Japan Academy Prize to:

Tsutomu Miyasaka
Specially Appointed Professor,
Toin University of Yokohama

for "Development of Organo-Metal Perovskite-Based Organic-Inorganic Hybrid Solar Cells"

Outline of the work:

Dr. Tsutomu Miyasaka has made a significant contribution to the field of solar energy by discovering organo-lead halide compounds as absorbers for perovskite solar cells, which exhibit energy conversion efficiencies comparable to those of the most efficient crystalline silicon solar cells. His research focuses on the development of new perovskite materials to enhance efficiency and device stability. Unlike commonly known metal oxides, the perovskite crystals utilized in solar cells are metal halides with an ABX_3 composition, typically comprising an organic-inorganic composite compound $CH_3NH_3PbX_3$ (where Pb denotes lead). These ionic crystals can easily form semiconductor films when the source liquid solution is coated on an electrode substrate and dried.

In 2009, Dr. Miyasaka published the inaugural report on photoelectric conversion using metal halide perovskites in *Journal of American Chemical Society*, marking a pivotal moment in the field. Subsequent research, including a 2012 paper in Science, demonstrated that a solidified layered structure of perovskite photovoltaic cells could achieve conversion efficiencies exceeding 10%. This groundbreaking work spurred global research efforts, leading to rapid improvements in efficiency. In recognition of his influential contributions, Clarivate Analytics (formerly Thomson Reuters) honored Miyasaka with the Citation Laureates award in 2017 for his highly cited papers, and in 2022, he received the Rank Prize in the UK.

Halide perovskites are distinguished by their exceptional optical properties and a physical characteristic known as "defect tolerance," which substantially mitigates defect-assisted charge recombination. This property ensures a high output voltage for power generation, thereby enabling high conversion efficiency. Dr. Miyasaka has demonstrated that perovskite crystal thin films possess efficient light-emitting properties, a phenomenon derived from this unique physical characteristic (*Chemistry Letters*, Chemical Society of Japan, 2012). By early 2018, the conversion efficiency of perovskite-based solar cells had surpassed those of compound semiconductor solar cells, such as Cu-In-Ga-Se (CIGS) and CdTe, achieving efficiencies over 26.0% and matching the market-leading crystalline silicon cells.

Dr. Miyasaka's key research achievements include:

I. During the solution-coating process to form a photovoltaic layer of perovskite polycrystalline thin film, he explored crystallization conditions to enhance crystal quality. This led to a film-forming method in an ambient humid atmosphere, increasing conversion efficiency to over 20% (Paper. No. 15).

II. He developed a defect passivation technology that protects carrier transfers against interfacial defects by forming a self-assembled organic molecule layer at the interface. This approach achieved nearly 23% efficiency and a voltage output approaching the theoretical limit (Paper. No. 3).

III. The focus on efficiency enhancement extended to all-inorganic perovskite compositions with high thermal stability. A cell using $CsPbI_2Br$ as perovskite, modified at the interface to minimize energy loss, achieved a high voltage of over 1.4 V. This cell demonstrated an efficiency exceeding 34% under low-intensity indoor lighting (Paper. No. 8).

IV. In collaboration with JAXA, Dr. Miyasaka has initiated research on the space applications of perovskite solar cells, revealing their high durability against high-energy radiation (electron and proton beams) in the space environment (Paper. No. 16). This finding has prompted numerous research institutes worldwide to validate its reproducibility.

In 2012, following the achievement of perovskite solar cells surpassing 10% efficiency, Peccell Technologies, Inc., a university venture company founded by Dr. Miyasaka in 2004, filed for 12 patents, setting a precedent in the field. Most of these patents have been granted.

Halide perovskite crystals offer several industrial advantages over silicon and other semiconductor materials used in solar cells, not only in terms of high conversion efficiency but also in various other aspects. These advantages have attracted numerous companies, both in Japan and internationally, towards the commercialization of perovskite-based technologies:

(1) The solution coating (printing) method used for film formation is a low-cost and high-speed process. The synthetic raw materials, including lead and iodine, are inexpensive and can be sourced domestically. The cost of perovskite semiconductor film is approximately 200 JPY/m^2, which is about 1/20 that of silicon.

(2) Since the process does not require high temperatures (below 120℃), perovskite photovoltaic films can be deposited on plastic substrates, facilitating the creation of lightweight and flexible devices.

(3) With a high optical absorption coefficient (10^5/cm), the absorber functions effectively with thicknesses less than 1 μm, significantly thinner than crystalline silicon absorbers, enabling the production of thin and highly flexible devices.

