoactivity (*i.e.*, sensitivity to radiation) were not dependent on the selection of the ligand, *any* ligand could be selected. Trikeriotis, however, explains that you can change the photoactivity by changing the ligands, which indicates that the ligands are sensitive to radiation.

61. Third, I understand that Dr. Banerjee points to Ober (EX1027) as evidence that the mechanism of photoactivity relates to a photoactive generator. However, Figure 1 of Ober shows that the **organic ligand shell is susceptible to "photochemical reactions,"** which means that it is "radiation sensitive":

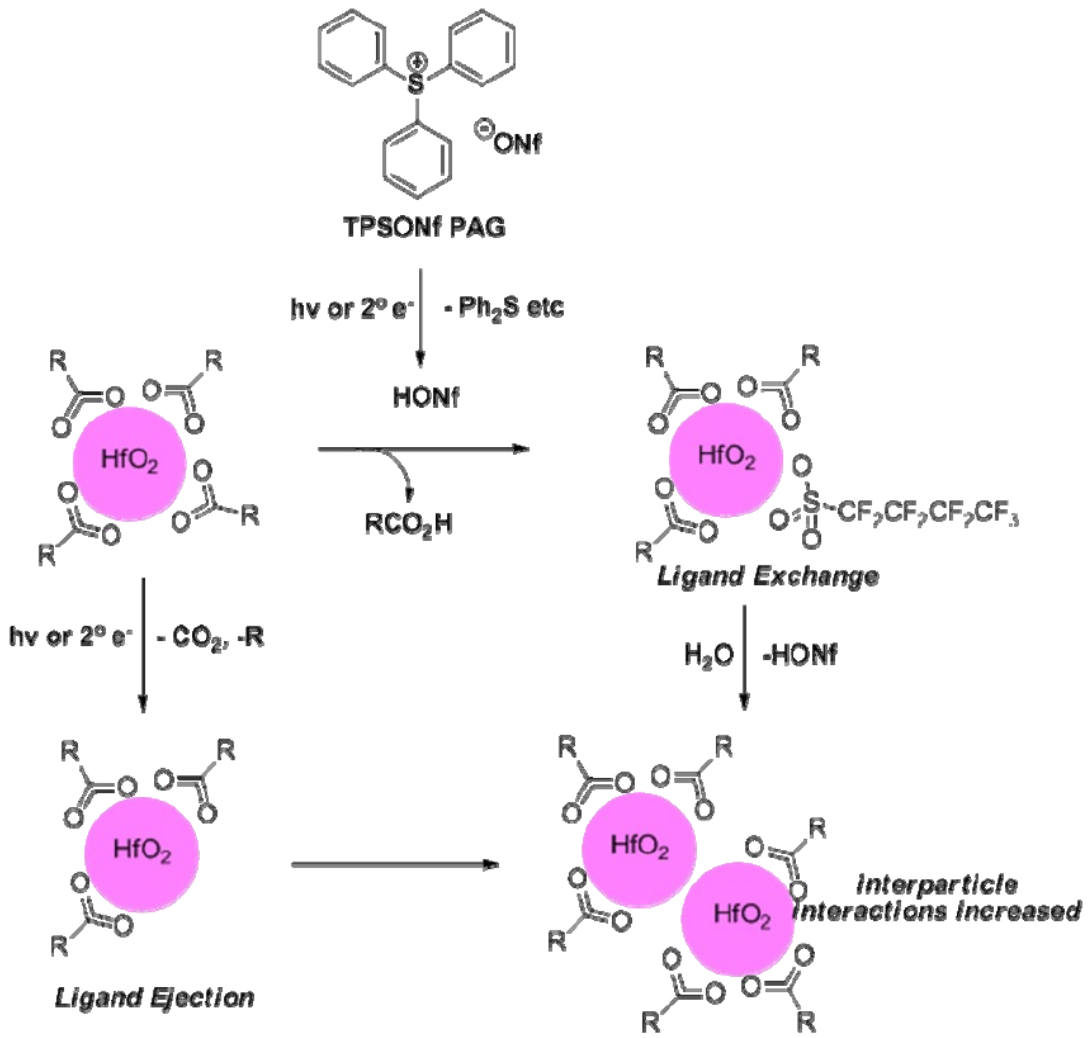


EX1027, 1 (Fig. 1, annotation in blue added). While Ober shows that photoactive compounds can be added, those too are **radiation sensitive ligands**:



EX1027, 2 (Fig. 2). As shown in Figure 2, photoactive compounds (shown in blue) are cleaved after exposure to radiation. *Id.* A ligand exchange occurs where the red ligands are displaced by the blue ligands. *Id.* Ober describes the red ligand as the "starting ligand" and the blue ligand as the "displacing ligand." *Id.*, 2. Accordingly, as Dr. Banerjee admits, *both* the red compound and blue photoactive compound are "ligands." EX1033, 181:19-182:7, 183:15-20. Thus, even under Dr. Banerjee's theory where only the photoactive compound is sensitive to radiation, the Trikeriotis coating materials comprise "radiation sensitive ligands."

62. However, even where a photoactive compound is present, both the red "starting ligand" and the blue "displacing ligand" are "radiation sensitive" as confirmed by later publications. EX1036, 5-6 (describing that the 'ligand exchange' method described in Ober "occur[s] simultaneously" with a mechanism that is based on radiation induced cleavage of the non-photoactive compound ligand). For example, Krysak showed that the Trikeriotis coatings can be patterned "with and without" a photoactive compound (which Krysak refers to as a photoacid generator, or "PAG"). *Id.* at 5. Krysak explains that the "PAG-free mechanism" works by "ligand ejection by photo decarboxylation." *Id.* That is, the carboxylic group of the organic ligand of the Trikeriotis coatings (without PAG) is cleaved by exposure to radiation which removes the ligand from the nanoparticle. This is shown in Krysak Figure 7:



Id. (Fig. 7). The "PAG-free" mechanism is shown on the left half of Figure 7 and the PAG "ligand exchange" method is sown on the right half of Figure 7. Krysak explains that even when PAG is present, both mechanisms take place simultaneously. *Id.*, 5. Accordingly, the organic ligands of Trikeriotis (i.e., the red "starting ligands" shown in Ober (EX1027)) are "radiation sensitive ligands."

D. Exposure Alters the Chemical Properties of the Prior Art Coatings

63. It remains my opinion that e-beam, UV, or EUV exposure of the Stowers 2008 coatings and the Trikeriotis coatings alters the chemical properties of those coatings which creates

differential dissolution rates between the exposed and un-exposed regions of the coating material. I understand that Dr. Banerjee opined that "UV or EUV radiation could alter a physical property of a material as opposed to a chemical property." EX2009 ¶¶40-41. I disagree.

64. Dr. Banerjee cites only a single reference for his opinion that a UV or EUV could alter a physical property—a 2007 patent that includes only a single sentence in the background section that allegedly supports his opinion: "The chemical or physical properties of the photoresist can be changed by being exposed to light." EX2006, 1:18-20. Dr. Banerjee did not identify even a single example where UV or EUV exposure changed only the physical properties of a photoresist. EX2009, ¶¶40-41. I was also unable to identify such an example. Dr. Banerjee stated during his deposition that he did not even "analyze what types of physical properties could change due to exposure to EUV or UV radiation." EX1033, 146:22-145:3.

65. Even the sole reference cited by Dr. Banerjee does not show an example of a physical change caused by UV or EUV exposure—it discloses a **chemical change** to the photoresist. EX2006, 1:53-63. Rather, it discloses the use of PMMA as a photoresist which, upon exposure to e-beam (i.e., neither UV or EUV radiation) is "cleaved into smaller fragments" and the "different solubility of the PMMA and the fragments formed from the PMMA by exposure in a s solvent is utilized for developing the photoresist." *Id.* Cleavage of the PMMA into fragments is a **chemical change**. Thus, there is no evidence that UV or EUV exposure could alter a physical property of a coating.

66. Dr. Banerjee also opines that "chemical changes that result in different dissolution rates can be driven by aspects of the lithography process besides the actual exposure to light/radiation," such as a post-exposure bake. EX2009, ¶¶42-43. Dr. Banerjee's logic is

unsound—if a baking step alone was sufficient to create a chemical change resulting in differential dissolution rates, an exposure step would be unnecessary. Indeed, Dr. Banerjee admitted during deposition that, in his hypothetical scenario where a post-exposure bake drives the "chemical changes that result in different dissolution rates," the exposure and post-exposure step "have to act in concert." EX1033, 139:20-140:7.

67. Dr. Banerjee cites publications describing chemically amplified resists which use a post-exposure bake to complete a chemical reaction that causes the difference in dissolution rates to support his opinion that a post-exposure bake can drive the "chemical changes that result in different dissolution rates." EX2009, ¶¶42-43; EX2005, 1:65-2:3; EX2007, 1. None of these references discuss the metal oxide photoresist of the '564 patent or any other metal oxide photoresists. Nevertheless, these articles show that irradiation causes a chemical change that is necessary to create the different dissolution rates. For example, an article by Moon & Kim describes that "**upon irradiation**, acids photogenerated from a PAG catalytically cleaves t-BOC groups during postexposure bake (PEB) at high temperatures." EX2007, 7. Thus, it is clear that the mechanism described in Moon & Kim **depends** "**upon irradiation**."

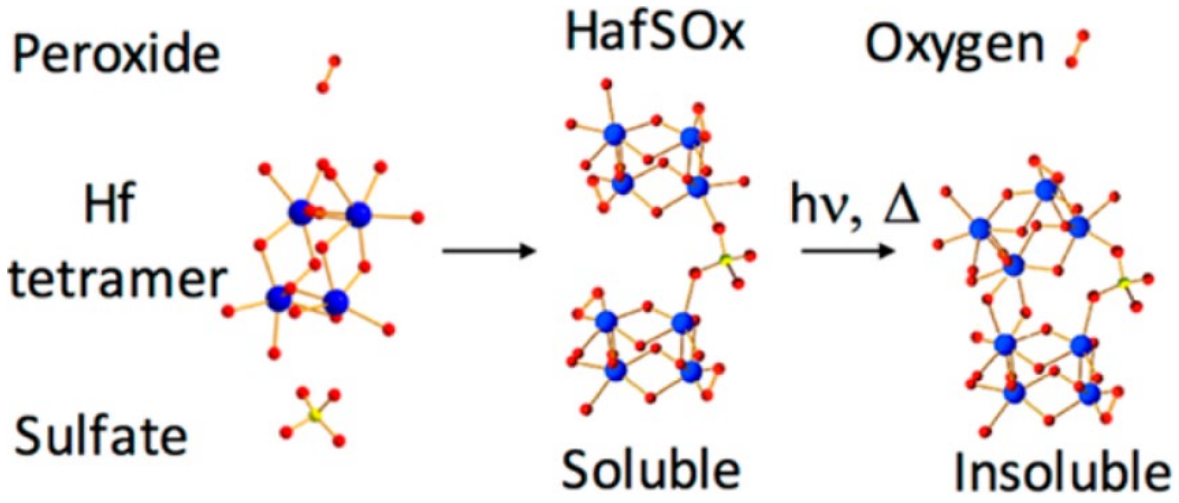
68. Moreover, as I describe below, Dr. Banerjee ignores publications demonstrating that the e-beam, UV, or EUV exposure of the Stowers 2008 and Trikeriotis coatings causes a **chemical change** which, in turn, cause different dissolution rates between the exposed and unexposed regions of the coating.

1. Exposure of Stowers 2008 Coating Alters Its Chemical Properties . . . Creating Different Dissolution Rates

69. As I described above, the Stowers 2008 coatings are the same in all material aspect to the '564 patent coatings. *Supra* § V.B. The '564 patent describes that the exposure to radiation

(i.e. e-beam, UV, or EUV exposure) "converts the irradiated coating material into a condensed material that is resistant to removal with a developer composition." EX1001, 4:3-6. Thus, exposure alters the chemical properties of the coating (i.e., causes condensation of the coating material), which is then less soluble than the unexposed regions (i.e., the exposed region is "resistant to removal" with a developing solution). It follows, therefore, that exposure of the Stowers 2008 coatings also alters its chemical properties creating different dissolution rates between the exposed and unexposed regions of the coating.

70. Dr. Banerjee also ignores later publications by Dr. Stowers showing how the material is chemically altered. For example, in 2018 Dr. Stowers explained that "[b]ecause regions of HafSOx exposed to radiation become insoluble in the subsequent pattern development step . . ., it may be deduced that the decomposition of Hf−peroxide bonds leads to the formation of a Hf−oxide network, which results in the insoluble HafSOx in exposed regions." EX1019, 5. This is depicted in the abstract figure:



Id., 1. As can be seen in this figure, exposure to UV/EUV/e-beam breaks the peroxide bond which induces condensation of the HafSOx material (*i.e.*, chemically alters the material) and the

condensed (exposed) material is insoluble (*i.e.*, has a different dissolution rate than the unexposed regions of the coating).

71. Accordingly, it is my opinion that exposure of the Stowers 2008 coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material.

