Radius J. Sci. Technol.
Volume 1, Issue 1, (2024) 241004 (13pp)

ORIGINAL ARTICLE

Oxide Double Perovskites: Bridging the Gap in Photovoltaic and Thermoelectric Applications

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Received: 25 September 2024 / Revised: 03 October 2024 / Accepted: 09 October 2024 / Published Online: 27 October 2024

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Abstract: In response to the 20th-century surge in urban migration and the growing need for digital devices, global energy consumption witnessed a steady rise, leading to an energy crisis. To address this, the researcher has turned their attention to renewable energy sources, such as solar cells and thermoelectric generators. However, the efficiency and stability of these materials remain challenging, especially when constructed from toxic or less abundant elements. Oxide double perovskite materials have gained attention due to their tuneable properties and unique crystal structure, making them suitable for photovoltaic and thermoelectric applications. This study aims to study oxide double perovskite materials' structural, optical, and thermoelectric properties for exploring their potential in energy conversion applications. This comprehensive study not only contributes to oxide double perovskite materials but also paves the way for future research endeavours. The imperative for both experimental and theoretical exploration is emphasized to unlock the full potential of these compounds in the dynamic landscapes of optoelectronics and thermoelectric devices, fostering sustainable and efficient energy conversion technologies.

[Keywords: Photovoltaic applications; Thermoelectric applications; Renewable energy technologies; Perovskite solar cells (PSCs); Perovskite thermoelectrics]

1. Introduction:

The world faces a growing demand for energy, and this demand is increasing rapidly because of an unprecedented rise in the world's population. However, our current reliance on conventional fossil fuels, such as coal, oil, and natural gas, is unsustainable. These fossil fuels are finite resources, probably they will run out with respect to time. Moreover, the burning of fossil fuels as a primary energy source is a major contributor to greenhouse gas emissions, which are the primary drivers of climate change and global warming. Climate change leads to a host of devastating consequences, including more frequent and severe weather events, rising sea levels, and disruptions to ecosystems. Additionally, the extraction, processing, and burning of fossil fuels release harmful pollutants into the air and water **[1]**. This results in air pollution, habitat destruction, and water contamination, all of which have severe consequences for both the environment and human health. This finite nature of conventional fossil fuels and their

harmfulness to the global environment and health underscores the critical need to explore and embrace renewable energy resources like solar, wind, and hydropower **[2, 3]**. Unlike fossil fuels, these sources are essentially inexhaustible, as they rely on natural processes that will continue indefinitely and hence offer a sustainable solution to our energy needs.

Furthermore, renewable energy contributes to energy access and equity. By diminishing dependence on costly and polluting energy sources, it can provide electricity to remote and underserved areas, thus mitigating energy poverty and improving the quality of life in these communities, advancing the cause of equitable and inclusive energy access **[4]**. In this way, transitioning from fossil to renewable energy sources is not merely an option but a necessity for ensuring a sustainable and resilient future for building a more sustainable and prosperous world for generations to come. This study will unlock the full potential of these compounds in the dynamic landscapes of optoelectronics and thermoelectric devices, fostering sustainable and efficient energy conversion technologies.

2. Photovoltaic (PV) Technology: Applications, Advancements, and Challenges

PV technology, converting sunlight into electricity, has diverse applications **[5]**. In residences and businesses, solar panels provide cost-effective, sustainable power, while solar farms generate electricity for wider use. PV-powered water pumping aids in agriculture, and in space exploration, PV panels are crucial for satellites and space probes. Integrated into electric vehicles, PV contributes to energy efficiency. The adaptability of PV underscores its role in sustainable energy solutions across various scales and sectors. PV technology has evolved significantly over the years, with several generations of solar cells showing advancements in efficiency, cost-effectiveness, and versatility. An overview of the various generations of solar cells and their key advancements is shown in Figure 1 and described below:

2.1 First Generation Solar Cells (1950s - Present):

The first generation of solar cells predominantly consists of crystalline silicon solar cells **[6]**. These cells are still the most widely used photovoltaic technology today. The core of first-generation technology is the use of single-junction silicon solar cells, which are highly efficient in converting sunlight into electricity. First-generation silicon solar cells have seen continuous enhancements in efficiency. Monocrystalline silicon-based solar cells have now achieved a record conversion efficiency of 26.1%, approaching its theoretical Auger efficiency limit of 29.4% **[7]**. Other variations like polycrystalline silicon and thin-film silicon technologies have also emerged. However, they have slightly lower efficiency. First-generation solar cells are durable and can last for decades with minimal maintenance. These solar cells encounter drawbacks, including diminished efficiency and expensive raw materials. They experience reduced efficiency in high temperatures, leading to potential power loss. Current investigations prioritize the advancement of novel materials and technologies aimed at enhancing efficiency and decreasing production expenses.

2.2 Second Generation Solar Cells (1980s - Present):

In contrast, second-generation solar cells, developed in the 1980s and continuing to evolve, are also known as thin-film solar cells. They address some limitations of first-generation cells, particularly in terms of cost and flexibility. Notable types include amorphous silicon (a-Si) solar cells and cadmium telluride (CdTe) solar cells **[8]**, known for their affordability, higher

tolerance to temperature changes, and moderate efficiency, ranging from 10-21% **[9-11]**. These second-generation solar cells are often used in applications where flexibility, lightweight, and aesthetics are important, but these aren't very efficient, and their performance can get worse as time goes on. People even don't completely know about their lifetime or durability. They want to improve how well they work, how long they last, and how affordable they are so they can compete better with regular solar cells.

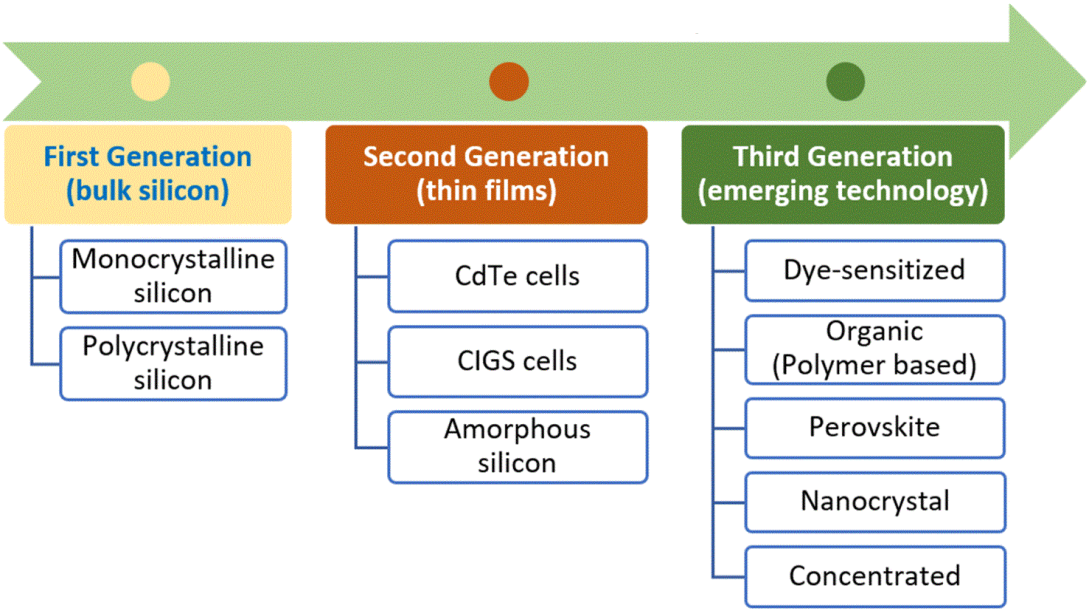

Figure 1: Categorizing Solar Cell technology advancements into three distinct generations **[18]**.

2.3 Third Generation Solar Cells (Emerging Technologies):

Third-generation solar cells are still in the research and development stage, and they aim to further improve efficiency and address some of the limitations of first and second-generation cells. Several promising technologies fall under this category, including organic photovoltaic (OPV) cells, leveraging organic materials for lightweight and flexible designs, even though with current efficiency is higher than previous generations **[11-14]**. Perovskite solar cells, based on perovskite materials, have shown rapid efficiency improvements and potential for surpassing traditional silicon cells while remaining cost-effective. Third-generation solar cells are expected to revolutionize the solar industry by offering higher efficiencies, lower costs, and greater versatility, but they are still undergoing research and development to make them commercially viable **[15]**.

2.4 Challenges of Photovoltaic Technology:

In the landscape of photovoltaic (PV) technology, its numerous advantages coexist with challenges impeding widespread adoption and efficacy **[16, 17]**. The reliance on sunlight introduces intermittency, prompting the need for robust energy storage solutions to address fluctuations in power generation. Despite cost reductions, the upfront investment for PV systems poses a significant hurdle, particularly in economically constrained regions. Environmental concerns arise from the production and disposal of PV panels, emphasizing the necessity for strides in sustainable manufacturing and recycling practices. Persistent efforts are directed towards enhancing the efficiency of PV panels, driving exploration into advanced materials, and innovative design strategies. As the share of solar power grows, grid integration

complexities surface, demanding the implementation of sophisticated management strategies. Overcoming these challenges, ranging from intermittency and cost to environmental impact, efficiency, and grid integration, mandates a continual commitment to research and innovation. This dedication is indispensable to unlock the full potential of PV technology and effectively address the escalating global energy demands.

3. Thermoelectric Devices: Advancements, Applications, and Challenges

Recent statistical data reveals that over 65% of the energy generated in various processes, such as burning fossil fuels or operating nuclear fission power plants, is wasted as heat energy (Figure 2) **[18]**. Thermoelectric (TE) materials stand out as effective solutions for recovering waste heat while minimizing environmental impact since they can convert waste heat into usable carbon-free electrical energy. A thermoelectric material is essentially a substance that captures heat and directly converts it into electricity, and the device in which TE compounds are employed for this purpose is referred to as a thermoelectric generator. TEGs represent environmentally friendly energy conversion devices with notable advantages over conventional methods.

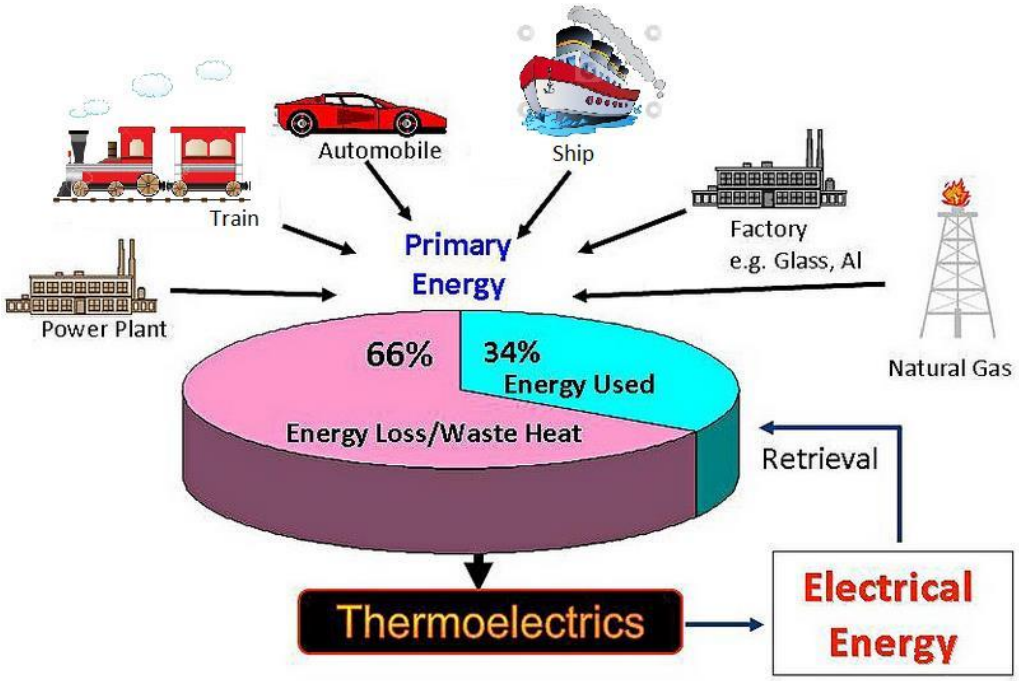

Figure 2: Illustration depicting the proportion of energy dissipated and energy utilized in different methods of energy generation. *Source:* *waste heat recovery technologies | Turbomachinery blog (softinway.com)*