(4) The bandgap energy of perovskites is tunable by altering the halogen composition, allowing for the adjustment of edge wavelengths. This property is crucial for developing multi-junction tandem cells that require wavelength optimization.

(5) The optical transparency of perovskite films enables the creation of semi-transparent solar cells, paving the way for bifacial power generation devices that absorb light from both sides.

(6) The defect tolerance of perovskite semiconductors ensures high conversion efficiency even under weak light conditions. This capability allows for power generation not only outdoors on sunny days but also on cloudy or rainy days, and even under indoor lighting.

Dr. Miyasaka has shown that a key factor in enhancing the efficiency of solar cells is the solution-based crystallization process, which improves the quality of perovskite polycrystalline films. Optimizing particle size and achieving a dense, flat crystalline film with minimal physical defects can significantly reduce structural defects and increase efficiency. Japan's industrial sector possesses strong technological capabilities in this chemical process. Therefore, Dr. Miyasaka's pioneering work in the field of perovskite solar cells is expected to significantly contribute to the realization of a carbon-neutral society and is deserving of the Japan Academy Prize.

List of Main Publications

(1) "Halide perovskite for indoor photovoltaics: The next possibility", Z. Guo, A. K. Jena, and T. Miyasaka. *ACS Energy Lett.*, 8, 90–95 (2023).

(2) "A semitransparent silver-bismuth iodide solar cell with Voc above 0.8 V for indoor photovoltaics", N. B. C. Guerrero, Z. Guo, N. Shibayama, A. K. Jena, and T. Miyasaka. *ACS Appl. Energy Mater.*, 6, 10274–10284 (2023).

(3) "Phenethylamine-based interfacial dipole engineering for high Voc triple-cation perovskite solar cells", G. M. Kim, H. Sato, Y. Ohkura, A. Ishii, and T. Miyasaka. *Adv. Energy Mat.*, 12, 2102856 (2022).

(4) "$FAPbBr_3$ perovskite solar cells with VOC over 1.5 V by controlled crystal growth using a tetramethylenesulfoxide", Y. Numata, N. Shibayama, and T. Miyasaka. *J. Mater. Chem. A*, 10, 672–681 (2022).

(5) "The high open-circuit voltage of perovskite solar cells: a review", Z. Guo, A. K. Jena, G. M. Kim, and T. Miyasaka. *Energy Environ. Sci.*,15, 3171–3222 (2022).

(6) "Dopant-free polymer HTM-based $CsPbI_2Br$ solar cells with efficiency over 17% in sunlight and 34% in indoor light", Z. Guo, A. K. Jena, I. Takei, M. Ikegami, A. Ishii, Y. Numata, N. Shibayama, and T. Miyasaka. *Adv. Functional Mat.*, 31, 2103614 (2021).

(7) "Artemisinin-passivated mixed-cation perovskite films for durable flexible perovskite solar cells with over 21% efficiency", L. Yang, Q. Xiong, Y. Li, P. Gao, B. Xu, H. Lin, X. Li and T. Miyasaka. *J. Mater. Chem. A*, 9, 1574–1582 (2021).

(8) "VOC over 1.4 V for amorphous tin-oxide-based dopant-free $CsPbI_2Br$ perovskite solar cells", Z. Guo, A. K. Jena, I. Takei, G. M. Kim, M. A. Kamarudin, Y. Sanehira, A. Ishii, Y. Numata, S. Hayase, and T. Miyasaka. *J. Am. Chem. Soc.*, 142, 9725–9734 (2020).

(9) "MACl–assisted Ge doping of Pb–Hybrid perovskite: A universal route to stabilize high perovskite solar cells", G. M. Kim, A. Ishii, and T. Miyasaka. *Adv. Energy Mat.*, 1903299 (2020).

(10) "Lead(II) propionate additive and a dopant-free polymer hole transport material for $CsPbI_2Br$ perovskite solar cells", S. Oez, A. K. Jena, A. Kulkarni, K. Mouri, T. Yokoyama, I. Takei, F. Uenlue, S. Mathur, and T. Miyasaka. *ACS Energy Lett.*, 5, 1292–1299 (2020).

(11) "Femto- to microsecond dynamics of excited electrons in a quadruple cation perovskite", E. Jung, K. Budzinauskas, S. Oez, F. Uenlue, H. Kuhn, J. Wagner, D. Grabowski, B. Klingebiel, M. Cherasse, J. Dong, P. Aversa, P. Vivo, T. Kirchartz, and T. Miyasaka, P. H. M. van Loosdrecht, L. Perfetti, and S. Mathur. *ACS Energy Lett.*, 5, 785–792 (2020).

(12) "Halide perovskite photovoltaics: background, status, and future prospects", A. K. Jena, A. Kulkarni, and T. Miyasaka. *Chem. Rev.*, 119, 3036–3103 (2019).

