RARE-EARTH ION DISTRIBUTION IN SOL-GEL GLASSES CO-DOPED WITH Al$^{3+}$

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For many applications it is essential that rare earth (RE) doped sol-gel glasses have high fluorescence yields, and therefore several mechanisms of fluorescence quenching in these materials must be overcome. We investigate interactions among RE ions that depend strongly on inter-ion distance and are exacerbated in sol-gels because dopants tend to cluster. Co-doping sol-gel glasses with Al$^{3+}$ improves RE fluorescence yield remarkably, and the generally accepted explanation for the past decade has been that Al$^{3+}$ disperses RE dopants in the matrix, reducing ion-ion energy transfer. Recently, numerical works [1,2] have attempted a detailed understanding of the role that Al$^{3+}$ plays. The first of these studies suggests the glass has regions of relatively high Al$^{3+}$ and RE concentration, compared to the rest of the network that is mostly undoped silica. This picture is different from earlier ideas, and requires reconsideration of previous evidence for Al$^{3+}$ dispersing RE clusters.

In this study, we use Tb$^{3+}$ to probe the effects of RE-RE interactions in sol-gel glass. We study energy transfer between Tb$^{3+}$ ions with pulsed laser experiments and use an analysis approach based on the Inokuti-Hirayama method. Assuming a multipolar interaction, we fit the decay curves to derive an effective local concentration for a series of samples with varying amounts of RE. When actual doping concentration is varied over two orders of magnitude, the effective local Tb concentration changes by about a factor of 10 (Fig. 1). Our results indicate that Al$^{3+}$ co-doping is only effective at dispersing RE ions when the ratio of Al:RE is 10:1 or greater. This result is consistent with ref. 2 but contradicts earlier work that used fluorescence line narrowing to demonstrate RE dispersal at much higher doping levels.


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Fig. 1. Effective concentration near emitting Tb$^{3+}$ centers is lower than sample concentration for doping levels above ~0.2%.