TEGs operate based on two fundamental principles: the Seebeck Effect and the Peltier Effect. The Seebeck effect allows them to generate a voltage when two dissimilar materials are connected at different temperatures, leading to the conversion of heat into electricity **[19]**. Conversely, the Peltier effect enables these devices to create a temperature difference at junctions when an electric current is applied **[20]**. These effects can be understood more deeply with the help of Figure 3. For optimal functioning, thermoelectric materials must possess specific properties. Semiconductors are the primary materials used in these devices, including bismuth telluride (Bi_2Te_3), lead telluride (PbTe), and silicon-germanium alloys **[22-24]**. These

materials should exhibit high electrical conductivity to maximize the Seebeck effect, coupled with low thermal conductivity to minimize heat transfer and maintain a temperature gradient.

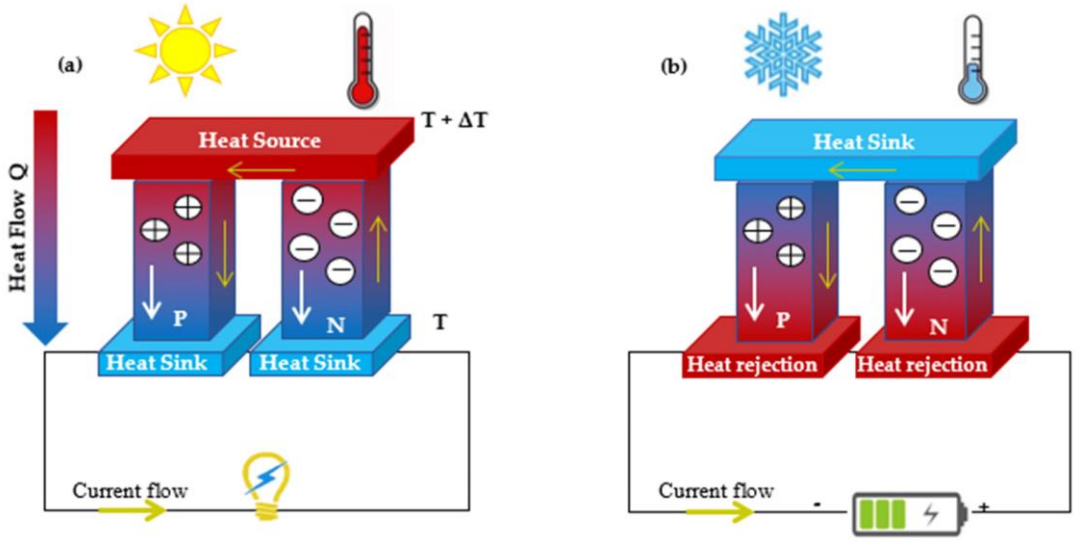

Figure 3: (a) Thermoelectric Generator (Seebeck Effect), and **(b)** Thermoelectric Refrigeration (Peltier Effect) Categorizing Solar Cell technology advancements into three distinct generations **[21]**.

In recent years, thermoelectric generators (TEGs) have undergone notable advancements, ushering in a new era of energy harvesting and waste heat utilization. Researchers have focused on developing advanced thermoelectric materials with superior efficiency, especially at higher temperatures. Nanostructured materials are designed to enhance thermoelectric properties **[25, 26]**. The use of nanocomposites and nanostructuring has facilitated better compatibility between different thermoelectric materials, optimizing their collective performance **[26, 27]**. These generators have been designed to harness energy from body heat, paving the way for applications in wearable electronics and medical devices **[28, 29]**. Research efforts have been directed toward designing TEGs capable of extracting energy from sources like industrial waste heat or ambient environmental heat. Moreover, TEGs have been integrated with other energy harvesting technologies, such as solar cells and piezoelectric devices, creating hybrid systems capable of extracting energy from multiple sources. The commercial landscape has witnessed a surge in interest and investment in thermoelectric devices such as portable electronic devices, waste heat recovery, harnessing energy from automotive exhaust, powering remote areas, and even space missions using radioisotope thermoelectric generators **[28-33]**.

Despite these advancements and vast applications, several challenges persist in the widespread adoption of thermoelectric devices. One key challenge is the need for materials with a high thermoelectric figure of merit (ZT), balancing the often-competing factors of electrical conductivity and thermal conductivity. Additionally, cost-effective and scalable manufacturing processes are crucial to make thermoelectric technology economically viable for various applications. Hence, efficiency improvements have been a constant focus, encompassing factors such as material efficiency, system design, and considerations related to heat transfer. However, researchers and industry players are increasingly focused on environmentally friendly materials and manufacturing processes, ensuring that the sustainability and life cycle impact of thermoelectric generators aligns with contemporary environmental standards.

This study is primarily focused on investigations of oxide-based double perovskite materials and characterising them to know their potential for photovoltaic and thermoelectric applications. The studied materials have helped in manufacturing photovoltaic and thermoelectric devices such as solar cells, LED, photodetectors, medical imaging, perovskite lasers, Neuromorphic devices, solar panels, security cameras, bar code scanners, thermoelectric generators, etc.

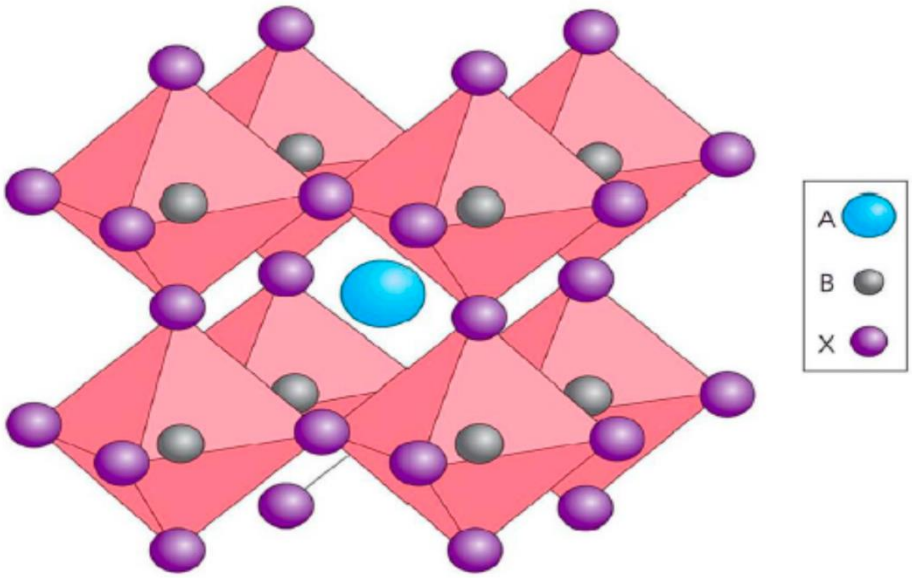

Figure 4: A cubic crystal perovskite composition ABX_3 **[34]**.

4. Perovskite Materials: History and Advancements

The term "perovskite" originates from the Russian mineralogist Lev Perovski, who first discovered a mineral Calcium Titanate with the chemical formula $CaTiO_3$ in the Ural Mountains of Russia in 1839 **[34]**. This mineral was named perovskite in his honour. It belongs to a large mineral family characterized by a cubic crystal structure in its ideal form. The general formula for a perovskite compound is ABX_3, where 'A' represents a large metal cation (ion with a positive charge) located at the centre of the cubic unit cell, 'B' is a smaller cation that occupies the corners of the unit cell, and 'X' is an anion (ion with a negative charge) that surrounds the central 'A' cation, forming octahedra with the 'B' cations at the corners (Figure 4) **[34]**. In the chemical formula of perovskite (ABX_3), when this chemical structure is used in photovoltaic applications like solar cells, A represents typically small organic or inorganic molecular cations, B is represented by metallic ions like lead, germanium or tin, and X is denoted by corresponding halides.

4.1 Types of Perovskite Materials:

4.1.1 Natural Perovskites:

These are perovskite minerals that are found naturally in the Earth's crust. While the perovskite crystal structure is quite common in nature, it can be found in various combinations of elements. Some natural perovskite compounds include:

a) **Perovskite ($CaTiO_3$):** It is the original perovskite mineral composed of calcium titanate and discovered by Lev Perovski. It is widely found in igneous rocks and can have various impurities, which may give it different colours.
b) **Bridgmanite ($(Mg,Fe)SiO_3$):** It is a high-pressure perovskite mineral that constitutes a significant portion of Earth's lower mantle.
c) **Loparite:** This is a natural perovskite mineral that contains rare earth elements and is often used as a source of these elements, which have important applications in various technologies, including electronics and green energy.
d) **Knopite:** It is another natural perovskite mineral, knopite, is composed of lead titanate ($PbTiO_3$).
e) **Stetindite:** This mineral is a natural perovskite containing strontium titanate ($SrTiO_3$).

These natural perovskite compounds have various properties and are of scientific and economic interest due to their unique combinations of elements and crystal structures.

4.1.2 Synthetic Perovskites (Hybrid Perovskites):

These are perovskite-like compounds engineered for specific applications through chemical synthesis. These are perhaps the most well-known perovskite materials in the context of photovoltaics. They also have the general formula ABX_3, where 'A' can be an organic cation, typically methylammonium ($CH_3NH_3^+$), formamidinium ($HC(NH_2)_2^+$), or other organic molecules, 'B' is typically a metal cation, such as lead (Pb^{2+}), tin (Sn^{2+}), or a combination of both, and 'X' is an anion, usually iodide (I^-) or bromide (Br^-) **[35-37]**.

4.1.3 Other Variants of Perovskites:

a. Double Perovskites:

Double perovskites, a subset of perovskite materials with the general formula $A_2BB'X_6$, deviate from the traditional ABX_3 structure by replacing the single B-site cation with two distinct cations, B and B'. This alteration allows for diverse and tuneable properties. The exploration of double perovskite oxides had its origins in 1961 when Longo and Ward **[38]** reported the ferromagnetic behaviour of Re-based double perovskites. Subsequent advancements were gradual until 1998, when Kobayashi *et al.* **[39]** published findings on the half-metallic properties of Sr_2FeMoO_6, where strontium, iron, and molybdenum contribute to intriguing magnetic and electronic behaviours, reigniting interest in this category of compounds. Furthermore, the demonstration of half-metallicity in Sr_2FeReO_6 was subsequently affirmed through first-principles calculations of the density of states **[40]**. The double perovskite compounds have demonstrated enhanced thermal stability and non-toxic properties, making them promising materials for efficient solar cells **[41, 42]**. The relationship between perovskite and double perovskite structures can be observed in Figure 5.

