

Semiconducting polymer LEDs

by David Braun

Technologies based on organic semiconductors may answer the increasing demands that consumers make in the areas of large-area electronics, lightweight displays, and portable computing. Advances in scientific understanding, technology, and device performance have occurred particularly rapidly in the area of polymer light-emitting diodes (LEDs). Material properties and economic considerations suggest that polymer LEDs are the devices most likely to win the race to applications that produce light on inorganic substrates such as glass and silicon, as well as plastic substrates.

The field of semiconducting polymers has its root in the 1977 discovery of the semiconducting properties of polyacetylene¹. This breakthrough earned Alan Heeger, Alan MacDiarmid, and Hideki Shirakawa the 2000 Nobel Prize in Chemistry for 'the discovery and development of conductive polymers'²⁻⁵. Other review articles capture how more than two decades of developments in the physical and chemical understanding of these novel materials has led to new device applications as active and passive electronic and optoelectronic devices ranging from diodes and transistors to polymer LEDs, photodiodes, lasers, and solar cells⁶⁻¹¹. Much interest in plastic devices derives from the opportunities to use clever control of polymer structure combined with relatively economical polymer synthesis and processing techniques to obtain simultaneous control over electronic, optical, chemical, and mechanical features⁵. This article focuses on the advances leading to polymer LEDs¹²⁻¹⁴.

Polymers as light sources

Perhaps the most exciting possibilities stem from the prospect of polymers as active light sources. In 1990, Burroughes and co-workers at the University of Cambridge discovered that a semiconducting polymer can also exhibit electroluminescence^{15,16}. This finding added polymers to the repertoire of organic luminescence originating in the 1987 work by Tang and van Slyke of Kodak Corporation^{17,18}.

Polymer LED devices consist of several layers of polymer materials sandwiched between a metallic cathode and an inorganic or organic anode, such as indium tin oxide (ITO) or a conducting polymer. Fig. 1 shows the cross section of a polymer LED and illustrates the enticing simplicity.

Improved performance

Device operation depends on device fabrication. Polymer LED fabrication usually begins with a transparent and insulating substrate coated with a transparent conductor. A spin-coating step deposits one or more polymer layers onto the substrate. A top metal electrode is applied using a vacuum evaporation process. The entire procedure is conceptually quite simple yet creates numerous possibilities. In part because polymer LED research is just plain fun, many research groups began to explore the prospect of making and improving polymer LEDs. One feature of electroluminescent polymers has a surprising effect at the instant that someone first views the visible light emission from one of these devices. Functioning devices have *macroscopic* dimensions on the order of square millimeters to square centimeters²³. The resulting light-emitting devices are *tangible*, and large enough to motivate some viewers to smile (Fig. 2) and, frequently, excite the viewer to pursue further study.

Immediately following the Cambridge announcement, the state-of-the-art began to advance quite rapidly as researchers around the world responded to the new opportunities. Using an electroluminescent polymer that is soluble *in the semiconducting state* (Fig. 3a) demonstrated device fabrication without the use of any high temperature

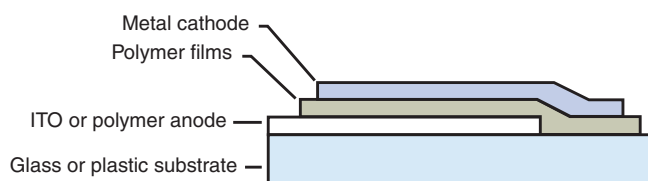


Fig. 1 Polymer or organic LED geometry. Applying a positive voltage to the anode relative to the cathode causes current to flow through the polymer film and light emission from the polymer film through the transparent bottom electrode and substrate. Devices made on plastic substrates can have flexible mechanical properties¹⁹⁻²².

