UPTAKE OF SO_2 BY SULFURIC ACID (pH = 2 to 70% wt H_2SO_4): OXIDATION BY H_2O_2 AND O_3

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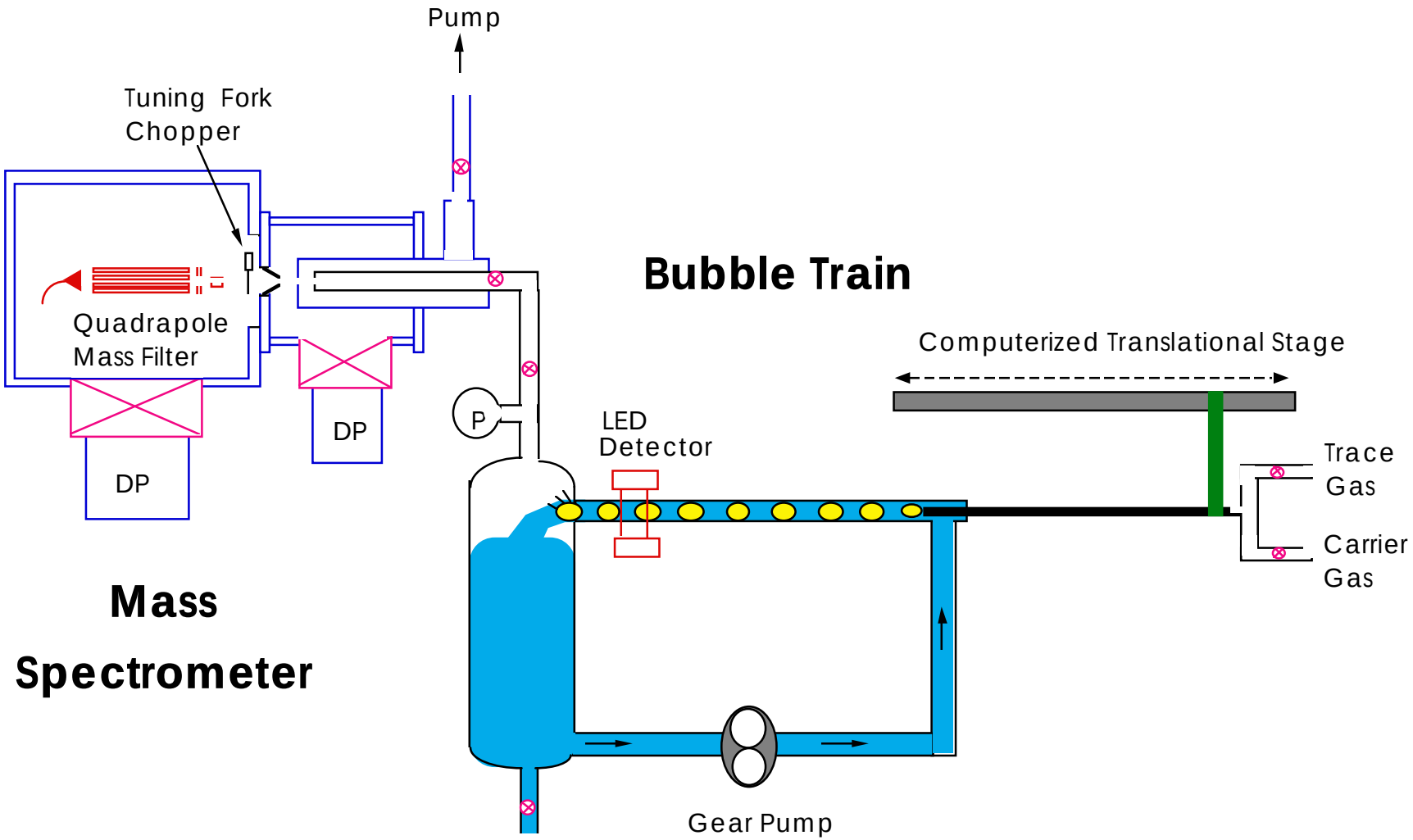
Recent observations of particle densities in aircraft wakes suggest a rapid oxidation of SO_2 to sulfate. Current models under-predict this fast oxidation by perhaps an order of magnitude, suggesting that some new unexplained gas phase or heterogeneous processes are occurring in the plume.