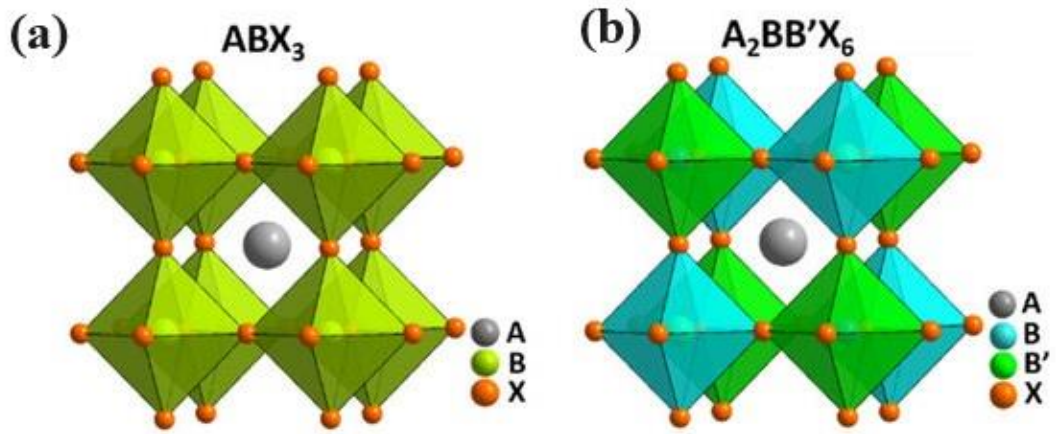

Figure 5: (a) Perovskite – ABX_3 and **(b)** Double Perovskite – $A_2BB'X_6$ structures.

b. Anti-Perovskites:

Anti-perovskites are represented by the ABX_3 structure which is a reversal in the general formula, leading to the occupation of X sites by an electropositive ion (a cation like an alkali metal). Simultaneously, A and B sites are filled with distinct types of anions. The term "anti" in its nomenclature indicates a reversal of the typical order of cations and anions compared to regular perovskite structures.

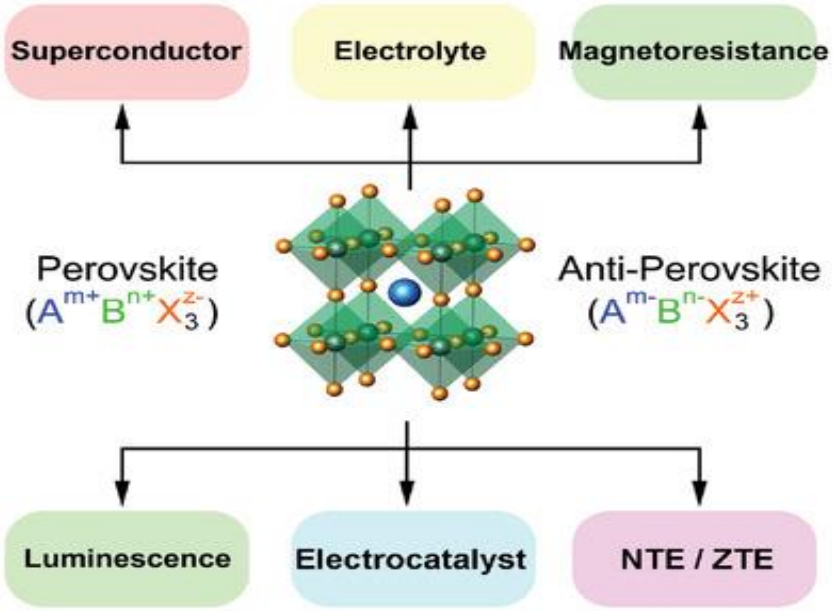

Figure 6: Antiperovskites with Exceptional Functionalities.

In an ideal cubic cell, the A anion is positioned at the corners of the cube, the B anion is situated at the octahedral centre, and the X cation is found at the faces of the cube (Figure 6) **[43]**. The unique arrangement offers potential advantages in electronic and ionic conductivity, making anti-perovskites promising for innovative materials and devices **[44]**. A number of factors are there, responsible for affecting the stability of antiperovskite structure. One of the factors is phase transitions where starting reactants are responsible for affecting the structure of antiperovskites. Kinetically prevention factor also plays a key role in forming the structure. These types of compounds are found to be hygroscopic as they react with water results the

formation of another compound. The presence of other elements in the structure results in more stability of the compound.

c. Double Anti-Perovskites:

These materials combine the characteristics of double perovskites and anti-perovskites. They have the general formula $A_2BB'X_6$, where A is a monovalent anion, B and B' are divalent anions, and X is a cation. This arrangement gives these materials interesting properties, and they are being explored for various applications, including as potential candidates for solid-state electrolytes in batteries. These properties make them promising candidates for use in advanced electronic devices and energy storage systems **[45]**.