(13) "Perovskite solar cells: Can we go organic-free, lead-free, and dopant-free?", T. Miyasaka, A. Kulkarni, G. M. Kim, S. Oez, and A. K. Jena. *Adv. Energy Mat.*, 1902500 (2019).

(14) "Performance enhancement of $AgBi_2I_7$ solar cells by modulating a solvent-mediated adduct and tuning remnant BiI_3 in one-step crystallization", A. Kulkarni, A. K. Jena, M. Ikegami, and T. Miyasaka. *Chem. Comm.*, 55, 4031–4034 (2019).

(15) "Stabilizing the efficiency beyond 20% with a mixed cation perovskite solar cell fabricated in ambient air under controlled humidity", T. Singh, and T. Miyasaka. *Adv. Energy Mat.*, 8, 1700677 (2018).

(16) "Tolerance of perovskite solar cell to high-energy particle irradiations in space environment", Y. Miyazawa, M. Ikegami, H.-W. Chen, T. Ohshima, M. Imaizumi, K. Hirose, and T. Miyasaka. *iScience*, 2, 148–155 (2018).

(17) "Ambient fabrication of 126 μm thick complete perovskite photovoltaic device for high flexibility and performance", T. Singh, M. Ikegami, and T. Miyasaka. *ACS Appl. Energy Mater.*, 1, 6741–6747 (2018).

(18) "Microstructural analysis and optical properties of the halide double perovskite $Cs_2BiAgBr_6$ single crystals", O.A. Lozhkina, A.A. Murashkina, M.S. Elizarov, V.V. Shilovskikh, A.A. Zolotarev, Yu.V. Kapitonov, R. Kevorkyants, A.V. Emeline, and T. Miyasaka. *Chem. Phys. Lett.*, 18–22 (2018).

(19) "Sulfate-assisted interfacial engineering for high yield and efficiency of triple cation perovskite solar cells with alkali-doped TiO_2 electron-transporting layers", T. Singh, S. Oz, A. Sasinska, R. Frohnhoven, S. Mathur, and T. Miyasaka. *Adv. Funct. Mater*, 1706287 (2018).

(20) "Highly efficient and stable low-temperature processed ZnO solar cells with triple cation perovskite absorber", J. Song, L. Liu, X.-F. Wang, G. Chen, W. Tian, and T. Miyasaka. *J. Mater. Chem. A*, 5, 13439–13447 (2017).

(21) "Controlled crystal grain growth in mixed cation-halide perovskite by evaporated solvent vapor recycling method for high efficiency solar cells", Y. Numata, A. Kogo, Y. Udagawa, H. Kunugita, K. Ema, Y. Sanehira, and T. Miyasaka. *ACS Appl. Mater. Interfaces*, 9, 18739–18747 (2017).

(22) "Poly(4-Vinylpyridine)-based interfacial passivation to enhance voltage and moisture stability of lead halide perovskite solar cells", B. Chaudhary, A. Kulkarni, A. K. Jena, M. Ikegami, Y. Udagawa, H. Kunugita, K. Ema, and T. Miyasaka. *ChemSusChem*, 10, 2473–2479 (2017).

(23) "Towards stable and commercially available perovskite solar cells", N. G. Park, M. Grätzel, T. Miyasaka, K. Zhu, and K. Emery. *Nature Energy*, 1, 16152 (2016).

(24) "Effect of electron transporting layer on bismuth-based lead-free perovskite $(CH_3NH_3)_3$ Bi_2I_9 for Photovoltaic Applications", T. Singh, A. Kulkarni, M. Ikegami, and T. Miyasaka. *ACS Appl. Mater. Interfaces*, 8, 14542–14547 (2016).

(25) "Low-temperature-processed ZnO-SnO_2 nanocomposite for efficient planar perovskite solar cells", J. Song, E. Zheng, X.-F. Wang, W. Tian and T. Miyasaka. *Solar Energy Mater. Solar Cells*, 144, 623–630 (2016).

(26) "High performance perovskite solar cell via multi-cycle low temperature processing of lead acetate precursor solutions", T. Singh, and T. Miyasaka. *Chem. Commun.*, 52, 4784–4787 (2016).

(27) "Steady state performance, photo-induced performance degradation and their relation to transient hysteresis in perovskite solar cells", A. K. Jena, A. Kulkarni, M. Ikegami, and T. Miyasaka. *J. Power Sources*, 309, 1–10 (2016).

(28) "Low-temperature SnO_2-based electron selective contact for efficient and stable perovskite solar cells", J. Song, E. Zheng, J. Bian, X. F. Wang, W. Tian, Y. Sanehira, and T. Miyasaka. *J. Mater. Chem. A*, 3, 10837–10844 (2015).