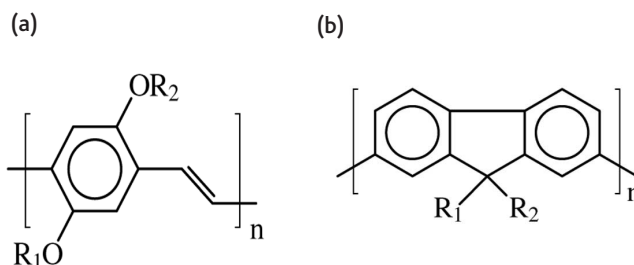


Fig. 3 Structures of two electroluminescent polymers (a) Poly(dialkoxy p-phenylene-vinylene) [dialkoxy-PPV]²⁵⁻²⁷ (b) Poly(dialkylfluorene) [DA-PF]^{12,28,29}.

processing, thereby allowing device preparation on flexible substrates such as plastic foils incapable of withstanding high temperatures^{19,20}. Secondly, choosing a lower work function cathode increased device quantum efficiencies above 1% (photon/electron) and reduced the threshold voltage necessary to generate light visible in well-lit ambient conditions^{19,20}. Such improved device performance finds guidance in bandgap engineering techniques used primarily in inorganic heterojunction devices. The energy band diagrams in Fig. 4 illustrate the operation of a single layer polymer LED. A calcium cathode produces more efficient



Fig. 2 Polymer LEDs made by Gang Yu, 5" x 5" and 5.1" diameter, to commemorate the 2000 Nobel Prize. These single pixel polymer LEDs are fixed digital images with a resolution of 80 dpi. (Courtesy of Alan Heeger and Gang Yu of DuPont Displays.)

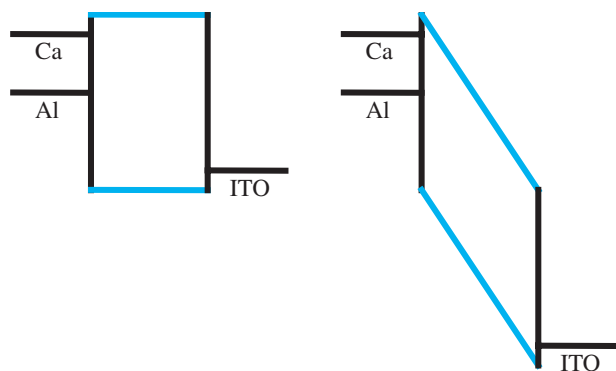


Fig. 4 Energy band diagrams of a single layer polymer LED showing the emitting layer at flatband (left) and under higher forward bias (right). Notice that potential barriers impede injection of both polarity carriers, and no potential barrier helps to confine carriers to the luminescent layer. A low work function cathode such as Ca leads to a smaller potential barrier for electron injection than an air stable cathode such as Al.

electroluminescence than an aluminum cathode, because its lower work function creates a lower potential barrier for electrons injected into the conduction band of the polymer. Other alkaline earth metals also improve electron injection²⁴.

Within four years³⁰ of the Cambridge discovery, polymer LEDs became available across the visible spectrum^{15,19,31-39}, with dramatically improved efficiencies^{20,40,41}, and on flexible substrates^{20,21}. In the meantime, polymer LEDs have advanced further in efficiency^{27,42,43}, devices emit all colors from the infrared to the ultraviolet^{7,44-46}, and can generate white light efficiently⁴⁷. Transparent devices exist^{48,49}, and polymer LEDs can emit polarized light^{50,51}. It is very likely that polymer LED displays will reach the market this year. Several publications and the references they contain provide a particularly complete snapshot of the rapidly advancing state-of-the-art^{7,12,13,22,52-55}.

Organic heterostructures for everything: device principle of the 2000s?

Advances in polymer LEDs originated in the desire to merge the many device prospects developed in the field of compound inorganic semiconductors with the growing repertoire of semiconducting polymers. Bandgap engineering traditionally applied to the inorganic materials uses careful crystal growth to obtain fine control of electronic and optical properties. The 2000 Nobel Prize in Physics recognizes the importance and impact of heterostructures⁵⁷. It is perhaps no coincidence that the same year's Nobel Prize in Chemistry recognizes the importance and impact of semiconducting polymers⁵. The title of this section, based on Kroemer's

similarly titled 1981 missive regarding inorganic semiconductors⁵⁶ hints that polymers present new options. Clever manipulation of chemical structure not only allows control of electronic and optical properties, but also enables independent control of chemical and mechanical features. The union of the two disciplines has yet to exhaust its promise. This section describes some fruits of collaborations between chemical synthesis, polymer processing, and bandgap engineering.