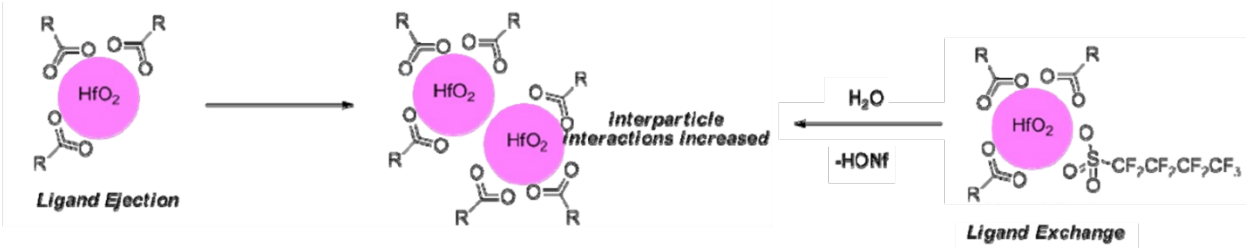
2. Exposure of Trikeriotis Coating Alters Its Chemical Properties . . . Creating Different Dissolution Rates

72. Trikeriotis explains that "different ligands on the surface of the nanoparticles give them unique properties, allowing these films to act as positive or negative tone photoresists." EX1024, 15. A POSA understands that in a positive resist, a development solution removes the exposed portion because it is **more soluble** in the development solution than he unexposed portion. Conversely, in a negative resist, a development solution removes the <u>un</u>exposed portion because it is **more soluble** in the development solution than he unexposed portion. Thus, because the Trikeriotis coatings can act as positive or negative tone resists, exposure to EUV, UV, or e-beam necessarily causes chemical changes that changes the solubility of the exposed region to make it more or less soluble than the unexposed regions (i.e. creates differential dissolution rates between exposed and un-exposed regions of the coating material).

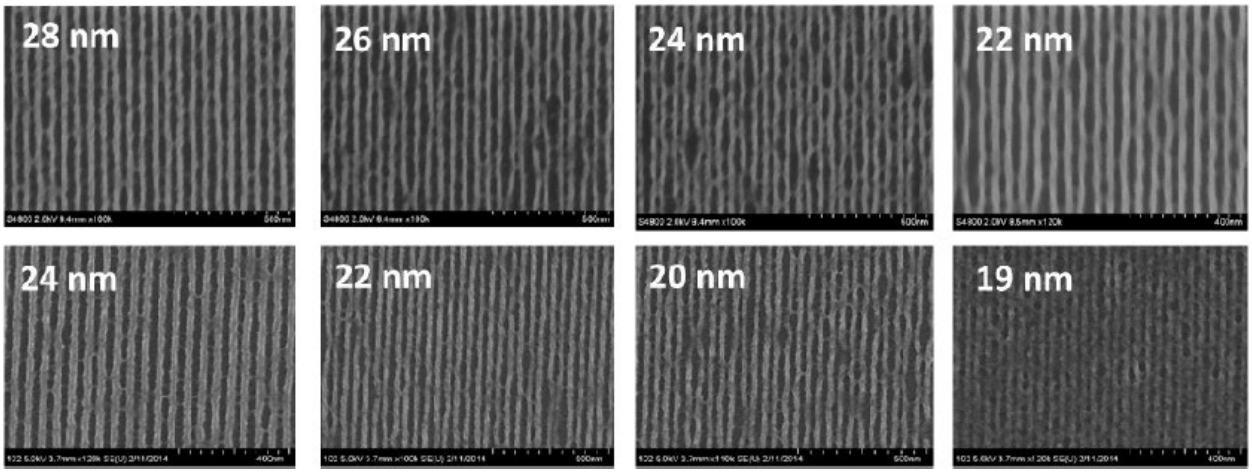
73. Dr. Banerjee also ignores later publications showing the photoresist mechanism. For example, Ober, which Dr. Banerjee discusses at length in his declaration, explains that exposure "creates a solubility change in exposed areas." EX1027, 1. As I explained above, Ober shows a ligand exchange mechanism where a starting ligand is displaced with a displacing ligand caused by exposure to radiation. *Supra* § V.C.2. Ober explains that the "ligand displacement is

associated with a solubility change." EX1027, 2. Thus, a chemical change (i.e., ligand displacement) cause a change in solubility to the exposed portions of the coating (i.e. differential dissolution rates between exposed and un-exposed regions of the coating material).

74. Moreover, Krysak, which explains the PAG-free mechanism of the Trikeriotis photoresists (discussed *supra* § V.C.2) shows the mechanism of differential solubility. Figure 7 shows that upon either the radiation-induced "ligand ejection" mechanism (i.e. the PAG-free chemical change) or the ligand exchange mechanism (i.e., the PAG-dependent chemical change), "interparticle interactions increase[]":



EX1036, 6 (adapted from Fig. 7). Coatings with *and* without PAG were patterned with EUV:



Id., 5 (Fig. 6, showing "SEM images of EUV patterned HfO_2-MAA/BZA with PAG (top row) and without PAG (bottom row), dose 38 mJ"). This shows that the exposed and unexposed portions of the coating have different solubilities.

75. Accordingly, it is my opinion that exposure of the Trikeriotis coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material.

E. The Prior Art Metal Oxide Coatings Have High Absorption Relative to Conventional Radiation Based Resists

76. It remains my opinion that the Stowers 2008 and Trikeriotis coatings have high absorption relative to conventional radiation based resists. I understand that Dr. Banerjee opines that the linear trend shown between density and absorption coefficient does not show that "higher density necessarily means higher EUV absorption." EX2009, ¶¶ 48-51. Dr. Banerjee makes leaps in logic and ignores data in other publications to arrive at this opinion.

77. First, as I explained in my first declaration, a POSA would have known that as the density increases, the EUV absorbance increases. Indeed, the trend is generally linear, with the EUV absorption coefficient becoming larger as the density increases. EX1002, ¶¶ 136-137. Dr. Banerjee takes issue with the outliers shown on the graph of EX1025, where some materials have higher density and lower calculated linear absorption coefficients than PMMA:

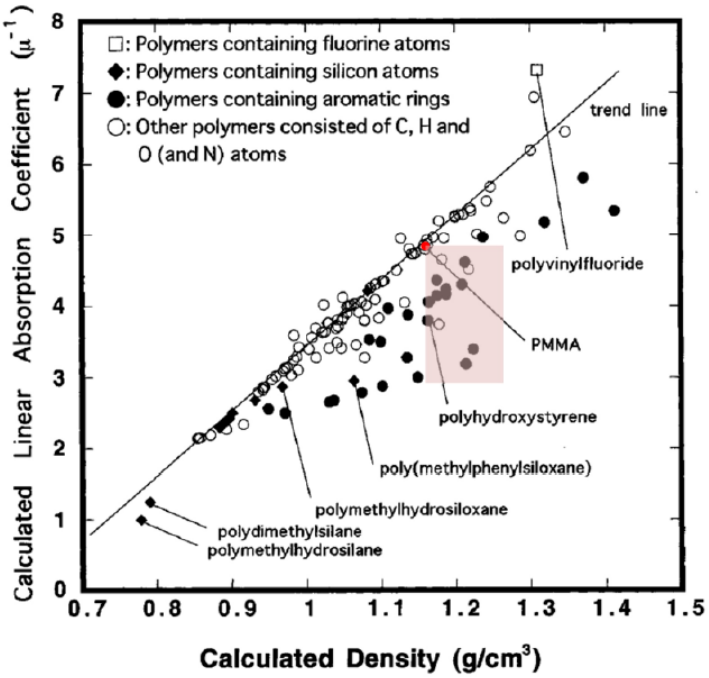
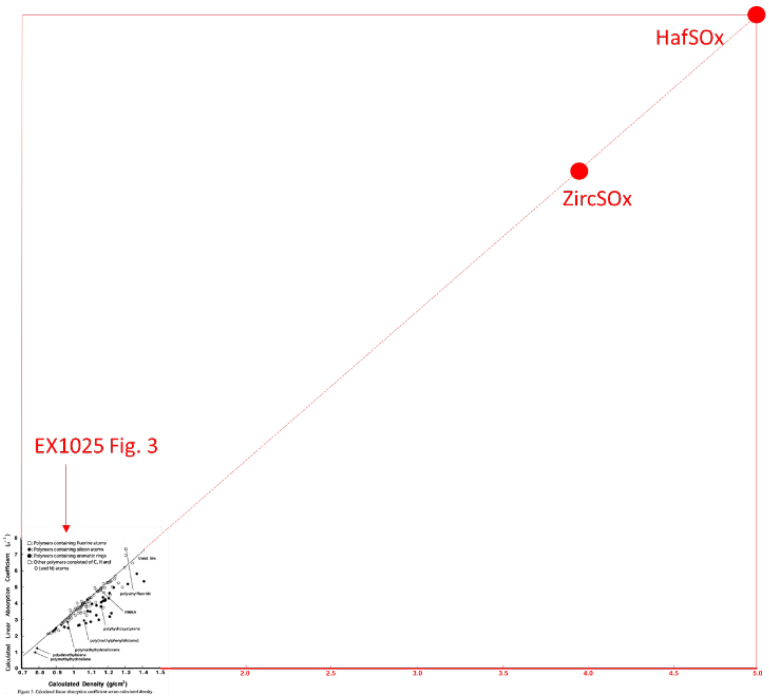


Figure 3. Calculated linear absorption coefficient versus calculated density.

EX2009, ¶ 50 (citing EX1025, 4 with Dr. Banerjee's "red annotation added to show PMMA datapoint, and pink annotation added to show data points with higher density and lower calculated linear absorption coefficient"). Dr. Banerjee cherry picks these alleged outliers, without considering the context of the metal oxide resists. For example, Dr. Stowers stated that the "because of the **metal content**," (which is present in the coatings of Stowers 2008 and Trikeriotis), "ZircSOx and particularly HafSOx have a higher density than other resists like HSQ (1.4 g/cm^3) or PMMA (1.19 g/cm^3)." EX1008, 90. Stowers explained that a "reasonable density of 4 g/cm^3 for ZircSOx and 5 g/cm^3 for HafSOx can be assumed." *Id.* Thus, the calculated density would be 3.3 to 4.2 times higher than PMMA and, in the context of Figure 3 of EX1025, would be off the x-axis of the chart. Instead, Dr. Banerjee focuses on materials that have a *similar density*. If we were to extend the x- and y-axis to include densities on the order of 4 to 5

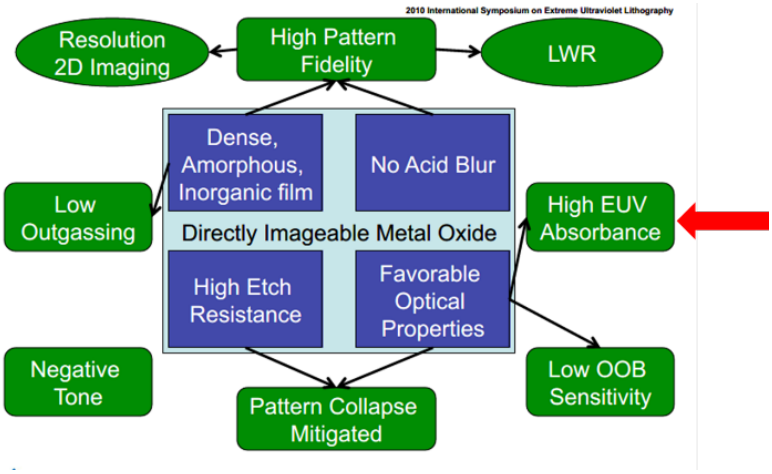
gm/cm^3, the metal oxide materials of Stowers 2008 and Trikeriotis would have a significantly higher calculated linear absorption:



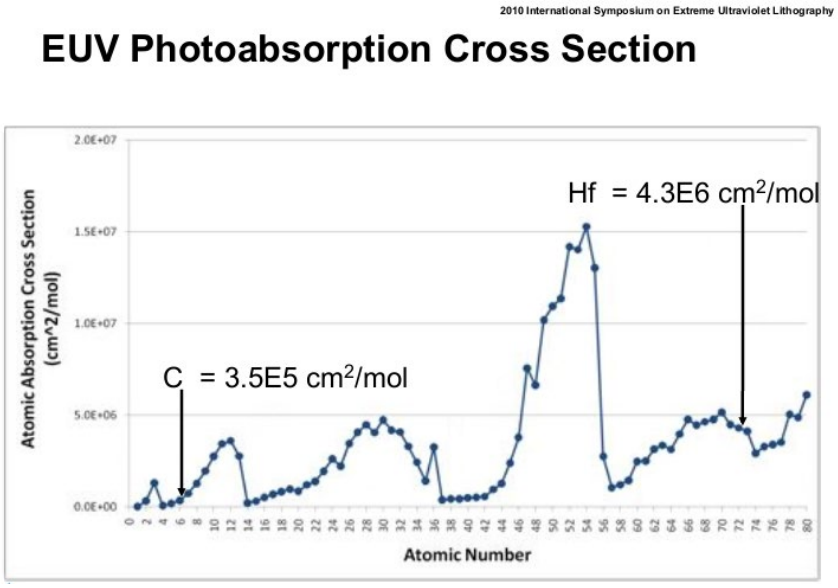
EX1025, 4 (adapted Figure 3, with red extensions to x-axis, y-axis, and trend line, and red dots showing location of 4 g/cm^3 and 5 g/cm^3 on the trend line). Indeed, when asked where HafSOx and ZircSOx would appear on the graph of EX1025 Figure 3, Dr. Banerjee pointed to a location of the chart on both the x- and y-axis. EX1033, 179:8-180:6.

