

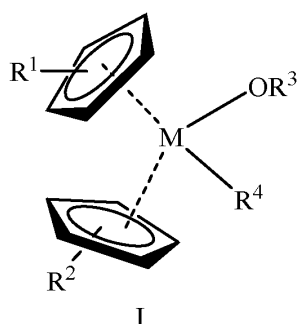
Claims

What is claimed is:

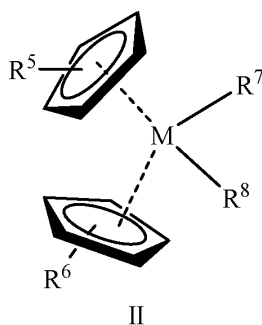
1. A thin film crystalline material comprising hafnium oxide and zirconium oxide, wherein the crystalline material exhibits ferroelectric behavior as deposited.
2. The crystalline material of claim 1, wherein
 - (i) greater than 40% of the total volume of the crystalline material is in a ferroelectric phase; and
 - (ii) less than 60% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
3. The crystalline material of claim 1, wherein less than 50% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
4. The crystalline material of claim 1, wherein less than 40% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
5. The crystalline material of claim 1, wherein less than 30% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
6. The crystalline material of claim 1, wherein less than 25% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
7. The crystalline material of claim 1, wherein less than 20% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
8. The crystalline material of claim 1, wherein less than 15% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
9. The crystalline material of claim 1, wherein less than 10% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
10. The crystalline material of claim 1, wherein less than 5% of the total volume of the crystalline material constitutes a non-ferroelectric phase component.
11. The crystalline material of claim 1, wherein
 - (iii) less than 60% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.
12. The crystalline material of claim 11, wherein less than 50% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.

13. The crystalline material of claim 11, wherein less than 40% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component
14. The crystalline material of claim 11, wherein less than 30% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.
15. The crystalline material of claim 11, wherein less than 25% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.
16. The crystalline material of claim 11, wherein less than 20% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.
17. The crystalline material of claim 11, wherein less than 15% of the total volume of the crystalline material as measured constitutes a non-ferroelectric monoclinic phase component.
18. The crystalline material of claim 11, wherein less than 10% of the total volume of the crystalline material as measured constitutes a non-ferroelectric monoclinic phase component.
19. The crystalline material of claim 11, wherein less than 5% of the total volume of the crystalline material as measured constitutes a non-ferroelectric monoclinic phase component.
20. The crystalline material of claim 1, wherein
 - (i) greater than 50% of the total volume of the crystalline material is in a ferroelectric phase;
 - (ii) less than 50% of the total volume of the crystalline material constitutes a non-ferroelectric phase component; and
 - (iii) less than 25% of the total volume of the crystalline material constitutes a non-ferroelectric monoclinic phase component.
21. The crystalline material of any of claims 1-20, wherein a hafnium oxide to zirconium oxide ratio is between approximately 1:3 and approximately 3:1.
22. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 6 atomic percent.
23. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 5 atomic percent.
24. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 4 atomic percent.
25. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 3 atomic percent.

26. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 2 atomic percent.
27. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content below approximately 1 atomic percent.
28. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content between approximately 1 atomic percent and approximately 6 atomic percent.
29. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content between approximately 1 atomic percent and approximately 5 atomic percent.
30. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content between approximately 1 atomic percent and approximately 4 atomic percent.
31. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content between approximately 1 atomic percent and approximately 3 atomic percent.
32. The crystalline material of any of claims 1-20, wherein the crystalline material has a carbon content between approximately 1 atomic percent and approximately 2 atomic percent.
33. The crystalline material of any of claims 1-32, wherein the crystalline material is derived from one or more metallocene precursor having Formula I:

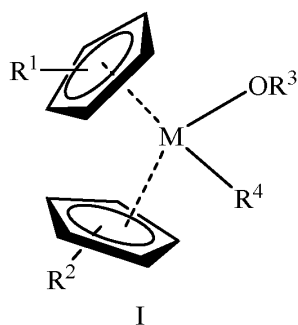


or Formula II:

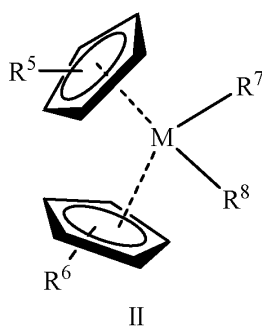


wherein (i) M is selected from Zr and Hf and (ii) R^1 , R^2 , R^3 , R^4 , R^5 , R^6 , R^7 and R^8 are each independently selected from a C_1 - C_6 linear alkyl, a C_1 - C_6 branched alkyl, a C_1 - C_6 halogenated linear alkyl and a C_1 - C_6 halogenated branched alkyl.

34. The crystalline material of any of claims 1-32, wherein the crystalline material is derived from one or more metallocene precursor having Formula I:

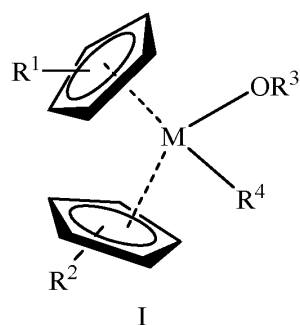


or Formula II:

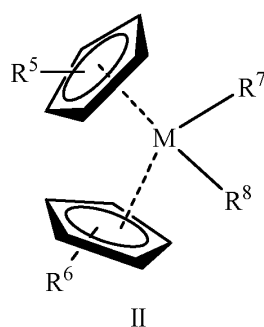


wherein (i) M is selected from Zr and Hf and (ii) R^1 , R^2 , R^3 , R^4 , R^5 , R^6 , R^7 and R^8 are each independently a C_1 - C_6 linear alkyl.

35. The crystalline material of any of claims 1-32, wherein the crystalline material is derived from one or more metallocene precursor having Formula I:



or Formula II:



wherein (i) M is selected from Zr and Hf and (ii) R¹, R², R³, R⁴, R⁵, R⁶, R⁷ and R⁸ are each a methyl group.

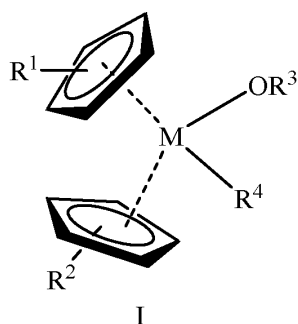
36. A method for depositing the crystalline material a material of any of claims 1-32 comprising:

- (i) providing a substrate at a deposition temperature;
- (ii) exposing the substrate to a first precursor that does not decompose at the deposition temperature;
- (iii) exposing the substrate to a first reaction gas;
- (iv) exposing the substrate to a second precursor that does not decompose at the deposition temperature; and
- (v) exposing the substrate to a second reaction gas,

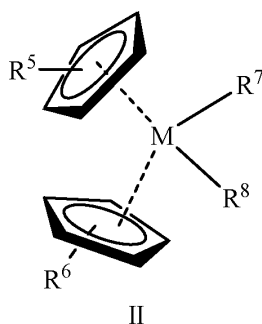
wherein one of the first precursor and the second precursor comprises zirconium and the other of the first precursor and the second precursor comprises hafnium.

37. The method of claim 36, further comprising at least one purging step.

38. The method of claim 36, wherein the first reaction gas and the second reaction gas are each independently a gas containing one or more of oxygen, water, hydrogen peroxide and nitrous oxide.
39. The method of claim 36, wherein the first reaction gas and the second reaction gas are each independently a gas containing oxygen.
40. The method of claim 36, wherein the first reaction gas and the second reaction gas are each independently a gas containing ozone.
41. The method of claim 36, wherein the first reaction gas and the second reaction gas are each independently a gas containing water.
42. The method of claim 36, wherein the first reaction gas and the second reaction gas are the same gas.
43. The method of claim 36, wherein the first reaction gas and the second reaction gas are different gases.
44. The method of claim 36, wherein the first precursor and the second precursor are each independently a precursor having Formula I:

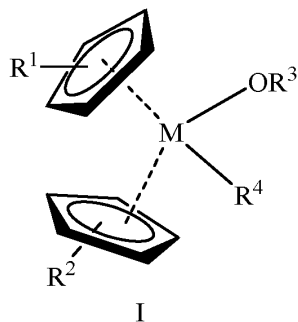


or Formula II:

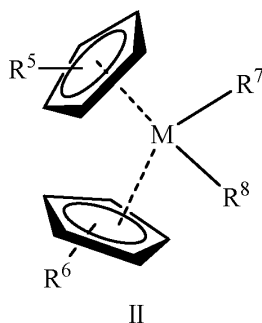


wherein (i) M is selected from Zr and Hf and (ii) R^1 , R^2 , R^3 , R^4 , R^5 , R^6 , R^7 and R^8 are each independently selected from a C_1 - C_6 linear alkyl, a C_1 - C_6 branched alkyl, a C_1 - C_6 halogenated linear alkyl and a C_1 - C_6 halogenated branched alkyl.