We have used the horizontal bubble train apparatus to investigate the uptake of gaseous SO_2 by sulfuric acid solution over a wide range of acidities (pH = 2 to 70% wt) at 293 K and in co-deposition with H_2O_2 and O_3.

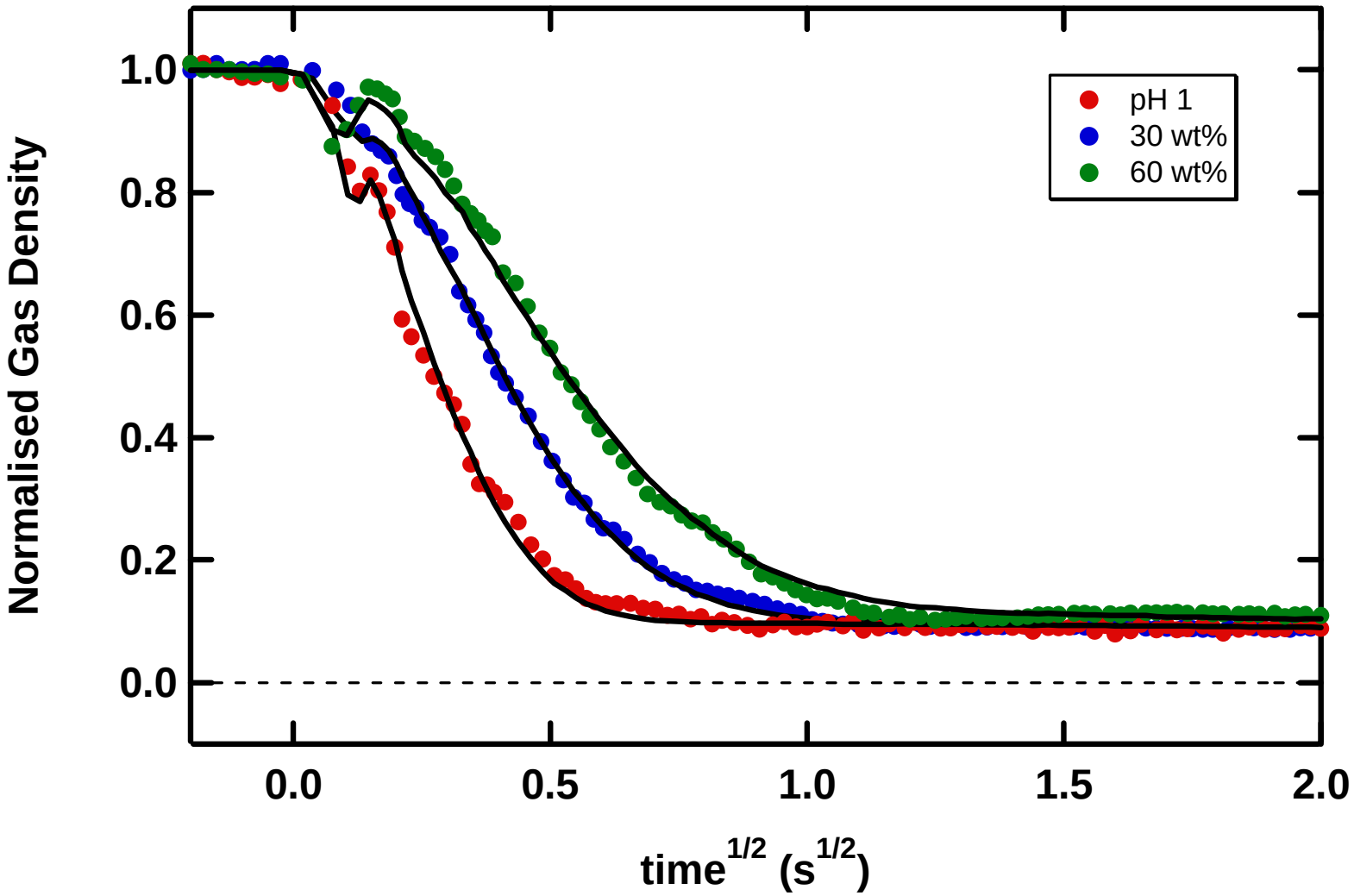
In the bubble train apparatus a low pressure flow of gas containing SO_2 is injected into the liquid forming well defined bubbles which move horizontally with the flow within a 0.4 cm i.d. tube. A computer controlled stepping motor determines the bubble travel distance in the tube and hence the gas-liquid interaction time. The uptake is determined by the change in the SO_2 gas density at the outlet end of the flow tube using a differentially pumped mass spectrometer. **Fig. 1**