78. This trend is confirmed by Dr. Stowers' later publication, which shows that a typical polyhydroxystyrene ("PHS") resist which has a density of 1.19 g/cm^3 while hafnium photoresists have a calculated density of 4.7 g/cm^3— ~ 4-fold increase. EX1022, 3. The hafnium photoresists had a corresponding ~4-fold increase in EUV absorption compared to the PHS resists: 4.1 µm^{-1} (for PHS photoresists) compared to 16 µm^{-1} (for hafnium photoresists). *Id*.

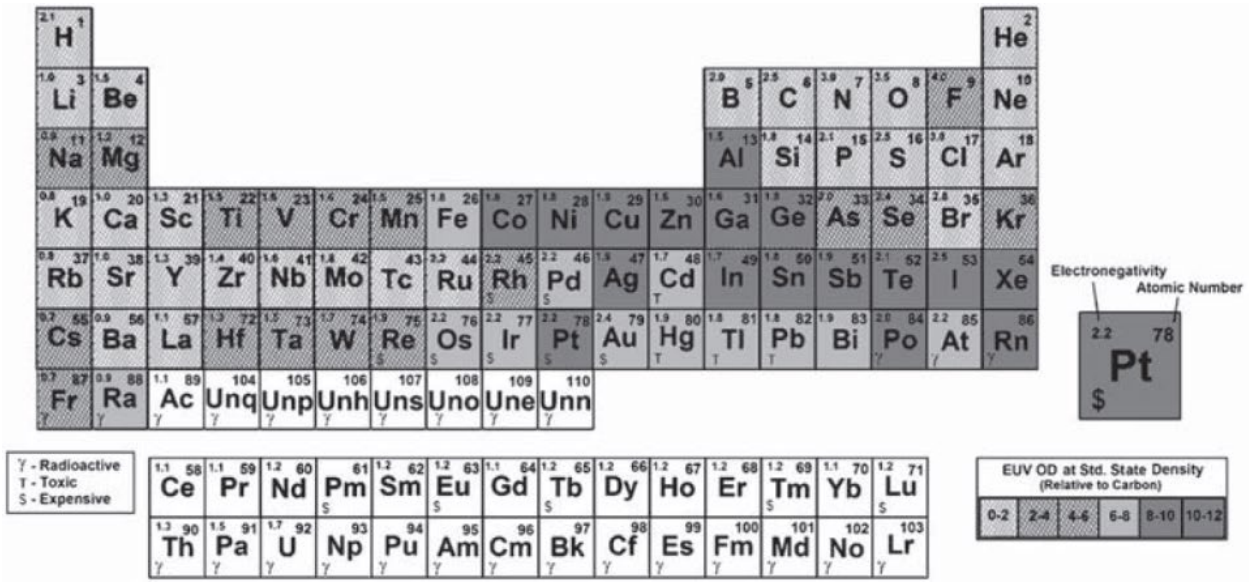
79. This makes sense, because as Dr. Stowers, and others, have recognized, metals have a higher EUV absorbance than carbon, and therefore **metal** oxide photoresists (such as those of Stowers 2008 and Trikeriotis) will necessarily have higher EUV absorbance than carbon-based photoresists. EX1044, 2 ("Absorption of EUV light in matter depends entirely on the atomic composition and the absorption cross section of elements."). For example, in a 2010 presentation, Dr. Stowers characterized metal oxide photoresists as "high EUV absorbance":



EX 1013, 3. In that same presentation, Dr. Stowers showed that the EUV absorption of Hafnium is significantly higher than that of carbon:



Id., 12. Others have also recognized that metals, such as hafnium, have a higher EUV absorbance than carbon:



EX1037, 5.

80. It is therefore my opinion that the Stowers 2008 and Trikeriotis coatings have high absorption relative to conventional radiation-based resists.

F. The Prior Art Metal Oxide Coatings Can Be Effectively Patterned at a Dose of No More Than 100 mJ/cm^2

1. The Stowers 2008 Coatings Can Be Effectively Patterned at a Dose of No More Than 100 mJ/cm^2

81. It remains my opinion that the Stowers 2008 coatings can be effectively patterned at a dose of no more than 100 mJ/cm^2.

82. As I described above, the Stowers 2008 coatings are the same in all material aspects to the ’564 patent coatings. *Supra* § V.B. The ’564 patent shows that the coatings can be patterned at a dose of 20-80 mJ/cm^2 (i.e., less than 100 mJ/cm^2). EX1001, 22:51-54, 23:37-38. It follows, therefore, that exposure of the Stowers 2008 coatings also can be effectively patterned at a dose of no more than 100 mJ/cm^2.

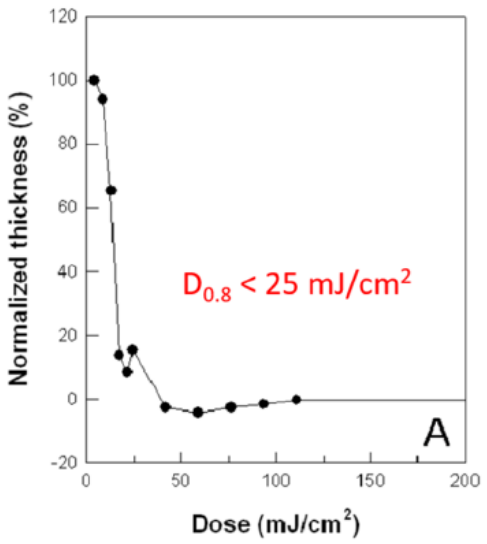
83. Additionally, as I explained in my previous declaration, Stowers 2008 shows that ZircSOx films could be patterned at a dose of 112 μC/cm^2. EX1002, ¶145 (citing EX1008, 89). In a later publication, Dr. Stowers explained that an e-beam dose of 249 μC/cm^2 for MSOx corresponds to an EUV dose of 25 mJ/cm^2. EX1022, 7. I understand that Dr. Banerjee opines that this correlation only applies to HafSOx material, while the dose I cited in Stowers 2008 was for ZircSOx material. EX2009, ¶¶52-54. I disagree. EX1022 generally relates to **M**SOx materials, where the “**M**” stands for metal, including zirconium <u>and</u> hafnium. EX1022, 3 (“We start with a metal oxide sulfate (MSOx) and introduce a peroxo complexing agent.”). Thus, the correlation e-beam to EUV dose of EX1022 relates to *either* HafSOX *or* ZircSOx. Moreover, the ’564 patent provides a similar correlation. EX1001, 2:62-63 (a dose of about 100 mJ/cm^2 or 300 μC/cm^2.”).

84. Even if Dr. Banerjee is correct (which he is not), Stowers 2008 also shows HafSOx films that were patterned at a dose of 60 μC/cm^2 or 300 μC/cm^2. EX1008, 99-100 (Figures 4-3 and

4-4). Thus, even under Dr. Banerjee's incorrect interpretation of EX1022, the Stowers 2008 coatings can be effectively patterned at a dose of no more than 100 mJ/cm^2.

2. The Trikeriotis Coatings Can Be Effectively Patterned at a Dose of No More Than 100 mJ/cm^2

85. Additionally, as I explained in my first declaration, shown in Figures 5 and 6 of Trikeriotis 2010, the Hf nanoparticle films can clearly be patterned in positive tone with DUV radiation dose of less than 100 mJ/cm^2. Figure 5A shows a sensitivity of less than 25 mJ/cm^2:



EX1024, Fig. 5A (positive tone contrast curve of the nanoparticle films under 254 nm UV light exposure; $D_{0.8}$ sensitivity notation added).

86. As can be seen in Trikeriotis Figure 5A, at a dose well under 100 mJ/cm^2, the exposed regions have been removed by the developer (*i.e.*, a normalized thickness close to 0%). I added the notation of $D_{0.8}$ < 25 mJ/cm^2 to show the dose required for 80% of the exposed film to be soluble (for a positive tone resist) or insoluble (for a negative tone resist). *See, e.g.*, EX1037, 43; EX1046, 1; EX1047, 186. In Trikeriotis Figure 5A, at a dose of less than 25 mJ/cm^2, the

normalized thickness is less than 20% (*i.e.*, more than 80% of the exposed film was soluble), and therefore the $D_{0.8}$ of the Trikeriotis film is 25 mJ/cm^2.

87. Accordingly, the Trikeriotis coatings can be effectively patterned at a dose of no more than 100 mJ/cm^2.

VI. OPINIONS IN SUPPORT OF PETITIONER'S OPPOSITION TO PATENT OWNER'S MOTION TO AMEND

A. New Ground 5: Proposed Amended Claim 13-18 And 20-23 Are Anticipated by Keszler (EX1038)

1. Keszler (EX1038)

88. WO2008/082448 ("Keszler") was filed on October 31, 2007 and published on July 10, 2008. I understand that Keszler is therefore prior art to the '564 patent regardless of the priority date.

89. As discussed below, proposed amended claims 13-18 and 20-23 are anticipated by Keszler.

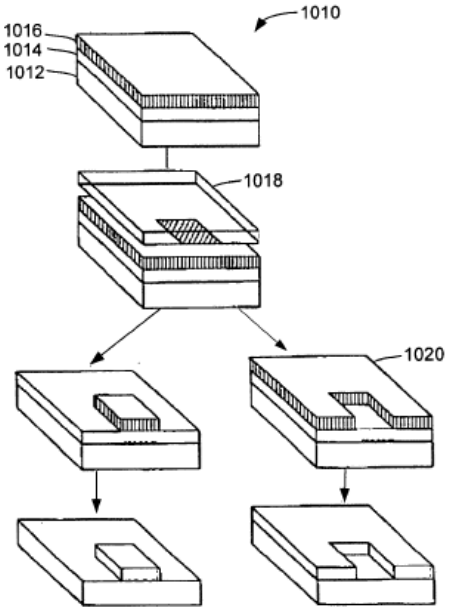
2. Proposed Independent Claim 13

a. Preamble: "structure comprising a substrate and a coating material on a surface of the substrate"

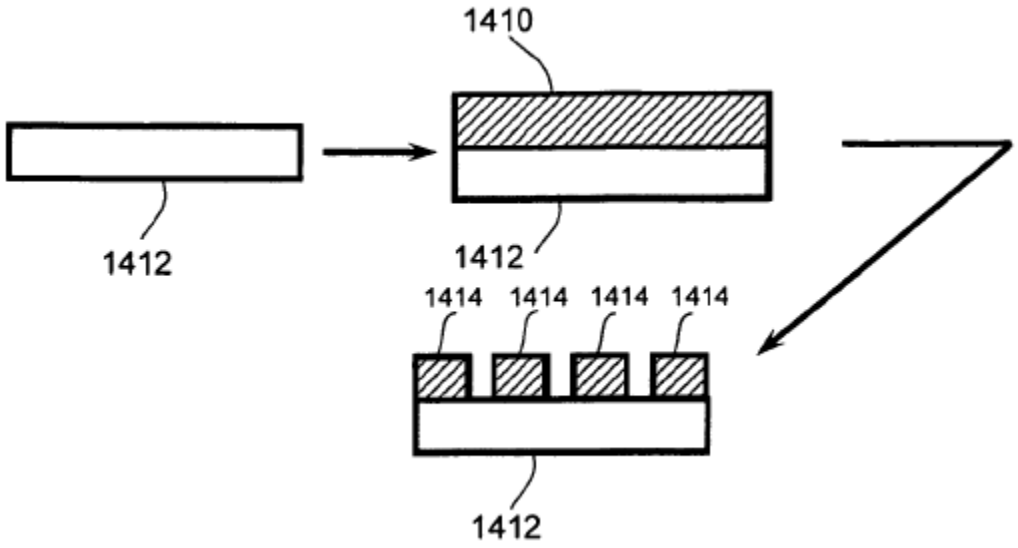
90. Keszler discloses "a method for patterning a device using a photoresist" by "providing a structure" which comprises a "resist layer." EX1038, 4:16-25. For example, Keszler example 4, discloses a method for "producing smooth, dense, and pinhole-free ultra-thin nanolaminate films as thin as about 2 nm" using "precursor solutions" that are deposited on "silica substrates with thermally grown SiO_2" via spin coating "with a spin rate of 3000 rpm for 30 seconds." *Id.*, 40:8-19. Similarly, Keszler example 7 discloses method where a precursor solution

containing peroxide (*e.g.*, the precursor solutions of example 4) is "spin-cast, liquid misted, or ink-jet printed onto an appropriate substrate." *Id.*, 42:8-10.