45. The method of claim 36, wherein the first precursor and the second precursor are each independently a precursor having Formula I:

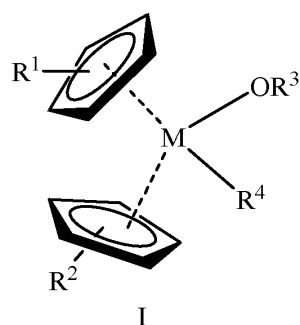


or Formula II:

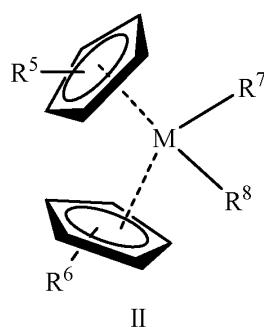


wherein (i) M is selected from Zr and Hf and (ii) R^1 , R^2 , R^3 , R^4 , R^5 , R^6 , R^7 and R^8 are each independently a C_1 - C_6 linear alkyl.

46. The method of claim 36, wherein the first precursor and the second precursor are each independently a precursor having Formula I:



or Formula II:



wherein (i) M is selected from Zr and Hf and (ii) R¹, R², R³, R⁴, R⁵, R⁶, R⁷ and R⁸ are each a methyl group.

47. The method of claim 36, wherein the method comprises an ALD process.
48. The method of claim 36, wherein the method comprises a CVD process.
49. The method of claim 36, wherein the deposition temperature is between approximately 200 °C and approximately 570 °C.
50. The method of claim 36, wherein the deposition temperature is between approximately 265 °C and approximately 500 °C.
51. The method of claim 36, wherein the deposition temperature is between approximately 280 °C and approximately 300 °C.
52. The method of claim 36, wherein the deposition temperature is below approximately 340 °C.
53. The method of claim 36, wherein the deposition temperature is approximately 300 °C.
54. The method of claim 36, wherein the substrate comprises silicon, germanium, group III-V materials, transition metal dichalcogenides, titanium nitride, titanium, tantalum, tantalum nitride,

tungsten, platinum, rhodium, molybdenum, cobalt, ruthenium, palladium, or mixtures thereof, or dielectrics like silicon oxide, silicon nitride, aluminum oxide, titanium oxide.

55. The method of claim 36, wherein the deposited crystalline material has a thickness of approximately 0.2 nm and approximately 20 nm.

56. The method of claim 36, wherein the deposited crystalline material exhibits remanent polarization without additional thermal processing.

57. The method of claim 36, wherein the deposited crystalline material has a remanent polarization (P_r) of greater than $8 \mu\text{C}/\text{cm}^2$ or a total loop opening of greater than $16 \mu\text{C}/\text{cm}^2$.

58. The crystalline material of any of claims 1-32, wherein there is hysteresis and remanent polarization in a polarization-electric field measurement.

59. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 0.2 nm to approximately 10 nm.

60. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 0.2 nm to approximately 5 nm.

61. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 0.2 nm to approximately 1 nm.

62. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 0.2 nm to approximately 0.5 nm.

63. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 15 nm or less.

64. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 10 nm or less.

65. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 5 nm or less.

66. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 3 nm or less.

67. A thin film comprising the material of any of claims 1-32, wherein the film has a thickness of approximately 1 nm or less.

68. A thin film comprising the material of any of claims 1-32 or claims 60-68, wherein the film has a remanent polarization (P_r) of greater than $8 \mu\text{C}/\text{cm}^2$ or a total loop opening of greater than $16 \mu\text{C}/\text{cm}^2$.

- 69.** The use of the thin film crystalline material of any of claims 1-32 for forming a thin film that exhibits ferroelectric behavior
- 70.** The use of the thin film of any of claims 60 to 69 as a ferroelectric material in a computing device.