Transparent conductors—materials that are simultaneously transparent and electrically conductive—are essential to modern semiconductor and optoelectronic technologies. These compounds are rare and have typically been realized by starting from a wide-band-gap insulator and introducing heavy doping to render the material conductive. Although this strategy is used in commercial devices, it makes it difficult to achieve both very high conductivity and sufficiently high optical transparency. Motivated by this limitation and by the early seminal works of Zunger *et al.* [Phys. Rev. Lett. 115, 176602, 2015] and Engel-Herbert *et al.* [Nat. Mater. 15, 204, 2016], we aim to develop an alternative approach: start from a gapped metal and make it transparent. However, in contrast to the above works, which mainly were proof of concept studies and limited to pristine stochiometric compounds, we aim to demonstrate that the properties of gapped metals can be widely tuned through controllable off-stoichiometry. Specifically, we posit that gapped metals—compounds whose Fermi level lies in the principal conduction band and that possess a large internal gap between principal band edges—can exhibit characteristically negative cation- or anion-vacancy formation enthalpies, resulting in spontaneous non-stoichiometry. Such spontaneous non-stoichiometry can occur even at low temperatures and is intrinsic to the compound, rather than a growth artifact. We expect this behavior to be generic: in n-type gapped metals, cation-vacancy formation allows conduction-band electrons to decay into acceptor states; the resulting negative electron–hole recombination energy can offset the positive energy cost of creating a vacancy by breaking chemical bonds. This Fermi-level-induced spontaneous non-stoichiometry can yield stable, non-stoichiometric compounds with distinct optoelectronic properties that are stable under a range of conditions. In this way, appropriate synthetic control can be used to produce transparent conductors with targeted transparency, conductivity, and stability. We note that high-performance transparent conductors require: (a) metals with a large internal gap between the valence and conduction bands; (b) a sufficiently high carrier density in the conduction band to provide electrical conductivity; and (c) a sufficiently low carrier density to limit the plasma frequency so that free-electron absorption does not compromise optical transparency. Furthermore, (d) the free carriers in the conduction band above the internal gap should not destabilize the compound by spontaneously creating Fermi-level-induced defects that suppress conductivity. Because such compounds cannot be reliably discovered by the trial-and-error Edisonian approach, we will perform fundamental theoretical analysis—namely, first-principles calculations—to identify design criteria for a new generation of transparent conductors. Our specific focus will be to understand off-stoichiometry in gapped metals and to determine which compounds can be synthesized. Importantly, this work may clarify which compounds are exceptions to the classical Daltonian paradigm (the law of definite proportions) and may open new applications for gapped metals.