91. Keszler provides schematic drawings of these processes where the film (*i.e.*, the "coating") is on a "substrate." For example, Figure 10 "is a schematic diagram illustrating positive or negative photoresist procedures" *Id.*, 10:21-22.



Id., 57 (Fig. 10). As is seen in figure 10, the substrate comprises (1) a silicon substrate (labeled 1012) with a silica layer (labeled 1014). *Id.* 28:15-17. The photoresist coating (labeled 1016) is applied to the substrate. *Id.* Similarly, Keszler Figure 14 "is a schematic diagram illustrating a single layer process for producing a patterned film on a substrate." *Id.*, 10:30-31.



Id., 59 (Fig. 14). In Figure 14, a "thin film" (labeled as 1410) is deposited on to a substrate (labeled 1412). A POSA would understand that the films deposited on the substrate are "coatings," as shown in Keszler Figures 10 and 14.

92. Keszler therefore discloses "structure comprising a substrate and a coating material on a surface of the substrate."

b. "coating material comprises metal ions with radiation sensitive ligands"

93. Keszler discloses that the photoresist comprises "**hafnium** and/or **zirconium** oxy hydroxy compounds." *Id.*, 4:16-25. Keszler provides such a "**hafnium** and/or **zirconium** oxy hydroxy compound" in Example 4:

> Precursor solutions containing $HfOCl_2 \cdot 8H2O$ or $ZrOCl_2 \cdot 8H2O$, H_2O_2 (30%) and H_2SO_4(aq) were prepared . . .. The precursor solutions contained a metal ion concentration of about 0.05 M to 0.4 M with 70% H_2O_2 and 50% sulfate relative to metal. The difference in **metal ion** concentrations allowed for thickness per coat variation from 3 nm to 25 nm."

Id., 38:6-63. Here, the precursor solutions comprise the hafnium or zirconium **metals**, which Keszler describes as a "metal ion." *Id.*

94. Indeed, the precursor solutions are aqueous solutions comprising $HfOCl_2 \cdot 8H2O$ or $ZrOCl_2 \cdot 8H2O$, which a POSA would understand to exist in solution as HfO^{+2} and ZrO^{+2} cations, respectively. Consistent with this understanding, Keszler explains that "that "the metal-containing cation may be provided using an aqueous solution of a hafnium oxy halide, such as hafnium oxy chloride ($HfOCl_2$ (aq))." *Id.*, 12:34-13:1.

95. These precursor solutions are then "applied to a substrate using any suitable technique, including printing, spin, spray, or dip-coating to form a patterned or blanket liquid film" *Id.* 26:14-16. In example 4, the precursor solutions were applied to the substrate "using a spin-coater with a spin rate of 3000 rpm for 30 seconds." *Id.*, 40:15-18. Since those precursor solutions in Keszler are spin-coated onto wafers to make films, the resulting films will necessarily also contain those same metal ions.

96. As shown above, the precursor solutions comprise peroxide in addition to the hafnium or zirconium ion. *Id.*, 38:6-63. Keszler discloses that "peroxide" (i.e. H_2O_2) is a radiation sensitive ligand. For example, Keszler explains that:

> A small coordinating species such as **peroxide** ($O2^{2-}$) can be incorporated into aqueous-based inorganic liquid suspensions and films to provide an additional, **new lithographic patterning method**. In this technique, energy (e.g., light, electron beam, heat) can be applied to films, inducing condensation reactions that alter the solubility of the film, thereby allowing lithographic patterning.

Id., 28:9-12. Keszler's explanation that the peroxide provides a "new lithographic patterning method" indicates that the peroxide is radiation sensitive because it imparts the ability for patterning via lithography (i.e., it is what allows a resist material to be patterned with radiation).

97. Moreover, the precursor solutions of Keszler Example 4 contains the same components as the precursor solutions used to make the coating materials in the Examples of the '564 patent and therefore necessarily have the same properties. Specifically, Keszler the '564 patent created precursor solutions having the following concentrations:

	'564 Patent (Hf Precursor Solution)	**'564 Patent (Zr Precursor Solution)**	**Keszler**
Metal Concentration	0.15 M	0.16 M	0.05 – 0.4 M
Peroxide Concentration	0.99 M – 1.32 M	0.11 M – 0.14 M	0.035 – 0.28 M [1]
Sulfate Concentration	0.12 M – 0.3 M	0.14 M – 0.36 M	0.025 – 0.2 M [2]
Peroxide : Metal	6.6 – 8.8 : 1	0.66 – 0.88 : 1	0.7 : 1
Sulfate : Metal	0.8 – 2 : 1	0.9-2.25 : 1	0.5 : 1

EX1038, 38:6-63; EX1002 ¶ 38 (using the corrected calculation noted during my July 18, 2024 deposition (EX2011, 132:6-124:12); *see also* EX1001, 21:10-67; POR, 34.

[1] The precursor solutions of Keszler Example 4 comprise 0.05 M to 0.4 M metal, with a 70% peroxide content on the basis of metal. EX1038, 38:6-63.

[2] The precursor solutions of Keszler Example 4 comprise 0.05 M to 0.4 M metal, with a 50% sulfate concentration on the basis of metal. *Id.*

98. Any differences in the final concentrations and ratios between the precursor solutions in Keszler in and the ’564 patent are immaterial. This is confirmed by publications by Drs. Stowers and Keszler (inventors of both Keszler and the ’564 patent). For example, EX1019 explains the role of peroxide in the HafSOx and ZircSOx. The authors found that for peroxide to metal ratios less than 0.5 “all of the peroxide coordinates to the [hafnium] clusters.” *Id.* However, when the ratio “exceeds 0.5, the free peroxide stretching mode . . . appears while the bound peroxide saturates in intensity” which “suggests that approximately two peroxide groups coordinate to each hafnium tetramer.” *Id.* Here, the ratio of peroxide to metal in both Keszler and the ’564 patent exceeds 0.5, indicating that the films of the ’564 patent examples would have the same properties as the films of Keszler. *See also supra*, ¶¶ 44-47. EX1019 also tested various ratios of sulfate : metal and found that “[a]ll films made with a ratio of sulfate to hafnium <2 exhibit increased oxide network formation with increased X-ray exposure.” EX1019, 5. Here, the ratio of sulfate to metal ratio in both Keszler and the ’564 patent is less than 2, indicating that the films of the ’564 patent examples would have the same properties as the films of Keszler.

99. Because the films of the ’564 patent are the same in all material aspects as the films of Keszler, they necessarily have the same properties. As the ’564 patent discloses, the peroxide ligands are “radiation sensitive.” For example, the ’564 patent explains that “upon absorption of the radiation, the peroxide functional groups are fragmented” and that “energy absorbed is transferred to the peroxide ligand which can result in the rupturing of the peroxide bond.” EX1001, 7:62-66; 11:39-42. The peroxide ligands of the Keszler films are therefore also “radiation sensitive,” an inherent property of the material.

100. Later publications related to HafSOx and ZircSOx further confirm that the Keszler peroxide ligands are radiation sensitive. For example, in 2010, Dr. Stowers explained that "the mechanism that leads to negative tone patterning is dependent on the presence of peroxide," which is based on "beam-induced decomposition and elimination of peroxide [that] promotes condensation reactions, cross-linking of metal-oxo bridges, and the production of three-dimensional framework structures." EX 1010, 21. Similarly, in a 2018 publication, Dr. Stowers explained that in HafSOx "[p]eroxide serves as the **radiation sensitive** species" which "decompose[s] via radiative . . . energy." EX1019, 1.

101. Keszler therefore discloses a "coating material compris[ing] metal ions with radiation sensitive ligands."

c. "coating material has an average thickness of about 25 nm"

102. Keszler discloses depositing the precursor solutions as "a thin film (from about 1 nm to about 100 nm)." EX1038, 32:3-15. For example, the precursor solutions of Example 4 can produce "films as thin as about 2 nm." *Id.* 40:1-2.

103. Keszler Example 4, provides a specific example of depositing the coating material at thicknesses "from 3 to **25 nm**." *Id.* 40:13-14. The variation in thickness was achieved a by varying the concentration of the metal ion in the precursor solution, which Keszler disclosed was from 0.05 M to 0.4 M. *Id.,* 38:6-63, 40:13-14.

104. Keszler therefore discloses a "coating material [that] has an average thickness of about 25 nm."

d. "wherein exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material"

105. Keszler discloses that patterns are formed by changing the chemical structure of the resist layer using UV, EUV, or e-beam and that this chemical change causes the exposed region of the coating to have a different dissolution rate than the unexposed region of the coating. For example, Keszler explains that:

> Patterns can be formed using a positive or negative photoresist. This process is illustrated schematically in FIG. 10 with reference to a laminate structure 1010 having a silicon substrate 1012, a silica layer 1014 and a resist layer 1016. For positive resists, the resist layer 1016 is exposed to an energy source, such as UV light, wherever the underlying material is to be removed. In these resists, exposure to the UV light changes the chemical structure of the resist layer 1016 so that it becomes more soluble in the developer.

Id. 28:15-21. Keszler explains that this change in the "chemical structure of the resist layer" can be the condensation or crosslinking. *Id.* 8:1-12. This crosslinking will change the solubility of the exposed region, which creates "differential dissolution rates between exposed and un-exposed regions of the coating material."

106. Moreover, as I explained above, because the films of the '564 patent are the same in all material aspects as the films of Keszler, they necessarily have the same properties. The '564 patent also explains that exposure to UV, EUV and/or electron-beam radiation. For example, the '564 patent discloses that "exposure to the radiation converts the irradiated coating material into a

condensed material that is resistant to removal with a developer composition." EX1001, 4:3-6. The mechanism of this condensation occurs by breaking the peroxide bonds in the coating material:

> The absorption of the radiation results in transfer of energy that breaks the peroxide —O—O— bonds so that at least some of the peroxide-based ligands are no longer available to stabilize the material. With the absorption of a sufficient amount of radiation, the exposed coating material condenses.

Id., 15:37-42. Figure 1 of the '564 patent provides a schematic of this mechanism:

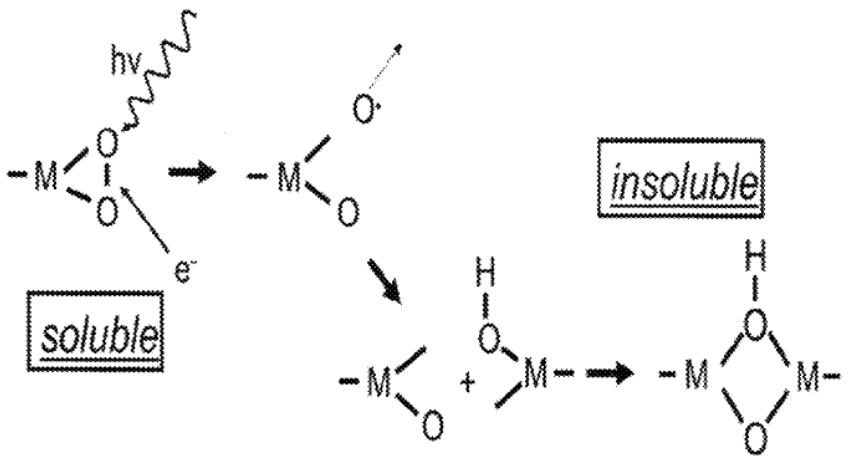
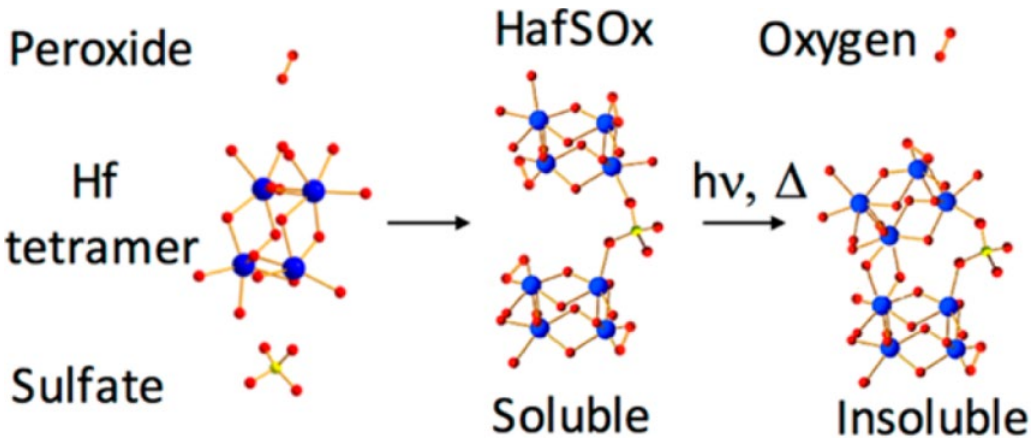


Fig. 1

Id., 3 (Fig. 1). Figure 1 shows "a metal cation bonded to a ligand, where M represents a metal atom and O represents an oxygen atom, undergoing a condensation reaction triggered by energy absorption from radiation." *Id.*, 3:6-9. Thus, the EUV, UV, or e-beam exposure breaks the peroxide bonds in the coating material causing the coating material to condense (*i.e.*, a chemical change). That change renders the exposed portion of the coating insoluble (*i.e.*, having a different solubility compared to the unexposed portions of the coating). Accordingly, EUV, UV, or e-beam exposure of the Keszler films will also cause the same condensation chemical change rendering the exposed portions of the coating insoluble.

107. This mechanism of action is further confirmed by later publications. For example, in a 2018 publication, Dr. Stowers explained that "[b]ecause regions of HafSOx exposed to radiation become insoluble in the subsequent pattern development step . . ., it may be deduced that the decomposition of Hf−peroxide bonds leads to the formation of a Hf−oxide network, which results in the insoluble HafSOx in exposed regions." EX1019, 5. This is depicted in the abstract figure:



Id., 1. As can be seen in this figure, exposure to UV/EUV/e-beam breaks the peroxide bond which induces condensation of the HafSOx material (*i.e.*, chemically alters the material) and the condensed (exposed) material is insoluble (*i.e.*, has a different dissolution rate than the unexposed regions of the coating).

108. Keszler therefore discloses that "exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material."

3. Proposed Claim 14: The structure of claim 13 wherein the metal ions comprise metal suboxide ions.

109. Keszler discloses that the "the metal containing ion may comprise a **metal suboxide**." EX1038, 13:20-21. As discussed above with respect to claim 13, the films of Keszler comprise **HfO^{+2}** or **ZrO^{+2}** which are metal suboxide ions. The '564 patent defines "metal suboxide cations" as "polyatomic cations with a metal element and covalently bonded oxygen atoms." EX1001, 5:47-49. The '564 patent explains that "[m]etal suboxide cations of particular interest include, for example, ZrO^{+2}. . ., [or] HfO^{+2}" confirming that the metal ions of Keszler are "metal suboxide cations."

110. Accordingly, Keszler discloses that "the metal ions comprise metal suboxide ions."

4. Proposed Claim 15: The structure of claim 13 wherein the metal ions comprise … Zr, … Hf, …

111. As discussed above with respect to claim 13, the films of Keszler include ZrO^{+2} or HfO^{+2} cations, in which the metal is Zr or Hf. Keszler therefore discloses this limitation.

5. Proposed Claim 16: The structure of claim 13 wherein the radiation sensitive ligands comprise peroxide ligands

112. As discussed above with respect to claim 13, the films of Keszler include peroxide ligands. Keszler therefore discloses this limitation.

6. Proposed Claim 17: The structure of claim 13 wherein the coating material further comprise polyatomic anions

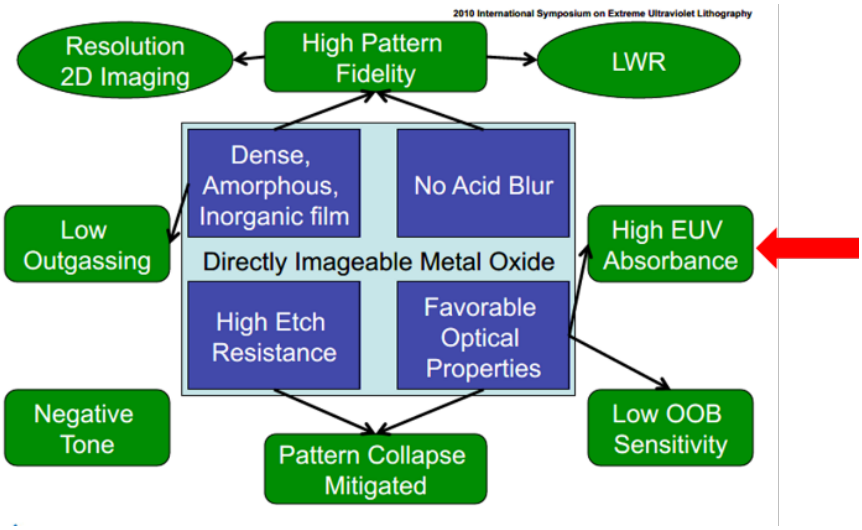
113. As described above, the Keszler films were prepared by spin coating a precursor solution that comprises "$HfOCl_2 \cdot 8H2O$ or $ZrOCl_2 \cdot 8H2O$, H_2O_2 (30%) and H_2SO_4 (aq)." EX1038, 38:6-63, 40:15-18. Keszler discloses that the "H_2SO_4 (aq)" is a polyatomic anion: "a polyatomic anion, such as sulfate (SO_4^{2-}), may be provided using an aqueous solution of sulfuric acid (H_2SO_4

(aq))." *Id.* 13:1-2. Accordingly, a POSA would understand that the H_2SO_4 exists in solution as SO_4^{2-}, which is a polyatomic anion. Keszler therefore discloses this limitation.

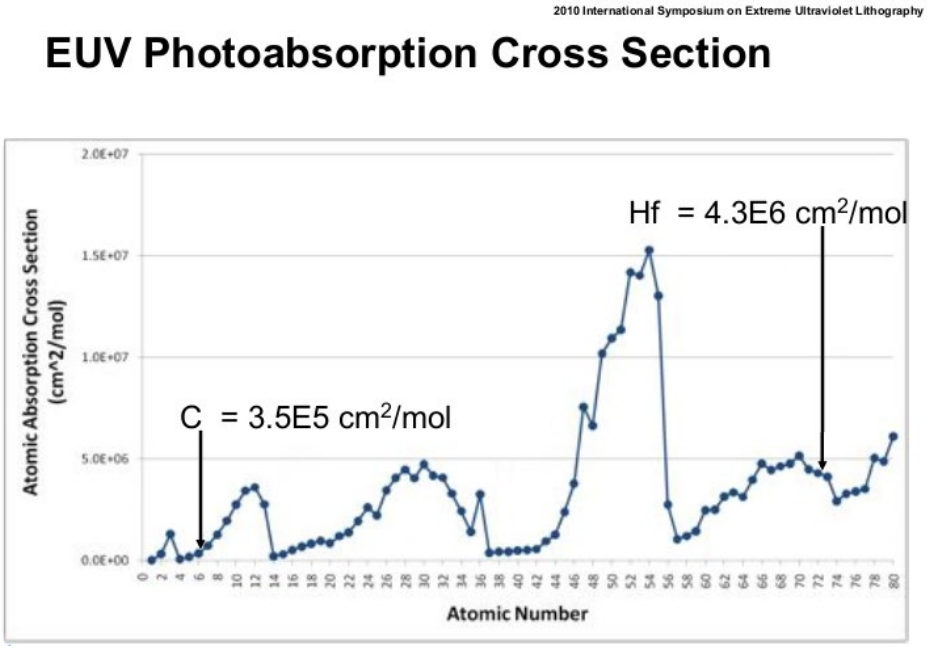
7. Proposed Claim 18: The structure of claim 13 wherein the coating material has high absorption of EUV radiation relative to conventional radiation-based resists

114. A material's ability to absorb EUV radiation is an inherent property of the material itself. A subsequent publication by Stowers demonstrates that the MSOx materials (including HafSOx) have approximately a 4-times higher absorption than a typical polyhydroxystyrene ("PHS") photoresist. EX1022, 3. Dr. Stowers showed that the EUV absorption was 4.1 μm^{-1} for PHS photoresists compared to 16 μm^{-1} for MSOx photoresists. *Id*. Accordingly, the hafnium or zirconium coating materials of Keszler also have high absorption of EUV radiation relative to conventional radiation-based resists.

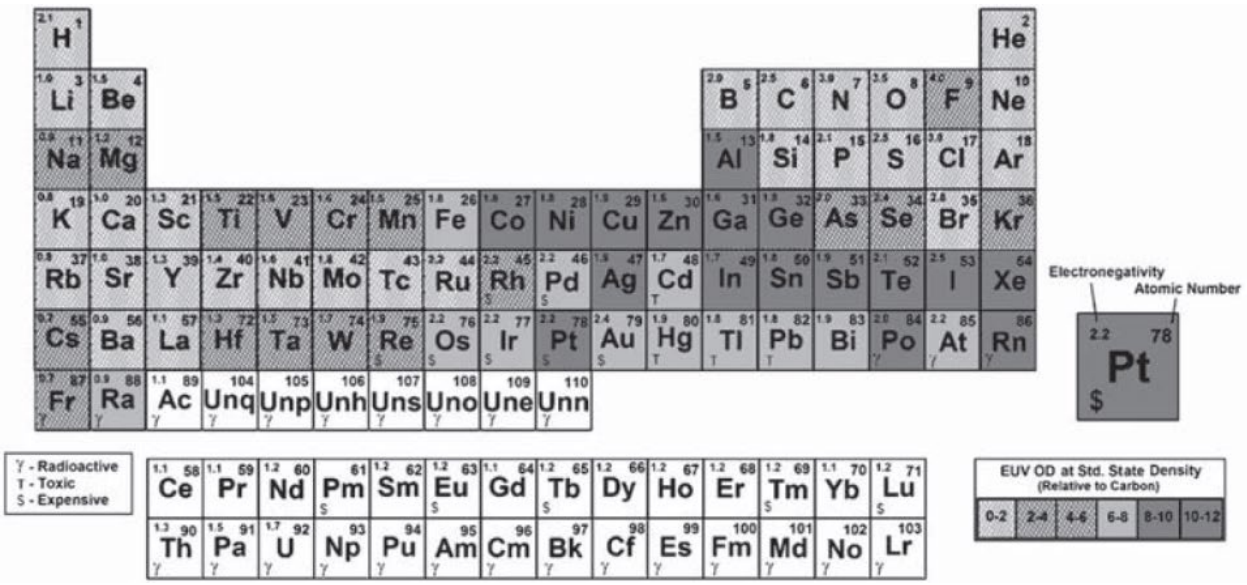
115. Additionally, Dr. Stowers, and others, have recognized that metals have a higher EUV absorbance than carbon, and therefore **metal** oxide photoresists, such as the HafSOx or ZircSOx disclosed in Keszler, will necessarily have higher EUV absorbance than carbon-based photoresists. For example, in a 2010 presentation, Dr. Stowers characterized metal oxide photoresists as "high EUV absorbance":



EX 1013, 3. In that same presentation, Dr. Stowers showed that the EUV absorption of hafnium is significantly higher than that of carbon:



Id., 12. Others have also recognized that metals, such as hafnium, have a higher EUV absorbance than carbon:



§. Accordingly, because the Keszler films incorporate a metal, which has higher EUV absorbance than a conventional, carbon-based resist, the Keszler films have high absorption of EUV radiation relative to conventional radiation-based resists.

116. Moreover, As I explained above, because the films of the ’564 patent are the same in all material aspects as the films of Keszler, they necessarily have the same properties. The ’564 patent discloses the coatings have high absorption relative to convention radiation-based resists. EX1001, 8:67-9:2 (“Relative to organic resists, the inorganic coating material provides distinct advantages with respect to absorption of selected radiation…”); 11:26-27 (“HfO2+ provides good absorption of electron beam material and extreme UV radiation.”). Accordingly, the exposed regions of the Keszler films necessarily have high absorption of EUV radiation relative to conventional radiation-based resists. Keszler therefore discloses this limitation.

8. Proposed Claim 20: The structure of claim 13 wherein the coating material has been heated to temperatures from 45° C. to 150° C.

117. Keszler discloses that the films of Example 4 "were heated "at 125 °C for 1-2 minutes" after deposition. EX1038, 40:17-19. Similarly, the Keszler films in Example 7 were "heated at modest temperatures of less than 100 °C" before exposure. *Id.*, 44:11-12. Keszler therefore discloses this limitation.

9. Proposed Claim 21: The structure of claim 13 wherein following exposure an exposed coating region is insoluble in an aqueous solvent

118. A POSA would understand proposed claim 21 to recite a negative tone resist. For example, Keszler explains:

> Negative resists behave in just the opposite manner [as positive resists]. Exposure to the UV light causes the negative resist to polymerize, and it therefore is more difficult to dissolve. As a result, the negative resist [] remains on the surface wherever it is exposed, and the developer solution removes only the unexposed portions.

Id., 28:25-28. Thus, when "[t]he solubility of the exposed areas decreases relative to the unexposed areas[,] [t]he unexposed areas can then be selectively etched, producing a negative-tone image in the film." *Id.*, 8:7-9.

119. Keszler explains that the "resist may be a positive resist or a negative resist." EX1038, 6:26. For example, Keszler discloses that the exposed portions of the negative tone resists are "insoluble in acid." *Id.* 29:2-5, 32:25-27. Kesler Example 7 develops the exposed HafSOx films "with 5-12 M HCl." *Id.*, 44:18-21. Acids, such as hydrochloric acid ("HCl") are undisputedly an aqueous solution. *E.g.*, *id.*, 32:21-26 (explaining "[e]xamples of acids or bases that can be used [for development] include hydrochloric acid" and that the film is "developed in aqueous acid … to etch the unexposed regions."). Keszler therefore discloses this limitation.

10. Proposed Claim 22: The structure of claim 13 wherein following exposure an exposed coating region is soluble in an organic solvent

120. A POSA would understand proposed claim 21 to recite a positive tone resist. For example, Keszler explains:

> For positive resists, the resist layer [] is exposed to an energy source, such as UV light, wherever the underlying material is to be removed. In these resists, exposure to the UV light changes the chemical structure of the resist layer [] so that it becomes more soluble in the developer. The exposed resist layer [] is then removed by the developer solution, leaving areas of the bare underlying material. In other words, "whatever shows, goes."

Id. 28:17-23; *See also* EX1001, 1:53-60; EX1023 ¶0037.

121. Keszler explains that the "resist may be a positive resist or a negative resist." EX1038, 6:26. As I explained above, because the films of the '564 patent are the same in all material aspects as the films of Keszler, they necessarily have the same properties. The '564 patent discloses that the exposed portions of the coatings are soluble in an organic solvent. *E.g.,* EX1001, 24:40-41. Accordingly, the exposed regions of the Keszler films will necessarily be soluble in an organic solvent. Keszler therefore discloses this limitation.

11. Proposed Claim 23: The structure of claim 13 wherein the coating material can be effectively patterned with UV radiation or EUV radiation at a dose of no more than 100 mJ/cm^2

122. Keszler discloses that the HafSOx materials were patterned with an "electron beam at dose levels ranging from greater than 0 μC/cm^2 to at least 400 μC/cm^2" and could be patterned as "at dose levels as low as 2 μC/cm^2." EX1038, 33:13-18. As the '564 patent explains, a dose of 100 mJ/cm^2 is equivalent to a dose of 300 μC/cm^2. EX1001, 3:60-66 ("The method can comprise

irradiating a layer of coating material with extreme ultraviolet light at a dose of no more than about 100 mJ/cm^2 or an electron beam at a dose equivalent to no more than about 300 μC/cm^2 at 30 kV.") Accordingly, a dose of 2 μC/cm^2 are equivalent to a UV or EUV dose less than 100 mJ/cm^2.

123. This is further confirmed by later publications by Dr. Stowers and Dr. Keszler. For example, Stowers confirmed that and an e-beam dose of 249 μC/cm^2 for MSOx—such as HafSOx—films have been "well characterized to correspond to an EUV dose of 25 mJ/cm^2." EX1022, 7. Thus, the doses of 2 μC/cm^2 and 400 μC/cm^2 disclosed in Keszler are equivalent to a UV or EUV dose less than 100 mJ/cm^2. This is further confirmed by Dr. Stowers' publication where he demonstrates that HafSOx can be patterned with a dose of less than 100 mJ/cm^2. *E.g.*, EX1022, 5 (EUV dose of 70 mJ/cm^2).

124. Moreover, As I explained above, because the films of the '564 patent are the same in all material aspects as the films of Keszler, they necessarily have the same properties. The '564 patent shows the coatings can be patterned at a dose of 20-80 mJ/cm^2 (i.e., less than 100 mJ/cm^2). EX1001, 22:51-54, 23:37-38. Accordingly, the exposed regions of the Keszler films necessarily can be effectively patterned with UV radiation or EUV radiation at a dose of no more than 100 mJ/cm^2. Keszler therefore discloses this limitation.

B. New Ground 6: Proposed Amended Claim 19 Is Rendered Obvious by Keszler

125. Proposed Claim 19 depends on proposed claim 13 and further states the structure has a "molar concentration ratio of radiation sensitive ligands to metal ion of at least 2." As discussed above, Keszler discloses a precursor solution comprising a metal ion (*i.e.*, HfO^{+2} or ZrO^{+2}), peroxide as the radiation sensitive ligand, and sulfate (*i.e.*, H_2SO_4), which is spin-coated onto substrates. EX1038, 38:1-19. The precursor solution of Keszler Example 4 comprises "a

metal ion concentration of about 0.05 M to 0.4 M with 70% H_2O_2 [the radiation sensitive peroxide ligand] … relative to metal." EX1038, 38:6-63. Accordingly, the ratio of radiation sensitive ligands to metal ion of the Keszler films is 0.7 : 1.

126. The films are deposited from a precursor solution. Keszler discloses that the inclusion of the peroxide in the film provides a "new lithographic patterning method." *Id.* 28:9-12. Accordingly, the peroxide in the films allows for the films to be patterned by radiation (*i.e.*, allows them to be photo-patternable). Therefore, it would have been routine and obvious to optimize the amount of peroxide in the film. The Keszler inventors had already begun optimizing the precursor solution by varying the metal concentration from 0.05 M to 0.4 M. A POSA would understand such optimization to be routine to identify the optimum conditions.

127. Indeed, this is precisely what the Keszler inventors (Keszler and Stowers) did and discovered that at ratios higher than those in the Keszler film, the concentration saturates, and the remaining peroxide is unbound. EX1019, 4. In this study, they published a study showing the differences in peroxide to metal ratios in HafSOx resist materials. EX1019, 4. As discussed above, EX1019 shows that that for peroxide to metal ratios less than 0.5 "all of the peroxide coordinates to the [hafnium] clusters." *Id.* However, when the ratio "exceeds 0.5, the free peroxide stretching mode … appears while the bound peroxide saturates in intensity" which "suggests that approximately two peroxide groups coordinate to each hafnium tetramer." *Id.*

128. Accordingly, it would have been obvious to modify the Keszler films to have a ratio of radiation sensitive ligands to metal ion of at least 2 of through routine optimization.

C. The Proposed Amended Claims Are Not Patentable Over Stowers 2008 and Trikeriotis

1. Proposed Amended Claim 13 Is Not Patentable Over Stowers 2008 and Trikeriotis

129. I understand that Inpria has proposed substitute claims that require the thickness to be "about 25 nm" as follows:

> 13. A structure comprising a substrate and a coating material on a surface of the substrate, wherein the coating material comprises metal ions with radiation sensitive ligands and wherein the coating material has an average thickness of about 25 nm~~from 5 nm to 30 nm,~~ wherein exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material.

Mot., 18. However, this amendment addresses only the thickness limitation. Merely amending the required thickness does not make proposed substitute independent claim 13 is patentable over Stowers 2008 or Trikeriotis. Accordingly, proposed amended claim 13 is not patentable for all the reasons in my previous declaration (EX1002) and this declaration and the thickness modification is anticipated by and/or obvious over Stowers 2008 and Trikeriotis.

a. Stowers 2008 Discloses HafSOx and ZirSOx Materials with a thickness of about 25 nm

130. Stowers 2008 discloses depositing HafSOx and ZircSOx materials in films ranging in thickness from 2 to 500 nm." EX1008, 18. Stowers 2008 also states that "HafSOx, a hafnium oxide sulfate, and its zirconium analog ZircSOx are spin-coated materials that exhibit extraordinary film qualities. These materials has (sic) been deposited in thickness ranges of 3 to 500 nm." *Id.*, 66; 68 (HafSOx and ZircSOx "[f]ilms have been demonstrated as thin as 3 nm with retention of film quality."); 70 ("HafSOx has been deposited as thin as 2-3 nm in a film…This serves to demonstrate a fundamental difference of SOx materials from traditional polymer

resists."). Stowers 2008 thus discloses HafSOx and ZircSOx films having thicknesses that overlap with the claimed thickness of "about 25 nm." I am not aware of any evidence of that the claimed thickness is critical to the operability of the claimed invention.

131. Moreover, a POSA would have recognized that film thickness is a variable that a POSA would have been motivated to optimize. For example, A POSA would have known that pattern collapse, which refers to the deformation of photoresist lines that occurs during the development process, was an issue that needed to be mitigated. *See, e.g.*, EX1012 at Abstract ("However for sub 100 nm line width patterns, the pattern collapse, generated during the drying step of the developing process, becomes a serious limiting phenomenon."). A POSA would have recognized that film thickness was one parameter that could be optimized to help avoid pattern collapse. *Id.*, 1 ("Two solutions allow avoiding pattern collapse: decreasing the resist thickness or change the dry process step."); *id.*, 8 ("we have shown that the mode of pattern collapse depends on the pattern height").

132. Given the ease with which film thickness can be varied, *e.g.*, by adjusting spin rates and times (EX1008, 6; EX1001, 14:30-32), finding an optimum thickness range to mitigate pattern collapse would have required nothing more than routine experimentation. Indeed, Stowers 2008 already teaches that depositing films "at 3000 rpm for 30 seconds result[ed] in films 30 to 35-nm thick." EX1008, 70; *see also id.*, 86 ("HafSOx has been deposited as thin as 2-3 nm in a film."). Thus, a POSA could have easily deposited the claimed coatings at a thickness of about 25 nm based on the teachings of Stowers 2008 with routine experimentation.

b. It would have been obvious modify the coatings of Trikeriotis to have a thickness of about 25 nm

133. Trikeriotis discloses that "film thicknesses ranging from 30 nm to 200 nm can be easily achieved." EX1024, 18. Although Trikeriotis was testing a variety of film thicknesses, including those thicker than 30 nm, Trikeriotis explained that "[t]he trend of ever decreasing feature sizes in subsequent lithography generations is paralleled by the need to reduce resist thickness to prevent pattern collapse." EX1024, 15. This is consistent with a POSA's understand that film thickness should be reduced to avoid pattern collapse. *E.g.*, EX1012, 8 ("pattern collapse depends on the pattern height"). Accordingly, as discussed above a POSA would have recognized that film thickness is a variable that a POSA would have been motivated to optimize.

134. Given the ease with which film thickness can be varied, *e.g.*, by adjusting spin rates and times (EX1008, 6; EX1001, 14:30-32), finding an optimum thickness range to mitigate pattern collapse would have required nothing more than routine experimentation. Indeed, Dr. Banerjee stated during his deposition that optimizing the thickness "is not rocket science[,] [i]t's not extensive experimentation by any means." EX1033, 75:5-11. Thus, a POSA could have easily deposited the claimed coatings at a thickness of about 25 nm based on the teachings of Stowers 2008 with routine experimentation.

2. Proposed Amended Claim 19 Is Not Patentable Over Stowers 2008

135. I understand that Inpria has proposed a substitute dependent claim that requires the molar concentration ratio of radiation sensitive ligands to metal ion of at least about 2 as follows:

> 19. The structure of claim 13~~1~~ having a molar concentration ratio of radiation sensitive ligands to metal ion ~~to radio-sensitive ligand ratio~~ of at least about 2.

Mot., 14. However, Inpria's proposed amendment only inverts the ratio of claim 8 of the '564 patent. Accordingly, proposed amended claim 19 is not patentable for all the reasons in my previous declaration (EX1002) and this declaration—the ratio modification is obvious in view of Stowers 2008.

136. Stowers 2008 discloses a metal oxide precursor solution that is deposited onto substrates that made by mixing $ZrOCl_2$ or $HfOCl_2$ with H_2O_2 (peroxide) and then adding H_2SO_4 (sulfuric acid). EX1008, 68. The peroxide to Hf (*i.e.*, radiation sensitive ligand to metal) ratio of in the Stowers 2008 precursor solution is 0.75 to 1. Stowers discloses that the addition of peroxide allows the films to act as a photoresist (*i.e.*, they become photo-patternable). *Id.* 143 (the dielectric materials were adapted to be photoresists by "changing the chemistries to a peroxide based system."). Accordingly, would have been routine and obvious to the optimize concentration of peroxide in the solution and/or film (and therefore the ratio of ligand to metal). A POSA would understand such optimization to be routine to identify the optimum conditions.

137. Indeed, this is precisely what Stowers later did. EX1019, 4. Stowers discovered that at ratios higher than those in the Stowers 2008 films, the concentration saturates, and the remaining peroxide is unbound. *Id.* This study showed the differences in peroxide to metal ratios in HafSOx resist materials. *Id.* As discussed above, EX1019 shows that that for peroxide to metal ratios less than 0.5 "all of the peroxide coordinates to the [hafnium] clusters." *Id.* However, when the ratio "exceeds 0.5, the free peroxide stretching mode … appears while the bound peroxide saturates in intensity" which "suggests that approximately two peroxide groups coordinate to each hafnium tetramer." *Id.*

138. Accordingly, it would have been obvious to modify the Stowers films to have a ratio of radiation sensitive ligands to metal ion of at least 2 of through routine optimization.

D. The Scope of Proposed Amended Claim 19 is Larger Than the Scope of Original Claim 8, Which It Amends

139. As part of my analysis, I was asked to evaluate the scope of the PO's proposed substitute claim 19 (Mot., 7), and compare that claim scope to original claim 8, which proposed substitute claim 19 amends.

140. Original claim 8 is directed to the "structure of claim 1 having a metal ion to radiosensitive ligand ratio of at least 2." EX1001 at claim 8. As such, claim 8 covers structures having a **metal ion to ligand ratio** of 2 to 1, 3 to 1, 4 to 1, etc.