(29) "Perovskite photovoltaics: Rare functions of organo lead halide in solar cells and optoelectronic devices", T. Miyasaka. *Chem. Lett.*, 44, 720–729 (2015).

(30) "The interface between FTO and the TiO_2 compact layer can be one of the origins to hysteresis in planar heterojunction perovskite solar cells", A. K. Jena, H.-W. Chen, A. Kogo, Y. Sanehira, M. Ikegami, T. Miyasaka. *ACS Appl. Mater. Interfaces*, 7, 9817–9823 (2015).

(31) "Excitonic feature in hybrid perovskite $CH_3NH_3PbBr_3$ single crystals", H. Kunugita, T. Hashimoto, Y. Kiyota, Y. Udagawa, Y. Takeoka, Y. Nakamura, J. Sano, T. Matsushita, T.Kondo, and T. Miyasaka, *K. Ema. Chem. Lett.*, 44, 852–854 (2015).

(32) "Efficient perovskite solar cell fabricated using an aqueous lead nitrate precursor", T.-Yu Hsieh, T. C. Wei, K.-L. Wu, M. Ikegami, and T. Miyasaka. *Chem. Commun.*, 51, 13294–13297 (2015).

(33) "Efficient and environmentally stable perovskite solar cells based on ZnO electron collection layer", J. Song, J. Bian, E. Zheng, X.-F. Wang, W. Tian, and T. Miyasaka. *Chem. Lett.*, 44, 610–612 (2015).

(34) "A switchable high-sensitivity photodetecting and photovoltaic device with perovskite absorber", H.-W. Chen, N. Sakai, A. K. Jena, Y. Sanehira, M. Ikegami, K.-C. Ho, and T. Miyasaka. *J. Phys. Chem. Lett*. 6, 1773–1779 (2015).

(35) "Emergence of hysteresis and transient ferroelectric response in organo-lead halide perovskite solar cells", H. W Chen, N Sakai, M Ikegami, and T Miyasaka. *J. Physical Chem. Lett.*, 6, 164–169 (2014).

(36) "Fully crystalline perovskite-perylene hybrid photovoltaic cell capable of 1.2 V output with a minimized voltage loss", A. Ishii, A. K. Jena, and T. Miyasaka. *APL Materials*, 2, 091102 (2014).

(37) "Quantum conversion enhancement with TiOx compact layer for ITO plastic film-based low-temperature-processed dye-sensitized photoelectrode", S. Morita, M. Ikegami, T.-C. Wei, and T. Miyasaka. *ChemPhysChem*, 15, 1190–1193 (2014).

(38) "Tri-functional Nb_2O_5 nano-islands coated on an indium tin oxide layer for a highly efficient dye-sensitized plastic photoanode", S. Morita, T.-C. Wei, M. Ikegami, and T. Miyasaka. *J. Power Sources*, 240, 753–758 (2013).

(39) "Efficient hybrid solar cells based on meso-superstructured organometal halide perovskites", M. M. Lee, J. Teuscher, T. Miyasaka, T. N. Murakami, and H. J. Snaith. *Science*, 338, 643–647 (2012).

(40) "Highly luminescent lead bromide perovskite nanoparticles synthesized with porous alu-

mina media", A. Kojima, M. Ikegami, K. Teshima, and T. Miyasaka. *Chem. Lett.*, 41, 397–399 (2012).

(41) "A high voltage organic-inorganic hybrid photovoltaic cell sensitized with metal-ligand interfacial complexes", A. Ishii, and T. Miyasaka. *Chem. Commun.*, 48, 9900–9902 (2012).

(42) "Organometal halide perovskites as visible-light sensitizers for photovoltaic cells", A. Kojima, K. Teshima, Y. Shirai, and T. Miyasaka. *J. Am. Chem. Soc.*, 131, 6050–6051 (2009).

(43) "Low-temperature fabrication of dye-sensitized plastic electrodes by electrophoretic preparation of mesoporous TiO_2 layers", T. Miyasaka, and Y. Kijitori. *J. Electrochem. Soc.*, 151, A1767–A1773 (2004).

(44) "The photocapacitor: An efficient self-charging capacitor for direct storage of solar energy", T. Miyasaka and T. N. Murakami. *Appl. Phys. Lett.*, 85, 3932–3934 (2004).

(45) "Tin-based amorphous oxide. A high-capacity lithium storage material", Y. Idota, T. Kubota, A. Matsufuji, Y. Maekawa, and T. Miyasaka. *Science*, 276, 1395–1397 (1997).