Sample uptake data as a function of acidity are shown in **Fig. 2.** Using known Henry's law for SO_2 allowed the characterization of the system parameters for acid solutions **Fig 3.**

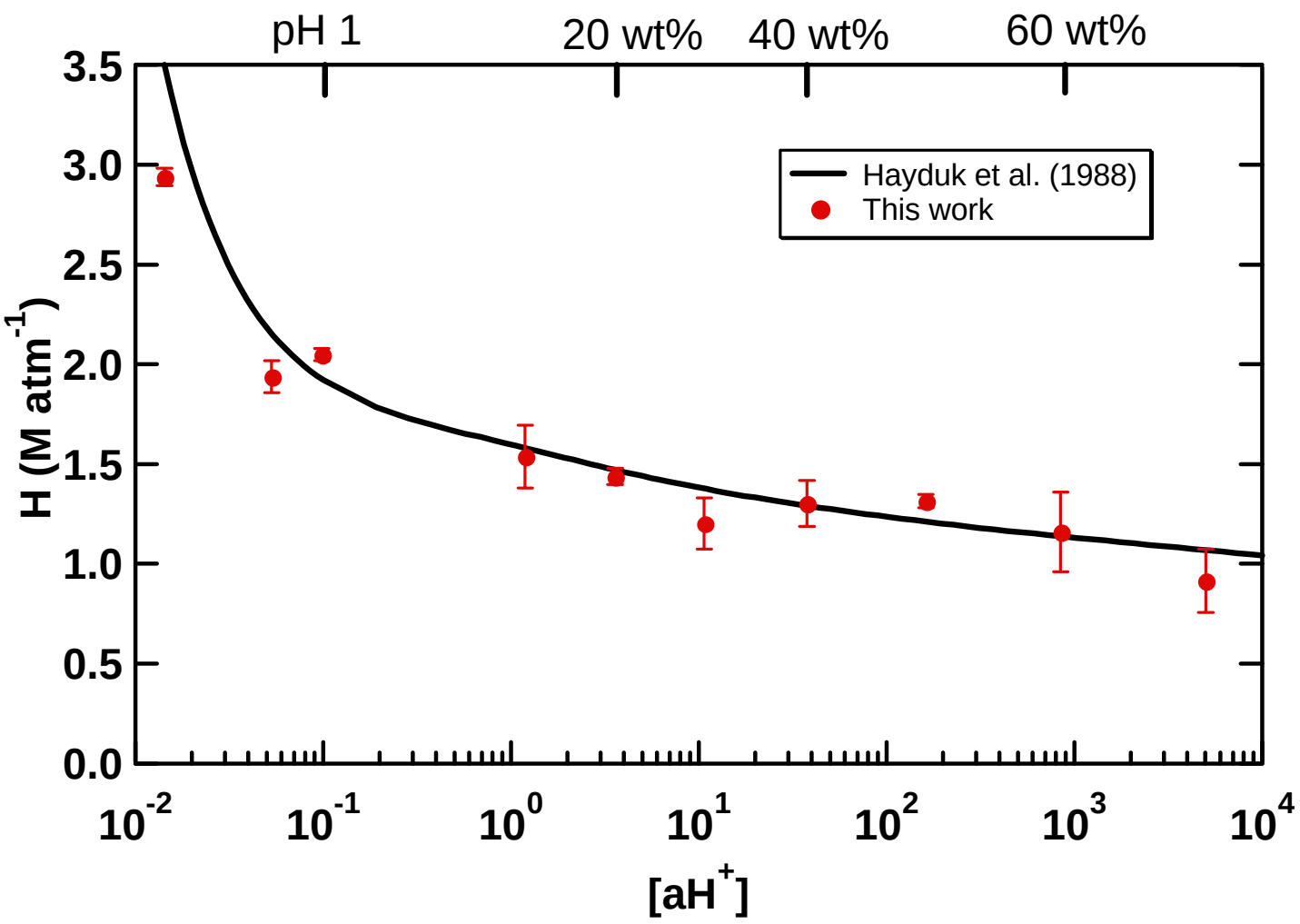
Horizontal Bubble Train Apparatus



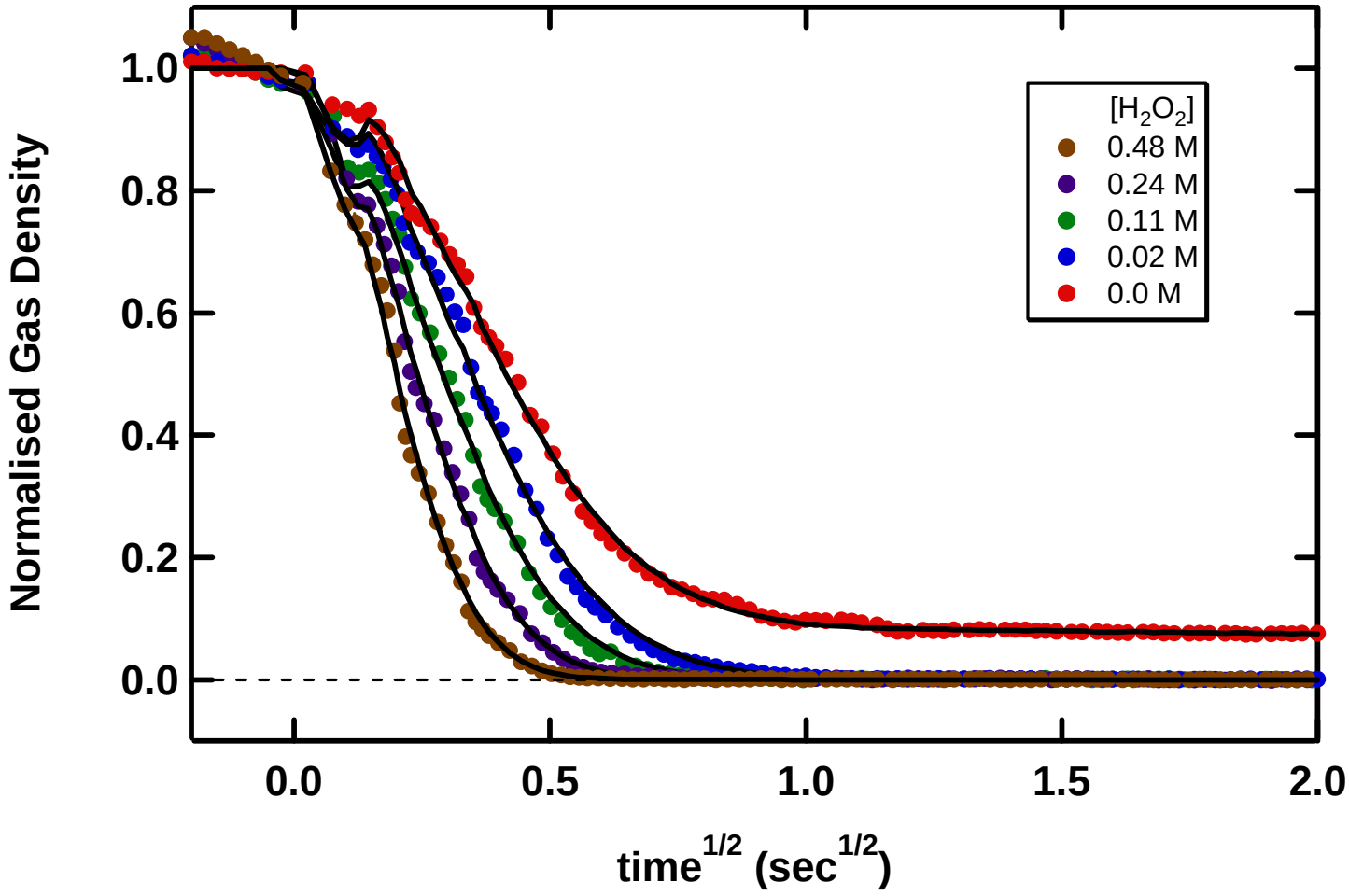