Metal/polymer junctions formed on soluble semiconducting polymers establish that soluble polymers can form diodes and permit exploration of basic device physics^{58,59}. By virtue of solubility, a polymer may have device performance comparable to less tractable relatives and require considerably less manipulation in post-synthesis processing. Novel applications and better device performance may result from taking advantage of the processing flexibility allowed by solubility and other modifications of the polymer. Understanding basic elements of device operation such as charge injection and transport can guide device engineers as they design and construct the devices, and relating the device physics to polymer properties can guide chemists to synthesize new materials with the desired capabilities.

Improved understanding of transport

Initial experiments suggested that tunneling injection from the electrodes primarily determined device efficiency^{19,60}. A decade of thoughtful analysis and careful experiments studying transport for devices of varying thickness as a function of temperature and electrode work function reveal more complex tradeoffs of temperature-dependent injection, trap assisted tunneling, hopping, and bipolar space-charge-limited processes⁶¹⁻⁷³.

The device structure pictured in Fig. 5 incorporates more features than currently realized with semiconducting polymers, and, therefore, represents a long term goal. For vacuum deposited molecules, the concepts of ETHB (electron transport and hole blocking layer) and HTEB (hole transport and electron blocking layer) were proposed by Tang and Van Slyke and extended by Adachi and Tsutsui in order to improve carrier injection from the electrodes and confine the carriers to the emission layer^{17,74}. The current versus voltage characteristics of LEDs made with the polyfluorene derivative F8BT (Ca/F8BT/PEDOT-PSS/ITO) show evidence of HTEB behavior and high electroluminescence efficiency⁷⁵.

Molecular electronic heterostructures have also been built with Langmuir-Blodgett deposition⁷⁶.

The extension proposed in Fig. 5 would further enhance carrier injection from the electrodes by means of graded bandgaps. The case shown has linearly graded bands, but even stepwise grading should work⁷⁷. With molecular beam epitaxy (MBE), gradual or statistical variations of layer composition may be used to realize the graded bands. Opportunities still remain to optimize the balance of carrier injection, mobility, and confinement to improve device performance. Brédas and others explain how chemical structure modifications influence electronic structure and band positions⁷⁸. Applying such knowledge will likely enable the engineering of optimal band diagrams similar to those portrayed in Fig. 5. For example, the polymer analog of statistical bandgap grading based on molecular self-assembly to improve the anode/polymer interface leads to improved device performance^{79,80}.

Statistical variations could arise from interlayer mixing. Other precedents exist in polymer processing. For example, graded index polymer optical fibers use gradual variations in index. For multilayers prepared using spin-coating, some degree of interlayer mixing can occur at the interfaces between the layers. The spin-coating process consists of at least two steps: applying a volume of the polymer in a solvent to the substrate and spinning the substrate to produce the thin and uniform polymer film. When applying a solution of polymer 2 (dissolved in solvent 2) on top of a film of polymer 1, solvent 2 will penetrate the film of polymer 1 to an extent determined by the solubility of polymer 1 in solvent 2. If polymer 1 has the function of *assisting* carrier injection into polymer 2, intermixing would tend to smooth out the interface, reduce the abruptness of the potential barrier that carriers encounter, and aid in the carrier injection process. If, however, polymer 1 has the function of *confining* carriers within polymer 2, an abrupt junction would be desired to maintain a potential barrier that impedes carrier flow. Suitable control of film morphology resulting from the polymer deposition process remains a challenge⁸¹.

Dramatic improvements in device performance results from optimizing cathode and anode interfaces. Metal fluoride layers at the cathode offer improved device efficiency⁸³, and ultrathin (<45 Å) layers of alkaline earth metals inserted at the cathode also improve operating lifetime^{24,84}. Molecules such as 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,2,4-