141. Proposed substitute claim 19 amends original claim 8 to claim the "structure of claim 13 having a "molar concentration ratio of radiation sensitive ligands to metal ion of at least about 2." Mot., 7. The claimed ratio of radiation sensitive ligands to metal ion of at least about 2, is equivalent to a metal ion to ligand ratio of no more than about 1 metal ion for every 2 ligands. Accordingly, proposed substitute claim 19 covers structures having a **metal ion to ligand ratio** of 1 to 2, 1 to 3, 1 to 4, etc. These metal ion to ligand ratios of 1 to 2, 1 to 3, and 1 to 4, etc., which fall within the scope of proposed substitute claim 19, do not fall within and are outside the scope of original claim 8.

142. Since proposed substitute claim 19 covers structures having metal ion to ligand ratios that are outside the scope of original claim 8, PO's amendments as included in proposed substitute claim 19 have enlarged the scope of original claim 8.

E. Lack of Written Description Support For the Proposed Amended Claims

143. Proposed substitute claim 13 is directed to a "structure comprising a substrate and a coating material." Mot., 6. The coating material "comprises metal ions with radiation sensitive ligands," wherein "exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material." *Id.*

144. A POSA would understand that the claim language of proposed substitute claim 13 includes functional requirements. Specifically, claim 13 includes a functional requirement that the metal ions have ligands that are "radiation sensitive." *Id.* Claim 13 also includes a functional requirement that "radiation alters the chemical properties" of the coating material. *Id.*

145. A POSA would understand that the technology that is the subject of claim 13 and the remaining proposed substitute claims is highly unpredictable. Specifically, the functional limitations of claim 13 relate to chemistry and chemical properties of certain compounds. Chemical reactions and properties can be highly variable and difficult to predict. For example, combining certain elements or functional groups into a molecule can affect the properties of the molecule in unpredictable ways. While a certain element or functional group may have known properties when found in isolation or when combined with certain other elements, those properties may be different when that same element is included in a different molecule. Similarly, the inclusion of different additives or compounds in a material can affect the properties of the material in unpredictable ways. One chemical property that can be difficult to predict is whether a specific ligand will be "radiation sensitive."

146. A POSA would understand that, as used in the '564 patent, a "ligand" is an atom or molecule with a functional group that binds to a central atom—typically a metal—in a

coordination complex. Given this understanding, a POSA would know that there are nearly an infinite number of atoms or molecules that could qualify as a ligand.

147. The specification of the ’564 patent identifies only one example of “radiation sensitive ligands”: ligands with peroxide groups. EX1001, 5:45-47, 6:9-14. But a POSA would understand that a large number of other ligands could potentially be “radiation sensitive.”

148. However, the specification of the ’564 patent does not provide any common structural features of ligands that result in “radiation sensitive” functionality. *See* EX1001. Aside from identifying the example of peroxide ligands, nothing in the specification discusses any structures of ligands that provide ligands with radiation sensitivity. *Id.*

149. As discussed above, due to the unpredictability of chemistry and chemical properties, combining certain elements or functional groups into a molecule can affect the properties of the molecule in unpredictable ways. As relevant here, combining different metal ions with different ligands could affect the radiation sensitivity of the ligands. As an example, A POSA would understand that a ligand that has radiation sensitivity as an independent molecule, or when used with one type of metal ion, may not have the same radiation sensitivity when used with a different metal ion.

150. The specification of the ’564 patent identifies only two examples of “metal ions with radiation sensitive ligands.” EX1001, 21:12-23:41. These two examples are (1) hafnium ions with ligands made from hydrogen peroxide, and (2) zirconium ions with ligands made from hydrogen peroxide. EX1001, 21:12-23:41. The disclosure of these two limited examples are insufficient to adequately describe the entire genus of “metal ions with radiation sensitive ligands.”

The specification of the ’564 patent also does not identify any common structures of “metal ions with…ligands” that result in the ligands having “radiation sensitive” functionality. *See* EX1001.

151. Proposed substitute claim 13 requires a coating material wherein “exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material.” Mot., 6.

152. The specification of the ’564 patent identifies only two examples of coating materials wherein “exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material.” EX1001, 21:12-23:41. These two examples are (1) a coating material made from hafnium ions with ligands made from hydrogen peroxide, and (2) a coating material made from zirconium ions with ligands made from hydrogen peroxide. EX1001, 21:12-23:41. The disclosure of these two limited examples are insufficient to adequately describe the entire genus of coating materials covered by claim 13.

153. The specification of the ’564 patent includes a section describing how the use of different ions can affect radiation absorption. EX1001, 11:21-42. The specification of the ’564 patent also describes that “absorption of a sufficient amount of radiation” can lead to chemical changes in the coating material. EX1001, 15:33-43. But the specification of the ’564 patent does not provide any actual examples of a coating material wherein there is “absorption of a sufficient amount of radiation” such that the chemical properties of the coating material are altered by the radiation—except for the two examples (hafnium and zirconium peroxide examples) identified above. *See e.g.,* EX1001, 21:12-23:41.

154. The specification of the ’564 patent also does not identify any common structures of coating materials that provide functionality wherein “exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material.” *See*

EX1001. Due to the lack of disclosure of any common structural features, the ’564 patent does not adequately describe the full scope of claimed coating materials.

F. Lack of Enablement For the Proposed Amended Claims

155. As discussed above, proposed substitute claim 13 is directed to a “structure comprising a substrate and a coating material.” Mot., 6. The coating material “comprises metal ions with radiation sensitive ligands,” wherein “exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material.” *Id*

156. I was asked to evaluate whether a POSA would be enabled to make and use the full scope of (1) the claimed “metal ions with radiation sensitive ligands,” and (2) coating materials wherein “radiation alters the chemical properties of the coating material” in view of the specification of the ’564 patent. In my opinion, the full scope of the claims is not enabled.

157. Claim 13 is directed to any combination of metal ions with any radiation sensitive ligands. Mot., 6. The specification of the ’564 patent provides examples of numerous metal ions that could ***potentially*** be used in connection with the material coating of claim 13. *See e.g.,* EX1001, 10:65-11:12. And a POSA would understand that the claims ***could*** cover metal ions that are not specifically listed in the specification. A POSA would also understand that there are nearly an infinite number of atoms or molecules that could qualify as a ligand.

158. The specification of the ’564 patent identifies only two examples of “metal ions with radiation sensitive ligands.” EX1001, 21:12-23:41. These two examples are (1) hafnium ions with ligands made from hydrogen peroxide, and (2) zirconium ions with ligands made from hydrogen peroxide. EX1001, 21:12-23:41. By reading the specification of the ’564 patent, a POSA would know how to make and use those specific examples.

159. However, a POSA would not know which other combinations of metal ions and ligands resulted in "metal ions with radiation sensitive ligands." In order to make and use the full scope of the invention, a POSA would have to repeatedly make photoresists using a combination of metal ions and ligands, and then test each and every combination in order to determine if the ligands were radiation sensitive. Given, (1) the sophisticated materials and equipment involved in the manufacture of photoresists, and (2) the near-infinite number of potential combinations of metal ions and ligands, the required testing would be a lengthy and arduous process, and would qualify as undue experimentation.

160. I have reviewed the deposition transcript of Dr. Banerjee, PO's expert, and I note that he agrees with me regarding the process required to determine whether a metal ion/ligand combination would be radiation sensitive. Specifically, Dr. Banerjee testified:

> Q Is there something in the '564 patent specification that would help you determine whether a particular ligand is a radiation sensitive ligand suitable for use in the structure of claim 1 of the '564 patent?
>
> A Yeah. If the POSA was a synthetic chemist, he or she could, you know, mix up this precursor solution and then expose that resist and then measure the etch rate and the developer and compare them between the exposed and the
>
> nonexposed parts. That would be one experimental technique that you could use.
>
> Q So a POSA would need to test by creating the precursor solution and exposing that resist and then measuring the etch rate and the development and compare to understand whether a ligand is radiation sensitive such that it is suitable for use in claim 1 of the '564 patent?

…

A I've not discussed any of that or analyzed any of that in my report, but in general the answer is yes. ***R&D takes time.***

EX1033, 159:13-160:12.

161. Similarly, the specification of the ’564 patent identifies only two examples of coating materials wherein “radiation alters the chemical properties of the coating material.” EX1001, 21:12-23:41. Again, these two examples are (1) coating materials made of hafnium ions with ligands made from hydrogen peroxide, and (2) coating materials made of zirconium ions with ligands made from hydrogen peroxide. EX1001, 21:12-23:41. By reading the specification of the ’564 patent, a POSA would know how to make and use those specific examples of coating materials.

162. But again, a POSA would not know which other coating materials would result in a material wherein “radiation alters the chemical properties of the coating material.” In order to make and use the full scope of the alleged invention, a POSA would have to repeatedly make photoresists, and then test each and every photoresist in order to determine if the ligands were radiation sensitive. This process would be even more arduous than the process above, because coating materials often include other additives or compounds (e.g., for viscosity control or stability) that could affect the coating material’s response to radiation. Accordingly, given (1) the sophisticated materials and equipment involved in the manufacture of photoresists, and (2) the near-infinite number of potential coating materials, the required testing would be a lengthy and arduous process, and would qualify as undue experimentation.

G. Indefiniteness of the Proposed Amended Claims

1. "about 25 nm"

163. Proposed substitute claim 13 includes a limitation directed to a thickness of "about 25 nm." A POSA, reviewing the specification and prosecution history of the '564 patent would not be able to determine the thicknesses that would fall within the scope of "about 25 nm" with reasonable certainty.

164. First, I have reviewed the specification of the '564 patent, and nothing in the specification informs a POSA as to the amount of deviation or variation that is permitted or contemplated by the use of the modifier "about." *See* EX1001. It is unclear whether "about" 25 nm would allow 1% deviation from 25 nm, 5% deviation, 10% deviation, 25% deviation, or some other range. Simply put, the range of thicknesses permitted by the "about 25 nm" limitation would not be clear to a POSA because the specification of the '564 patent provides no detail on how to evaluate the claim language.

165. Second, my opinion is confirmed by the prosecution history of the '564 patent. Specifically, I note that, during the prosecution of the '564 patent, the PO included claims directed to a material coating thickness range that included "about 30 nm":

> 1. (Original) A structure comprising a substrate and a coating material on a surface of the substrate, wherein the coating material comprises metal ions with radiation sensitive ligands and wherein the coating material has an average thickness from about 5 nm to about 30 nm, wherein exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material.

EX1007, 404.

166. The examiner rejected the claim and explained that the term "about" was indefinite:

> 4. Claims 1-7 and 21-25 are rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which the inventor or a joint inventor, or for pre-AIA the applicant regards as the invention. Instant claims 1-7 and 21-26 recite the term, "about". The term "about" in claims 1, 6, 21, 22 and 25 is a relative term which renders the claim indefinite. The term "about" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. Appropriate correction is required.

EX1007, 489.

167. The examiner explained further:

> C) Examiner respectfully disagrees that in the instant disclosure that the term, "about" is clear and not indefinite. First, the instant disclosure fails to define the meaning of the term, "about" in the claims or specification. The term "about" is not defined by the claim, the specification does not provide a standard for ascertaining the requisite degree, and one of ordinary skill in the art would not be reasonably apprised of the scope of the invention. The courts have stated in determining the range encompassed by the term "about", one must consider the context of the term as it is used in the specification and claims of the application. Ortho-McNeil Pharm., Inc. v. Caraco Pharm. Labs., Ltd., 476 F.3d 1321, 1326, 81 USPQ2d 1427, 1432 (Fed. Cir. 2007). The instant disclosure recites the following on page 33, lines 13-15:
>
> The coating material was produced according to the method of Example 2 with a ZrO^{+2}-based coating composition. The coating material was deposited at a thickness form 10-50 nm. The Zr-based coating material was irradiated with patterned 193-nm

EX1007, 496.

168. While the PO stated that it disagreed with the examiner's rejection, it ultimately rewrote the claims to remove "about." EX1007, 504. This history, including the ultimate removal of "about" from the claim language, confirms my opinion that a POSA would not be able to determine the scope of the term "about 25 nm."

