Combinatorial Search and Optimization of New ZnO-based Phosphors

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Combinatorial synthesis and screening of very large numbers of organic compounds has been widely applied in the pharmaceutical industry for drug discovery [1]. More recently, combinatorial arrays (libraries) of inorganic materials with known or potential superconductivity [2] or giant magnetoresistance [3] or photoluminescence [4-6] have been synthesized and screened. Thus, the combinatorial method, which implies discovery of new materials and optimization of their properties by high-throughput synthesis and consequent screening, is becoming a key technology in materials science today.

The combinatorial method promises to be especially fruitful in those multi-component systems where the properties are difficult to predict theoretically. Phosphors, with their diversity of possible mechanisms of luminescence, represent a typical example of such systems. It was demonstrated in our earlier works [6, 7] that integrated materials libraries having different dopants in different concentrations can be manufactured by combining pulse laser deposition (PLD) with a masking system. Unlike other methods, the PLD allows one-step fabrication of the libraries.

The pulse laser deposition method was employed in this work to fabricate thin-film combinatorial libraries of ZnO-based phosphors on different substrates. Fabrication of both pixel libraries and spread libraries has been accomplished – see Fig. 1. Screening of the libraries included examination of both photoluminescence (PL) and low-voltage cathodoluminescence (CL) properties. The measurements were done through the pixels of different compositions or through the spread with continuous concentration change.

Combinatorial exploration of twelve different binary and ternary ZnO: dopant systems in a broad range of dopant concentrations resulted in identification of bright luminescence in ZnO: (Y, Eu), ZnO: V, ZnO: W and ZnO:(W, Mg) systems.

The ZnO: (Y, Eu) emitted in red (611 nm in PL, 618 nm in CL), ZnO: V emitted in yellow (550-560 nm at different dopant concentrations), ZnO: W emitted in blue (475-500 nm at different dopant concentrations) and ZnO:(W, Mg) emitted in blue (465-500 nm at different dopant concentrations).

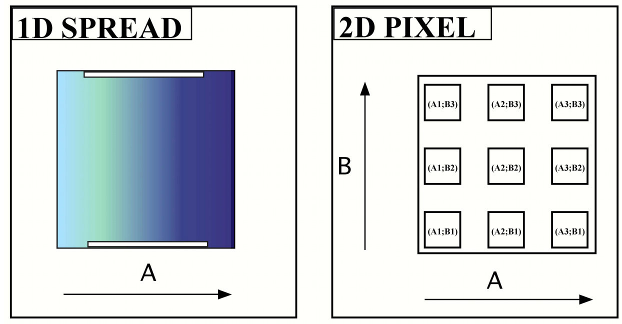

Fig. 1. Two simplest designs of combinatorial libraries. In the 1-dimensional spread design concentration of component A increases continuously along one axis. In the 2-dimensional pixel design concentration of components A and B increases stepwise, forming a 2-dimensional array of pixels. The libraries of both types have been fabricated in this work.

Careful “zooming in” i.e. fabrication and screening of more detailed libraries near the identified promising compositions revealed concentration dependencies of CL and PL luminescence efficiencies and emission wavelengths, which in turn allowed us to find optimum phosphor compositions for the above-mentioned systems.

The efficiency of the newly discovered and optimized blue ZnO: (W, Mg), blue ZnO: W, red ZnO: (Y, Eu) and yellow ZnO: V phosphors in low-voltage cathodoluminescence is high and allows their prospective use in advanced flat panel display and lighting applications.

References

1 Bunin B.A., Plunkett M.J., Ellman J.A., Proc. Natl Acad. Sci. USA 9 (1996) 4798.
2 Xiang X.D., Sun X., Briceno G., Lou Y., Wang K.A., Ckang H., Wallaca-Friedman W.G., Chen S.W., Schultz P.G., Science 268 (1995) 1738.
3 Briceno G., Chang H., Sun X., Schultz P.G., Xiang X.D., Science 270 (1995) 273.
4 Danielson E., Golden J.H., MacFarland E.W., Reaves C.M., Weinberg W.H., Wu X.D., Nature 389 (1997) 944.
5 Wang J., Yoo Y., Gao C., Takeuchi I., Sun X., Chang H., Xiang X.D., Schultz P.G., Science 279 (1998) 1712.
6 Mordkovich V. Z., Jin Zhengwu, Yamada Y., Fukumura T., Kawasaki M., Koinuma H., Solid State Sciences 4 (2002) 779.
7 Matsumoto Y., Murakami M., Jin Z., Ohtotomo A., Lippmaa M., Kawasaki M., Koinuma H., Jpn J Appl Phys 38 (1999) L603.