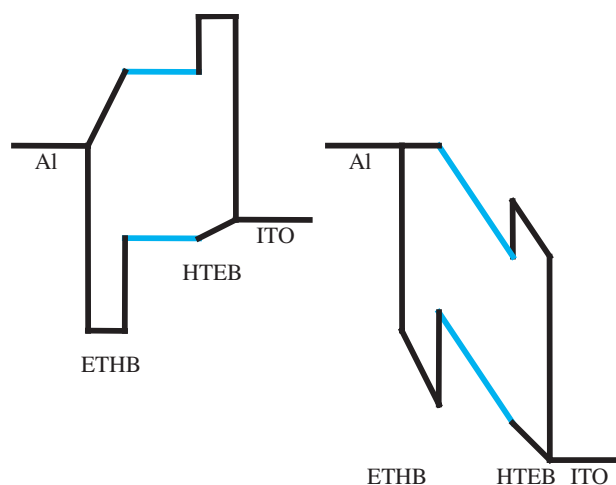


Fig. 5 Energy band diagrams of a multi-layer polymer LED showing the emitting layer at flatband (left) and under higher forward bias (right). ETHB refers to an Electron Transport and Hole Blocking Layer, and HTEB refers to a Hole Transport and Electron Blocking Layer. This design makes use of energy gap grading to assist carrier injection from the electrodes into the emitting layer. Proper selection of the ETHB valence band energy and the HTEB conduction band energy produces potential barriers that confine injected carriers to the emitting layer.

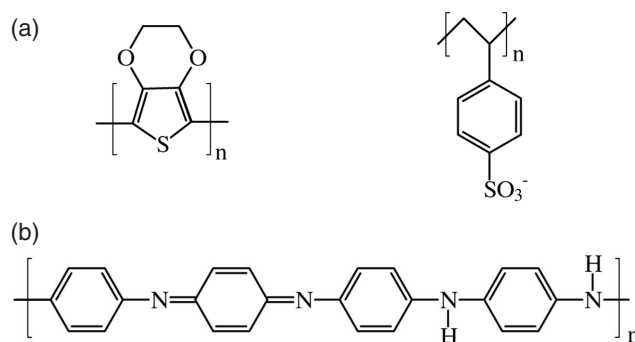


Fig. 6 Structures of polymer anodes (a) Poly(3,4-ethylenedioxythiophene)/Poly(styrenesulfonate) [PEDOT-PSS]⁹² and (b) Poly(aniline) [PANI].

oxadiazole (PBD)⁸⁵⁻⁸⁷ and surfactants such as anionic ether sulfates⁸⁸ can function as electron transport layers (ETLs) to improve electron injection from the cathode, even with air stable cathodes. Polymer ETLs also improve efficiency^{41,89}. Conducting polymers can function as the anode alone²¹ or assist hole injection from an ITO anode. Fig. 6 shows the structures of two popular polymer hole transport layers (HTLs), PANI^{90,91} and PEDOT-PSS⁹², that improve operating efficiency and operating lifetime⁹³. Fig. 7 contains the resulting high performance polymer LED characteristics. Similarly, blending polyfluorene (Fig. 3b) with HTL molecules can improve efficiency⁹⁴. Other triphenylamine copolymers with PPV⁹⁵ or DA-PF⁹⁶ can improve carrier mobility and LED performance.

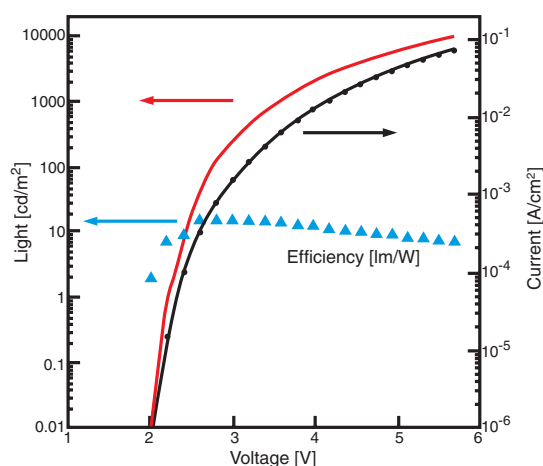


Fig. 7 High performance polymer LED characteristics for yellow PPV derivative. Brightness, power efficiency, and current versus applied voltage. (Data courtesy of Hermann Schenk and Heinrich Becker of Covion Organic Semiconductors.)

Doping adds an additional degree of freedom to increase performance. Doping applied to organic LEDs can increase electroluminescence efficiency and stability under stress¹⁸. More evidence comes from doping semiconducting polymers with mobile ions to improve carrier injection and transport of heterojunction LEDs with doped carrier confinement and injection layers⁹⁷. These light-emitting electrochemical cells (LEECs) demonstrate that appropriately chosen dopants can permit efficient electron injection even from relatively air-stable cathodes without sacrificing luminescence efficiency. Suitable injection and confinement layers would improve on the LEEC concept with non-mobile dopant species placed in the desired layer of the device. A LEEC based on the multilayer device geometry would be another approach to speed up the LEEC devices whose temporal response depends on moving ionic species.

