Introduction:

What is a glass:
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• Produced by cooling or by a liquid without crystallization
• Extremely high viscosities, \( T_g \), temperature, \( T_g \)

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• Extremely high viscosities, \( T_g \), temperature, \( T_g \), yielding amorphous (non-crystalline) materials
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Experimental investigations:

Glass temperatures, \( T_g \), homogeneous ice freezing, \( T_{hom} \), and ice melting temperatures, \( T_m \), of crystallized and bulk samples made of various organic, organic and inorganic/organic aqueous solutions have been investigated with a differential scanning calorimeter (DSC), see Fig. 1.

Inorganic/organic mixtures: mixture of HS and AS (MSA, 1.1 by mass) mixtures of HS and raffinose, of HS and silicic acid, and of HS and glycerol and AS (mixed model compounds of the water soluble organic fraction, Decesari et al. 2006; molar mass between 60-700 gmol-1) are observed for organic in model aerosols, e.g., Maltini et al. 2002.

Glasses are amorphous (non-crystalline) materials that behave mechanically as solids. They are produced by cooling or by a liquid without crystallization. Glasses have extremely high viscosities, \( T_g \), which yields amorphous (non-crystalline) materials.

\[ \Delta x (\text{water}) = \frac{10 E (1 - 10^{-4})}{10 E (1 - 10^{-4})} = 2.5 \times 10^{-3} \text{a} \text{t} \text{d} \text{ay} \]

Do atmospheric aerosols form glasses? Do atmospheric aerosols form glasses?

Fig. 1. Enthalpy and entropy change during the phase transitions of a crystallization/melting (first order phase change) and a glass transition.

If aqueous aerosols form glasses, then inhibition of:
• Water uptake
• Chemical reactions
• Crystal growth (e.g., ice)
• Ice nucleation

Fig. 2. DSC thermograms, panels (a)-(c), and state diagrams, panels (d)-(f), of 3 different concentrated aqueous glucose solutions: 0.25%, 0.50%, and 0.55% weight fraction. Panels (a)-(c): heating/cooling cycles from 300 K to 533 K with 10 K/min. Panel (d)-(f): The colored curves depict the course of the DSC experiments linked to the panel on the left side. Circles: Tg; Squares: Tm; Triangles: Tg1, Tg2; Hash: Tg, g2.

Glass temperatures of organic and multi-component solutions:

Fig. 4. State diagrams for several organic-water systems as a function of water activity, \( w_T \). Ice melting temperatures, \( T_m \); squares: homogeneous ice freezing temperatures, \( T_{hom} \); filled square triangles and filled right pointed triangles: \( T_m \). Solid lines: Glass curves; Dashed and solid black lines: Ice melting and homogeneous ice freezing curves, according to Knop et al. 2000, respectively. The large black circles denote the \( T_g \) for each solute, i.e., interaction between \( T_g \) and \( T_m \) curves (see also Fig. 6).

Fig. 5. State diagrams of multi-component solutions as a function of the solute weight fraction. Circles: \( T_g \); squares: \( T_m \); filled triangles: \( T_g \); open triangles: \( T_m \). Solid colored curves: Glass curves. Black circles: see Fig. 4.

Conclusions and Outlook:

1. Glass formation was observed in most of the investigated aqueous solutions (except AN and AS, cooling rate to low)!

2. It is unlikely that organic aerosols form glasses under atmospheric conditions.

3. Aqueous organic and multi-component solutions undergo glass transitions important at atmospheric temperatures and relative humidities.

4. \( T_g \) of such solutions depend predominantly on the molar mass of the solutes with a larger molar mass leading to a higher \( T_g \). To a lesser extent \( T_g \) also depends on the hydrophilicity of the molecules, with more hydrophilic molecules showing higher \( T_g \).

5. The \( T_g \) curves of the multi-component solutions are located between the \( T_g \) curves of the binary solutions close to that binary system with the lower \( T_g \).

6. Chemical reactions are impeded in v miscous aerosol particles as might be even inhibited completely in glassy aerosol particles. This may increase the lifetime of the aerosol with respect to gas reactions.

7. Water uptake is diminished or even fully inhibited for highly viscous or glassy aerosols, respectively.

8. Ice nucleation is inhibited at the homogeneous ice nucleation threshold when the aerosol is in a glassy state which leads to higher ice supersaturations than expected for liquid aerosols, mostly for \( T > 202 \text{K} \). Verification organic-enriched aerosols might lead to cirrus clouds with smaller ice particle number densities when compared to inorganic aerosols, which will influence the radiative effect of cirrus.

9. Knowledge of the chemical composition of the aerosol must be improved.

10. Water uptake and chemical reactions will be investigated by modeling and experimental studies.

References: