

# Review of: "Analysis method of binary concentration-inhomogeneous systems"

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**Potential competing interests:** No potential competing interests to declare.

I would like to explain further about this manuscript, from a modern perspective. My task as a reviewer is to seek for the least foundation upon which the manuscript makes sense. And then comment on the same foundation.

At least, the three mathematical equations made sense. But these were not considered new by the author. So, my understanding about this work should move further starting on this.

Then the question comes what the  $C(x, T)$  is, in eq. (1). It is clear from the manuscript that  $C(T)$  in general, is the measured heat capacity curve against temperature, as perhaps provided in DSC. This is also implied by Fig. 1.

Continuing looking at Fig. 1 I found the curves like the typical results of glass transition. The  $\alpha$ 's denotes results of different composition of a binary system. I got the idea from this figure that the author believed samples of different composition would exhibit different DSC curves during glass transition like this. This is the first argument I encountered.

For a binary mixture to show only one  $T_g$ , it must be perfectly miscible. Fortunately, the author is talking about the problem of bulk radical polymerization of MMA, a typical system where the polymerized product and the monomer are miscible. So, this question is passed.

But, as Prof. Godovsky had asked decades ago, why should the heat capacities of homogeneous and inhomogeneous system of the same averaged composition differ? This is difficult to answer in the 1990s. The theory of glass transition back in the 1990s had not fully developed to provide a general functional form for the heat capacity curve. As we understand now, even for a homogeneous liquid, the behavior of heat capacity jump during glass transition depends heavily on the thermal history, and the effect of physical aging. Another characteristic of glass-forming liquid, the fragility parameter, is also not theoretically clear. But fragility affects the height as well as the span of the heat capacity jump. Although fragility is less tunable for single component mixture, it depends on composition for the case of mixtures. Apologies for not having the time to provide supporting references for these statements, but I am sure the current research community of glass transition agrees with these. To sum up, the variation of the heat capacity vs temperature curves depends on more factors than just the composition.

Even if unique correspondence between composition and  $C(x, T)$  can be established, the problem of ill-posedness, as also noticed by the author, is nontrivial.

All these criticisms, however, are about possible practical complexity that may render the proposed method useless. The

logic of the proposed method itself makes sense. As a complete work, the author should have done experiments to justify or remove the worry in the practical aspects. A small note which only proposed an idea might not be acceptable for formal publication even in the standard in the 1990s. It is understandable why most of the other reviewers consider this manuscript not enough for publication. (I know it is now not practical for the author, but I comment only for the sake of fairness)

However, I would like to provide yet another encouraging argument. Glass transition has now been understood as the percolation of dynamically cooperative regions. Length-scales of dynamic heterogeneity diverges in general during glass transition. Therefore, in the modern sense the glass transition process is all about heterogeneity--not the heterogeneity in composition, concentration, etc., but rather the spatial correlation of fast or slow dynamics, a concept which includes but is wider than the heterogeneity in concentration/density. The intuition of the author to look for heterogeneity when considering the glass transition problem makes a lot of sense even today, and even excellent in the 1990s. Of course we all hate the ill-posedness nature of Fredholm integral equations, but we some how face them on a daily basis when we deal with soft condense matter in almost all aspects: viscoelasticity, non-Fickian diffusion, generalized Langevin equation, and of course the heterogeneous static and dynamic properties of liquids. An idea should not be killed just because of its ill-posedness when it comes to the study of glass transition.

A lot of scientific contributions with great originality by Russian scientists were buried in non-English Russian journals. In this manuscript all references are in Russian. Regretfully I am not the generation of Chinese who study Russian as the second language. Therefore I cannot comment on the fairness of citation in this manuscript.