Doping the emitting polymer layer with suitable materials offers control of color, can produce narrow bandwidth luminescence⁹⁸, and quite bright and efficient devices⁹⁹. The use of electrophosphorescence in the emitting layer offers extremely efficient luminescence^{100,101}, with power efficiencies above 40 lm/W reported^{42,102} and up to 80 lm/W expected¹⁰³. In addition to the successful polymers in Fig. 3, new materials become available¹⁰⁴⁻¹⁰⁶.

Increased efficiencies and lifetime support display applications

Initially considered a question mark large enough to stop the polymer LED show, understanding of device degradation

under operation has enabled improved device uniformity, reliability, and operating lifetimes above the 10 000 hours required for many applications. The polymer degrades via photo- or electro-oxidation^{29,107-109} and polymer chain reorientation¹¹⁰. As researchers identify specific polymer defects, they design new more efficient and robust polymers^{27,29}. As devices degrade¹¹¹, black spots form caused by cathode pinhole defects^{112,113} and electromigration^{114,115}, and light output decreases as the voltage needed to supply a constant current increases¹¹⁶. The polymer/ITO interface is unstable¹¹⁷, and shelf life is a concern¹¹⁸. Nevertheless, careful packaging can protect the sensitive, low work function cathodes sufficiently to obtain long operating lifetimes and shelf life¹¹⁹ even for large area (8 cm²) LEDs²³.

Substantial progress toward high efficiency LEDs and better understanding of application-specific and systems-level operation has made higher resolution matrix displays feasible. Polymer LEDs switch on and off fast enough for video display applications, limited by RC time constants associated with pixel and electrode geometry¹²⁰. The transient response sheds light on carrier transport^{121,122}. Pioneer has demonstrated a 5.2" quarter VGA passive matrix color OLED display with 320 (x3) x 240 pixels¹²³. Higher resolution displays require active matrix solutions with two, three or four transistors per pixel¹²⁴⁻¹²⁶ for acceptable power dissipation¹²⁷, uniformity¹²⁸, and crosstalk¹²⁹.

Active matrix circuits use transistors made from single crystalline silicon, poly-crystalline silicon, amorphous silicon, or conducting polymers^{130,131}. eMagin and Semiconductor Energy Laboratory have made active matrix microdisplays¹³². Consider also the integration of electroluminescent polymers with silicon circuits^{133,134} or polymer circuits¹³⁰. For optical communications, a polymer LED on a silicon substrate could couple light into an optical waveguide coupled to a photo-detector¹³⁵. Kodak/Sanyo demonstrated a 2.4" diagonal active matrix OLED display with a resolution of 852 x 222 pixels¹³⁶. DuPont Displays (Fig. 8) and Semiconductor Energy Laboratory¹²⁴ have made 4" diagonal prototype active matrix OLED displays with VGA resolution. Samsung SDI and Universal Display have announced a 8.4" SVGA color active matrix OLED display¹³⁷, and Sony has demonstrated a 13" diagonal prototype active matrix OLED display with a resolution of 800 x 600 pixels²².

Ink-jet printing allows patterning of injection or emitting polymer layers¹³⁷⁻¹⁴¹, with industrial programs announced by

Seiko-Epson¹⁴², CDT¹⁴², Philips¹³⁷, Toshiba, and others²². Roll-to-roll display fabrication on plastic substrates is under development¹³⁷, and a low temperature (60°C) lamination process has been demonstrated¹⁴³. Other printing techniques applied to passive matrix polymer LED displays include dye diffusion¹⁴⁴, stamped dye diffusion¹⁴⁵, screen printing¹⁴⁶⁻¹⁴⁸, spray coating, and laser-induced thermal imaging¹³⁷. Polymer LEDs can also work as backlights for LCD displays, for example in a smartcard¹⁴⁹.

