Letter to the Editor

Response to Cantù et al.

The criticisms presented in the preceding letter from Cantù et al. are essentially attributable to a serious misunderstanding of the structural parameters shown by Hirai et al. (1996a). This is evidently derived from some unsuitable expressions in a part of the structural parameters (scattering densities) in the above paper, which should be corrected by Hirai et al. Such corrections of the expressions do not affect the contents of the above paper, since the results in the above paper were derived from concrete scattering data analyses and modeling method. Those corrections can answer all criticisms of Cantù et al. as follows.

As shown by Hirai et al. (1996a), our analyses were carried out by using Eq. 6 on the basis of a reasonable model, namely a double-shelled ellipsoid of rotation composed of a shell and a core. Clearly we measured the relative scattering intensities experimentally and used Eq. 6 for a profile fitting of the experimental scattering curve calibrated in relative scale. As is well known for solution small-angle scattering (Stuhrmann and Miller, 1978), only a difference between the average scattering densities of the solute and solvent contributes to effective scattering intensities. This is also evidently shown in Eq. 6 in the above paper (Hirai et al., 1996a), namely, that Eq. 6 contains average excess scattering densities (contrasts) of model components (shell and core), not average scattering densities of them. Therefore, evidently relative values of contrasts have physical meanings and change profiles of model scattering functions obtained by Eq. 6. When we had carried out the shell-modeling analyses, we had given the average scattering density of the solvent to be 1, which does not mean the normalization of scattering density by that of the solvent owing to the above reason. If we subtract 1 from the values of the scattering densities shown in the paper, the relative values of contrasts can be obtained. The alternation of the expression from the average scattering densities to the contrasts should be done by Hirai et al., since this change does not affect other structural parameters and results obtained from the optimized models, which is ensured by the expression of Eq. 6.

In addition, we showed very well agreements of the simulated scattering curves with the experimental ones by using our prolate double-shelled ellipsoid models, not by using a hard sphere or a hard ellipsoid, which have also clearly shown in our other papers (Hirai et al., 1996b, c). To confirm and show the validity of the structural parameters of our obtained model, we also compared several kinds of structural parameters obtained from the model scattering function with those from the experimental one by using gyration radii, distance distribution functions, deviation factors, and empirical values estimated from geometrical considerations. Those additional comparisons successfully indicated that the structural parameters obtained by using our shell-modeling analysis are very reasonable and self-consistent.

The comment on the distance distribution analysis has no meaning since in the report (Hirai et al., 1996) the distance distribution were obtained from the experimental and modeling scattering functions independently. This comment is also solved by the above correction in the expression from the scattering densities to the contrasts.

Thus the major part of criticisms in the letter clearly results from the values of the scattering densities shown by M. Hirai et al. After the above correction is done by Hirai et al., the criticisms made by Cantù et al. will be fully answered, and will have no more meaning.

The warning of a possibility of an oblate micellar structure of gangliosides can be eliminated by a simple modeling, as shown in a standard textbook (Glatter and Kratky, 1982) and by fairly good agreement of the model with experimental results in the report (M. Hirai et al., 1996b, c). In our cases (Hirai et al., 1996a, b, c) the micellar shapes must be an prolate shape, since we showed very well agreements of the simulated scattering curves with the experimental ones by using their prolate double-shelled ellipsoid models, not by using a hard sphere or a hard ellipsoid.

The above warning would be based on the results shown by the authors of the preceding letter. A similar conformational change of ganglioside micelles was reported by the authors (Corti et al., 1996), where they showed four x-ray scattering curves at two different temperatures and presented some structural parameters of an oblate ellipsoidal structure of the G_{M1} micelle in comparison with their previous results. In the above paper they did not explicitly show the scattering function and condition in the modeling scheme; however, they may have used a scattering function similar to Eq. 6 (Hirai et al., 1996a). Unfortunately, they did not concretely show any validity of the structural parameters of their models by comparing several kinds of structural parameters obtained from the model scattering functions with those from the experimental ones, which is very important for readers to confirm the validity of a presented model as shown by Hirai et al. They only showed the model scattering functions with the experimental ones in a normal plot, and a significant deviation between the model and experimental scattering functions can be recognized by readers in the q range below 0.05 $Å^{-1}$, which reflects mostly the micellar shape. Such an inconsistency could have been checked simply by calculating the gyration radius

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and distance distribution function from the model and experimental scattering functions.

There is no doubt that the shell-modeling method is a very powerful tool for reasonably determining the internal structure of micelles, especially for a monodispersed system such as ganglioside systems, when based on the concrete scheme of the shell-modeling method.

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