Uptake of SO_2 in acidic solutions



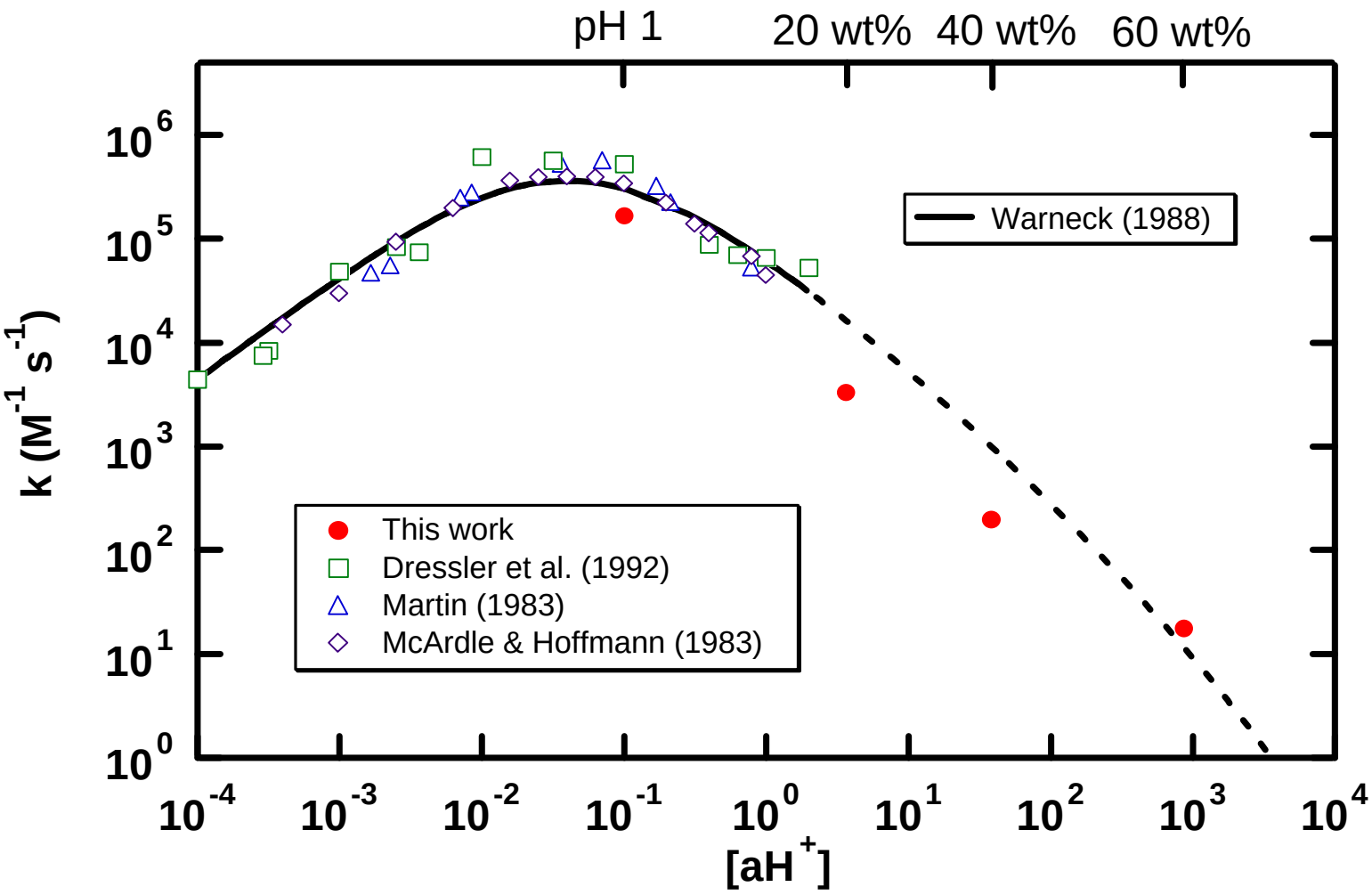
Effective Henry's law Coefficient as a Function of Acidity