5. Stability Criteria:

Solar cells must function effectively in various conditions, withstanding atmospheric elements like moisture, light, oxygen, and water, as well as internal factors such as heating degradation due to material properties **[46]**. The primary concern in the perovskite sector currently revolves around long-term instability, posing challenges to the swift discovery of materials **[47]**. The Goldschmidt tolerance factor, denoted as 't', is widely utilized to predict perovskite structure stability based solely on the chemical formula, represented by,

$$t = (R_A + R_x) / \sqrt{2}(R_B + R_X)$$

Here, R_A, R_B, and R_X correspond to the ionic radii of A, B, and X ions, offering insights into the crystal structure symmetry of perovskites. A t value of 1 signifies the ideal cubic perovskite structure, while $0.89 < t < 1$ indicates octahedral tilting, lowering the symmetry.

The octahedron factor μ, calculated as R_A/R_X, is crucial in predicting the stability of the MX_3 (where M = Metal) octahedron in metal halide perovskite formation. If $\mu > 0.442$, octahedra may form **[48]**. For a stable antiperovskite structure, the tolerance factor must fall between 0.71 and 1. Tolerance factor values between 0.71 and 0.9 suggest an orthorhombic or tetragonal crystal, while values between 0.9 and 1 indicate a cubic structure. Table 1 represents the Tolerance Factor (t) and Corresponding Perovskite/Anti-perovskite Structures.

Table 1: Tolerance Factor (t) and Corresponding Perovskite/Anti-perovskite Structures

Tolerance factor (t)	Perovskite / Anti-perovskite structures
$t = 1$	Ideal cubic perovskite structure
$0.89 < t < 1$	Octahedral tilting
$0.71 < t < 1$	Stable anti-perovskite structure
$0.71 < t < 0.9$	Orthorhombic or tetragonal
$0.9 < t < 1$	Cubic structure

The stability and reactivity of compounds are strongly influenced by the formation energy. When compared to higher formation energy, lower formation energy signifies a stable and less reactive compound. There occurs an issue with the long-term stability of double perovskite oxides that can be avoided by adopting a B-site doping strategy. Elements such as Ti, Nb, Mo, and Cr can be implemented for this particular purpose. The passivating perovskite grains method can be adopted to avoid the stability problem. The effective method which is known as minimizing water adsorption technique can be taken into account to get rid of the stability issue.

6. Limitations of Double Perovskite Oxides:

The power conversion efficiency for these compounds is found to be lower than that of lead-based perovskite solar cells. Most of the double perovskite oxides show large bandgaps generally above 2 eV. Most double perovskites with 3D crystal structures have lower dimensional electronic structures. Chemical defects are carried by these compounds generally. The B-site sublattice in double perovskite can cause spin frustration. In- and Tl-based double perovskites with direct bandgaps can experience parity-forbidden transitions. A more focused exploration into enhancing the efficiency of perovskite materials through innovative synthesis techniques, targeted doping strategies, bandgap engineering, or structural modifications. Additionally, a concerted effort to bridge the gap between theoretical predictions and practical applications by leveraging computational methods, high-throughput calculations, and machine learning approaches could accelerate the discovery of highly stable and highly efficient perovskite materials for energy conversion technologies.

7. Conclusions:

In essence, the domain of oxide double perovskites shows immense promise across renewable energy, catalysis, spintronics, and thermoelectric applications owing to their eco-friendly characteristics. Oxide-based double perovskites have also gained significant attention in the field of photovoltaic applications due to their excellent light-absorption capabilities and potential for highly efficient solar cells. These materials continue to intrigue researchers due to their diverse properties and potential to address critical challenges within clean energy and advanced technological fields. The discovery and exploration of perovskite materials have resulted in a rich array of compounds with diverse structures and properties. Despite their environmentally friendly nature, these perovskites face challenges such as low electrical conductivity and high thermal conductivity, limiting their widespread utilization in devices. Recent advancements in oxide double perovskites bring hope for more efficient devices, yet their efficiency still trails behind traditional materials. Although progress in structural modifications and computational modelling enhances their potential, further research in synthesis and customized designs is essential for their widespread integration into sustainable energy technologies.

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