Evaluation of the Hydration State of Polyethylene Glycol Solutions by Wide-range Dielectric Spectroscopy.

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***Abstract*—Hydration in aqueous polyethylene glycol (PEG) solutions with various molecular weights was evaluated using a broadband dielectric spectroscopy (100 MHz – 18 THz). With the increase of the molecular weight, the restriction strength of hydration waters becomes stronger, while the number of hydration waters decreases. The evaluation results explain an application that the contact lens package solution to employ PEG with low molecular weights.**

I. INTRODUCTION

Water is a major prerequisite for life, which influences the structure and function of biomolecules. In the biomolecular aqueous environment, due to the polarizability of both the biomolecule and the water molecule, water molecules will be restricted to the surface of biomolecules by forming hydrogen bonds (HB), forming a bioprotective shell surrounding a biomolecule[1]. This effect has been wildly applied in biomedical preservation, such as preventing objects from dehydration, protecting objects from certain reactions with other molecules and so on.

Since the dynamical water HB structure fluctuates from sub-picoseconds to tens of picoseconds, the dielectric spectroscopy in the microwave-terahertz (THz)-far infrared (FIR) range is an optimum experimental technique to observe the dynamics of waters directly. Compared with the other typical tools, such as nuclear magnetic resonance (NMR) and the depolarized light scattering (DLS), the dielectric spectroscopy has two advantages: no requirement of the isotopic substitution or chromophore, and high sensitivity to tiny changes of HB network. Shiraga et al.[2] achieved a detailed characterization of the hydrogen-bond network of waters around disaccharides with a broadband dielectric spectrum.

PEG is one of the materials employed in biomedical preservation techniques. The contact lens package solution employs PEG to prevent the lens from dehydration and maintain the high-water content for its ready-to-wear condition. PEG with the molecular weights (Mw) from 100-500 Dalton (Da) is preferred in the application. In this study, a broadband dielectric spectrum was collected to evaluate PEG water solutions with various Mw. The obtained variation trend of the hydration numbers and the local force of the hydration shell can explain the application preference.

II. RESULTS

The hydration number was calculated based on an obtained broadband dielectric spectroscopy. *Figure 1* shows the fraction of the hydration water in the solutions with different PEG Mw. The result demonstrates that the ratio of hydration water decreases along with the increasing Mw, while the relaxation time of hydration water increases due to the stronger restriction force. As shown in *Table 1*, defining the retardation factor $\xi = \tau_H/\tau_{slow}$ (τ_H and τ_{slow} are the relaxation time of hydration waters and bulk waters, respectively), it is relatively weak at the lowest Mw PEG ($\xi = 2.09$), and increases up to ξ=2.25. Hydration number per monomer and per unit area were also evaluated. It further proves that the larger PEG molecule with a longer poly coil and a more complex structure has a poor ability to let each monomer interact with the maximum number of water molecules. Therefore, the contact lens package solution prefers PEG with low Mw aiming at a large number of locked waters. Considering the opposite trending restriction strength, patents only offer a preferred PEG Mw (100 – 500 Da) range, instead of a specific best choice.

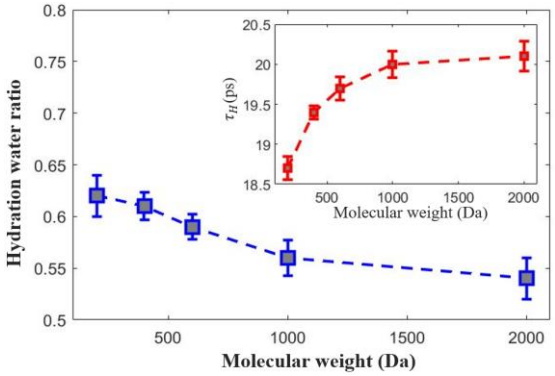

Fig. 1. Hydration water ratio of PEG solutions with different molecular weights. The inset exhibits the relaxation time of the hydration water at different molecular weights.

Tab. 1. Hydration state of PEG solutions with different molecular weights.

		200 Da	400 Da	600 Da	1000 Da	2000 D
Hydration number	1. Fraction	0.62	0.61	0.59	0.56	0.54
	2. Per monomer	6.06	5.93	5.76	5.47	5.28
	3. Per unit area ($Å^{-2}$)	0.68	0.58	0.54	0.48	0.43
4. Retardation factor $\xi = \tau_{H2}/\tau_{slow}$		2.09	2.17	2.20	2.23	2.25

III. SUMMARY

In this study, using broadband dielectric spectroscopy, we evaluated and compared the hydration state of PEG solutions with different Mw. The PEG with lower Mw possesses the higher efficiency to be bound more water molecules, but the formed hydration bond is not as sound as those with higher Mw. Considering the two opposite trending effects, manufacturers need to decide their best choice from this range, according to their needs of the number of locked waters and the strength locking waters.

REFERENCES

[1]. Laage, Damien, Thomas Elsaesser, and James T. Hynes. "Water dynamics in the hydration shells of biomolecules." Chemical Reviews 117.16 (2017): 10694-10725.
[2]. Shiraga, Keiichiro, et al. "Characterization of the hydrogen-bond network of water around sucrose and trehalose: Microwave and terahertz spectroscopic study." The Journal of chemical physics 146.10 (2017): 105102.