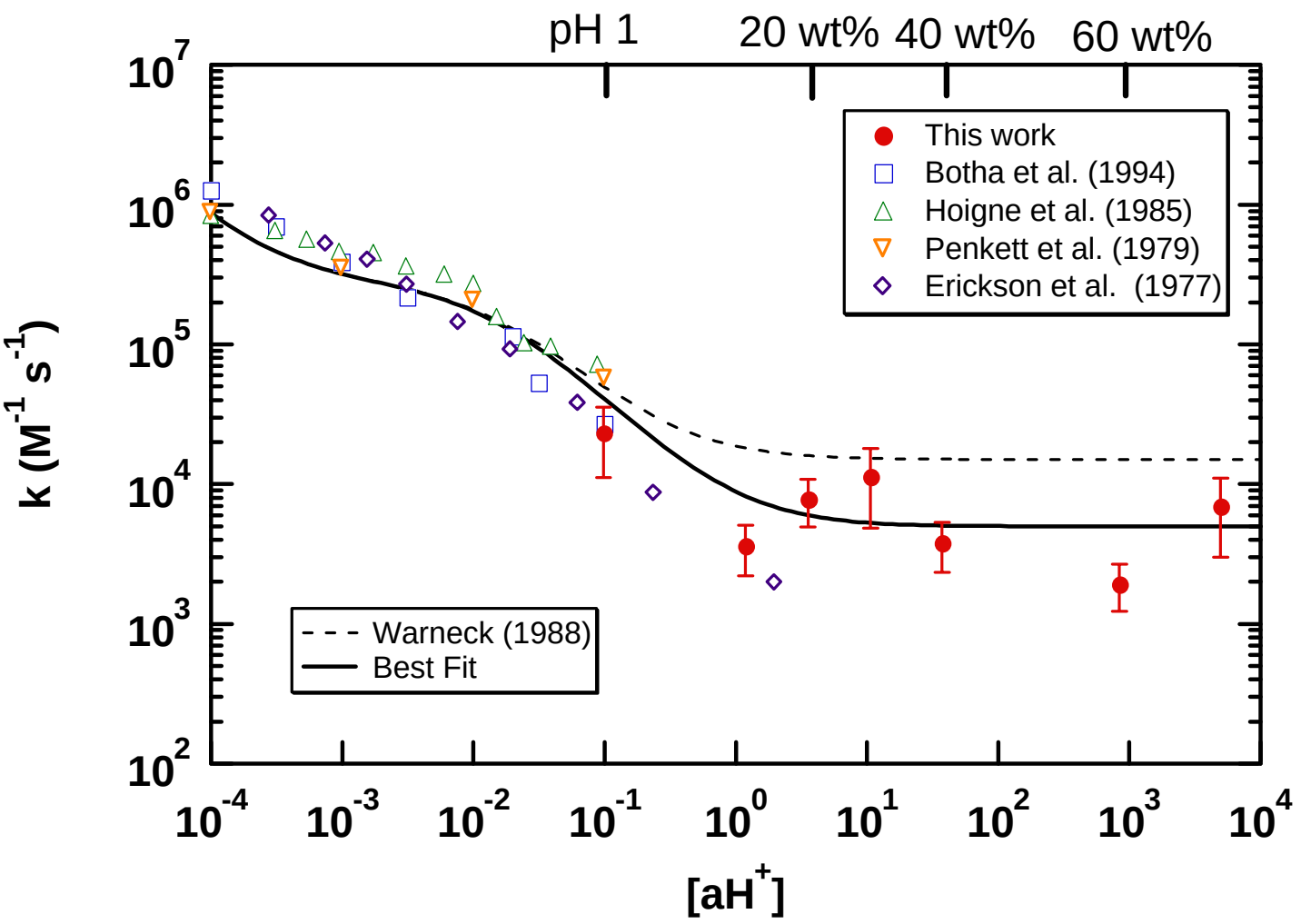
Uptake of SO_2 in 40 wt% H_2SO_4 in the presence of H_2O_2



Oxidation of SO_2 by H_2O_2 as a Function of Acidity



Oxidation of SO_2 by O_3 as a Function of Acidity



Uptake of SO_2 at high acidity is governed by Henry's law solubility. In the presence of added oxidants such as O_3 or H_2O_2, in the liquid or at the gas-liquid interface SO_2 can be further oxidized to S(VI) leading to enhanced uptake. The analysis of the SO_2 uptake observed in the bubble apparatus would reveal if there were significant SO_2/oxidant reactions occurring at the interface.

Fig. 4 shows uptake as a function of H_2O_2.

Figures 5 and 6 show SO_2/H_2O_2, and SO_2/O_3 reaction rate constants as a function of acidity obtained in our studies and compared to literature values obtained in bulk-phase experiments.

Conclusion: The two most likely paths of SO_2 oxidation that is with H_2O_2 and O_3 do not exhibit significant reaction enhancement at the gas-liquid interface