Many device features received and continue to receive careful attention as polymer LEDs approach widespread commercialization. The improved quality of newer materials has allowed more reproducible experimentation and higher performance device characteristics¹⁵⁰. Companies including Covion Organic Semiconductors and Dow Chemical sell high-quality luminescent polymers in large industrial quantities¹⁵¹ and groundbreaking industrial development of polymer LED materials continues at Cambridge Display Technology, Covion Organic Semiconductors, Dow Chemical, and DuPont^{152,153}. Other groups are working to overcome technical hurdles and commercialize this technology^{44,52,154}. Ultra-flat ITO anodes with RMS surface roughness below 1 nm reduce leakage currents and improve device efficiency¹⁵⁵. ITO surface treatments including UV ozone¹⁵⁶ and oxygen plasma¹⁵⁷⁻¹⁶¹ improve performance. To reduce reflections of ambient light entails a circular polarizer or other optical films^{162,163}, and

other optical techniques can improve external brightness¹⁰³. Note that caveats concerning device efficiencies require careful attention to measurement protocol¹⁶⁴.

Conclusion

Semiconducting polymers benefit from versatility and economy: enormous versatility in terms of structure, properties, and, ultimately, applications; economies in terms of their cost in large scale production; and their light weight in transportation and use. In some ways, the polymers can ease the implementation of projects that push inorganic semiconductor technology to its limits. For example, quasi-one-dimensional structures such as quantum wires can be fabricated in inorganic semiconductors, usually by means of fairly sophisticated techniques, so that semiconductor real estate costs dollars per square micron. Taking advantage of organic chemistry means that synthesis of advanced materials such as intrinsically quasi-one-dimensional semiconductor chains can take place literally in a pot on a hot plate for dollars per square meter.

Semiconducting polymers blend several attractive material properties that enable inexpensive and unique applications. They offer the ability to tailor electronic and optical properties, as do many inorganic semiconductors. They also offer simultaneous control over mechanical and chemical features. This article has reviewed how semiconducting

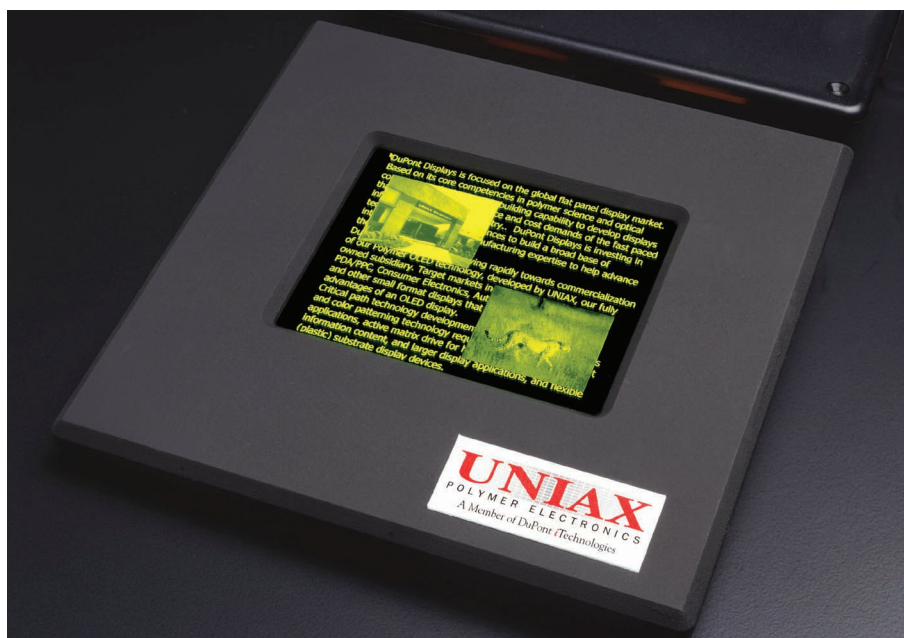


Fig. 8 Diagonal 4" VGA monochrome active matrix polymer LED display with a resolution of 200 dpi and 6 bit grey scale. (Courtesy of Alan Heeger and Gang Yu of DuPont Displays.)

polymer LEDs enable large area lighting and multimedia displays. Semiconducting polymers also have the potential to deliver the next generation of flat and flexible image sensors¹⁶⁵, printed integrated circuits^{130,166}, lasers¹⁶⁸, superconductors¹⁶⁷, and inexpensive solar cells¹⁶⁹. The progress described in this article combined with the already known and yet to be discovered properties of the semiconducting polymers add yet one more argument to the

compelling case that our society should not burn up so much petroleum. Materials derived from petroleum have much more appealing and unique uses. **MT**

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