2. "metal ions"

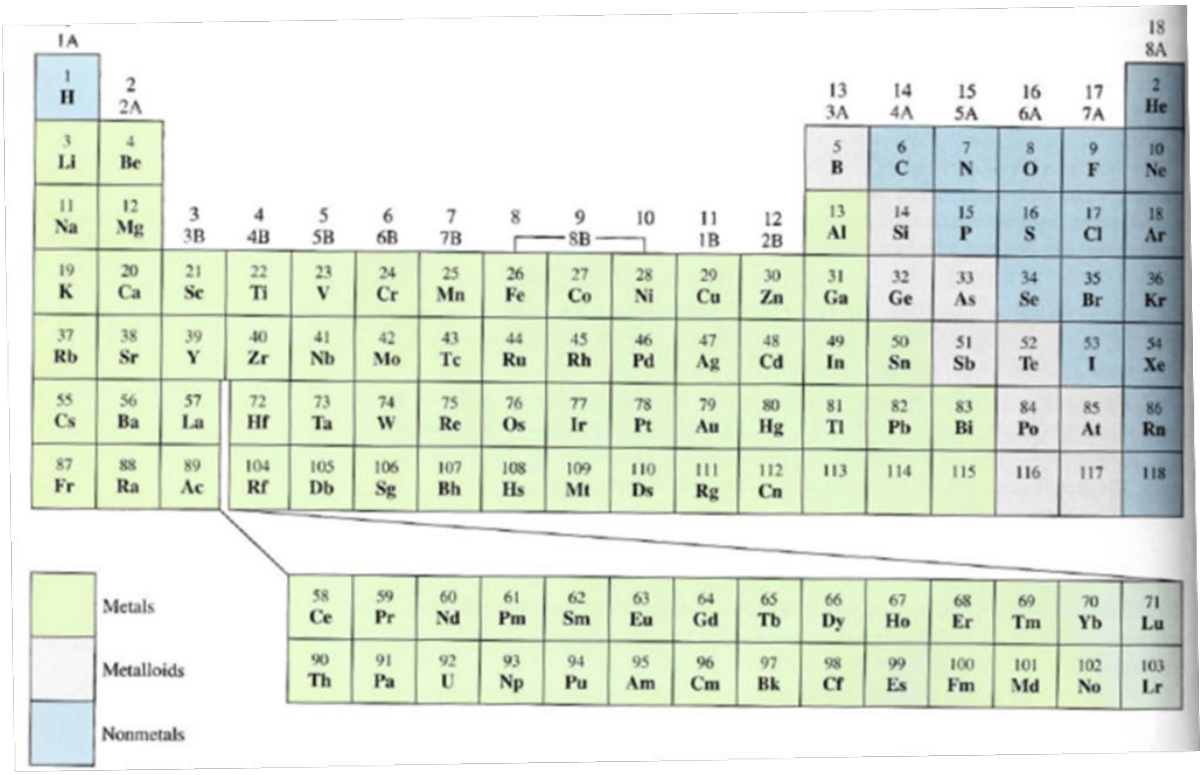
169. Claim 13 includes a limitation directed toward "metal ions." Mot., 6. A POSA would understand that the term "metal ions" simply refers to any ions of a "metal" element. This is supported by substitute claim 15, which further requires that "the ***metal*** of the metal ions comprises…." Mot., 6-7.

170. A POSA would understand that each element is traditionally characterized as a "metal," "nonmetal," or "metalloid," each of which have different properties:

> The elements can be divided into three categories – metals, nonmetals, and metalloids. A **metal** is a good conductor of heat and electricity while a **nonmetal** is usually a poor conductor of heat and electricity. A **metalloid** has properties that are intermediate between those of metals and nonmetals.

EX1039, 3 (emphasis in original).

171. The standard periodic table that is well known to those in field, and even to students just beginning to learn about chemistry, identifies each element as a metal, metalloid, or nonmetal:



Id.

172. Typically, a POSA would understand the term "metal" to refer to elements identified as "metals" on the periodic table.

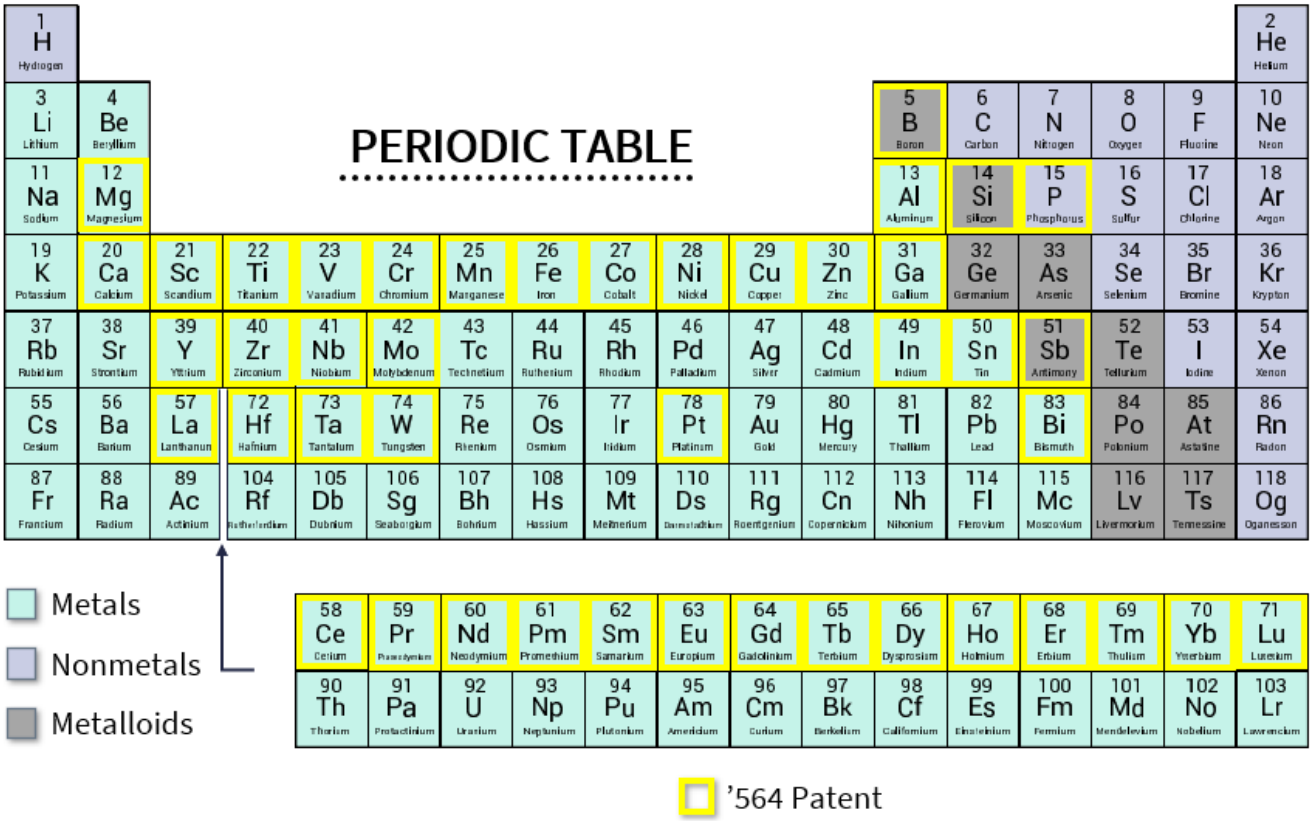
173. However, the specification of the '564 patent provides some examples of elements that it refers to as "metals" that a POSA would understand to be either metalloids or nonmetals. For example, the '564 patent describes boron, silicon, and antimony as examples of metals, even though a POSA would understand those elements to be metalloids. And the '564 patent describes phosphorus as an example of a metal, even though a POSA would understand phosphorous to be a nonmetal.

> Further tuning of the composition for radiation absorption can be adjusted based on the addition of **<u>other metal ions</u>**. **<u>For example</u>**,

> one or more ions (cations or anions) comprising titanium, zinc, calcium, indium, tin, **antimony**, bismuth or combinations thereof can be added to the precursor solution to form a coating material with an absorption edge moved to longer wavelengths, to provide, for example, sensitivity to 248 nm wavelength ultraviolet light. **Also**, one or more ions (cations or anions) comprising magnesium, **boron**, calcium, aluminum, **silicon**, ***phosphorous*** or combinations thereof can be used to increase the absorption cross section at shorter wavelengths.

EX1001, 11:21-42 (emphasis added).

174. The annotated periodic table below shows the traditional breakdown of metal, nonmental, and metalloid elements as well as the examples of "metals" that are included in the '564 patent.



175. As is clear, the examples in the specification of the '564 patent includes some, but not all, metalloids in its list of exemplary metals, and similarly includes the nonmetal phosphorus in the same list.

176. As such, a POSA would understand that the use of "metal" in claim 13 must be broader than just those elements traditionally defined as "metals." This is confirmed by substitute claim 15, which expressly includes "antimony (Sb)" as one of the possible claimed "metals." Mot., 6-7.

177. Yet, the specification does not define either "metal" or "metalloid"—offering no further guidance as to the appropriate scope of "metal." Accordingly, a POSA would be unable to determine which additional elements should fall within the scope of "metals." For example, a

POSA would not know whether ions of additional, unlisted metalloids like germanium, arsenic, and tellurium would qualify as "metal ions" within the meaning of claim 13. Similarly, a POSA would not know whether ions of additional, unlisted nonmetals like sulfur or selenium would qualify as "metal ions" within the meaning of claim 13. Accordingly, a POSA would determine that the term "metal ions" is indefinite.

3. "high absorption"

178. Claim 18 includes a term of degree limitation requiring the coating material to have "high absorption…relative to conventional radiation-based resists." Mot., 7. The '564 patent specification provides no guidance as to how to evaluate whether a coating material has "high absorption," rendering this limitation indefinite.

179. First, the identified claim limitation requires a comparison between the absorption of the claimed resist as compared to "conventional radiation-based resists." Mot., 7. Numerous conventional resists are known in the art, and such resists have different absorption characteristics. Yet, the specification of the '564 patent provides no description of which conventional resists to use for this comparison, nor how many must be used. *See* EX1001. Nor does the specification provide a description of the absorption levels of "conventional radiation-based resists" that could be used as a baseline for comparison. *Id.* Accordingly, the specification lacks any guidance as to how to determine the baseline absorption information to be used in the comparison required by claim 18.

180. Moreover, the specification provides no guidance as to what level of increased absorption qualifies as "high absorption" as required by claim 18. EX1001. The term "high absorption" is not found anywhere in the specification. And while a POSA would understand that

the absorption of the claimed resist must be higher than the absorption of a conventional resist, a POSA would have no way to determine how much higher is needed to qualify as "high absorption"—e.g., whether a 10%, 20%, 50%, etc. increase in absorption was needed.

181. Given the lack of any meaningful way to measure "high absorption," a POSA would be uncertain as to the scope of claim 18.

4. "effectively patterned"

182. Claim 23 includes a term of degree requiring the coating material to be "effectively patterned." Mot., 7-8. The '564 patent specification provides no guidance as to how to evaluate whether a coating material can be "effectively patterned," rendering this limitation indefinite.

183. While a POSA would understand whether or not a coating material could be "patterned" at all—a POSA would have no way to determine whether a coating material could be "effectively patterned." The specification of the '564 patent does not provide any guidance as to how to determine "effective patterning." EX1001. There is no test for effective patterning described in the specification, nor is there any description of characteristics that can be used to determine whether such patterning is "effective." *Id.* Instead, whether a coating material could be "effectively patterned" could only be a subjective determination which could differ according to the evaluator. For example, one technician may find certain patterning characteristics to qualify as "effectively patterned" while another technician looking at the same resist might disagree. This uncertainty in claim scope renders claim 23 indefinite.

5. "exposure" and "heat[ing]" method steps

184. Proposed substitute claim 13 is generally directed to a "structure comprising a substrate and a coating material. Mot., 6. However, it includes a limitation that recites "wherein

exposure of the coating material to UV, EUV and/or electron-beam radiation alters the chemical properties of the coating material creating an exposed coating material with differential dissolution rates between exposed and un-exposed regions of the coating material." Mot., 6.

185. A POSA would understand that this "wherein" clause requires a method step—*e.g.*, "exposure of the coating material to UV, EUV and/or electron-beam radiation." However, in my opinion, a POSA would not recognize any of the structures recited in claim 13 as performing the claimed exposure step. Specifically, the "exposure" step would necessarily be performed by a system that creates radiation—for example, the devices described at EX1001, 15:65-16:12—and then directs the radiation onto the coating material. This system is not part of the structure found in claim 13, which is limited to a substrate and a coating material. Mot., 6.

186. Similarly, proposed substitute claim 20 is directed to the same structure as claim 13 but includes a limitation reciting "wherein the coating material has been heated to temperatures from 45° C. to 150° C." Mot., 7.

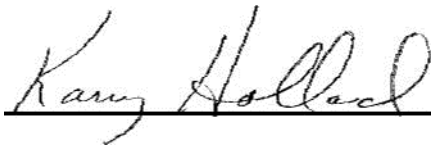
187. A POSA would understand that this "wherein" clause requires a method step—*e.g.*, "heat[ing]" the coating material "to temperatures from 45° C. to 150° C." Again, in my opinion, a POSA would not recognize any of the claimed structures as performing the claimed heating step. Specifically, the "heat[ing]" step would necessarily be performed by an unclaimed heating element which would include a controllable heat source. This heating element is not part of the structure found in claim 13, which is limited to a substrate and a coating material. Mot., 7.

VII. CONCLUSION

188. I declare that all statements made herein of my knowledge are true, and that all statements made on information and belief are believed to be true. I further declare that all

statements made herein were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code.

189. In signing this declaration, I understand that the declaration will be filed as evidence in a contested case before the Patent Trial and Appeal Board of the United States Patent and Trademark Office. I acknowledge that I may be subject to cross examination in the case and that cross examination will take place within the United States. If cross examination is required of me, I will appear for cross examination within the United States during the time allotted for cross examination.

DATE: October 23, 2024 By: 

Karey Holland, Ph.D.