Determination of Mean Activity Coefficients of Metal Sulfates in Aqueous Sulfuric Acid-Sulfate Systems

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Mean activity coefficients of Na_2SO_4 , Li_2SO_4 and MgSO_4 were determined at 298 K by means of a graphic integration method applying the Gibbs-Dühem equation to aqueous H_2SO_4 - Na_2SO_4 , H_2SO_4 - Li_2SO_4 and H_2SO_4 - MgSO_4 systems. The mean activity coefficients of these metal sulfates decreased with an increase in H_2SO_4 concentration. This activity coefficient decrease was most distinct at low concentrations of these metal sulfates. The effect of H_2SO_4 on the mean activity coefficients of these metal sulfates was compared. It was found that the magnitude of $\gamma_{\pm}$ of these sulfates studied was in the order of $\text{Li}_2\text{SO}_4 > \text{Na}_2\text{SO}_4 > \text{MgSO}_4$ at the same molality.

1 Introduction

A knowledge of the activities of electrolytes and their constituent ions is important in understanding the reaction kinetics in many hydro-metallurgical processes such as the leaching of ores and the purification of leach solutions, as described in a previous paper¹⁾.

Measurements of the activities of acid and the constituent ions in aqueous HCl - MCl_n and H_2SO_4 - M_nSO_4 systems (M : alkali or alkaline earth metals) have been made in authors' laboratory²⁻⁶⁾. If the activities of H_2O and H_2SO_4 in aqueous H_2SO_4 - M_nSO_4 system are known, the activity of M_nSO_4 can be determined by applying Gibbs-Dühem equation. In a previous study, the authors have measured the activities of H_2SO_4 in aqueous H_2SO_4 - M_nSO_4 system by measuring the electromotive force of a cell consisting of a Pt-Pt black electrode and a PbO_2 - PbSO_4 electrode. The activities of H_2O in the same solution system were also determined by means of an isopiestic method⁶⁾.

This paper describes the activity coefficients of M_nSO_4 (including Na_2SO_4 , Li_2SO_4 and MgSO_4) as calculated by using Gibbs-Dühem equation.

2 Theoretical Background for the Determination of Activity Coefficients of Sulfates in Aqueous H_2SO_4 - M_nSO_4 system

The theoretical background and experimental procedures for the determination of the activity of H_2SO_4 in aqueous H_2SO_4 - M_nSO_4 system, applying electromotive force measurement techniques, were described in a previous paper¹⁾. The activity of H_2O in the same solution system, as determined by an isopiestic method, and the theoretical background as well as experimental procedures were reported in a separate paper⁶⁾.

The activity coefficients of M_nSO_4 in aqueous H_2SO_4 - M_nSO_4 system can be calculated by applying the Gibbs-Dühem relation, using activity data determined separately.

Denoting H_2O , H_2SO_4 and M_nSO_4 by suffixes w , 1 and 2, respectively, the Gibbs-Dühem equation for this particular solution system can be expressed as:

$$55.51 \, d \ln a_w + m_1 \, d \ln a_1 + m_2 \, d \ln a_2 = 0 \quad (1)$$

The activity of H_2O in aqueous H_2SO_4 - Na_2SO_4 (or Li_2SO_4) system can be expressed as a function of the osmotic coefficient of water, ϕ by equation (2).

$$55.51 \ln a_w = -3(m_1 + m_2)\phi \quad (2)$$

a_1 and a_2 of the same solution system are given by equations (3) and (4), respectively.

$$a_1 = 4 \gamma_{\pm,1}^3 m_1^2 (m_1 + m_2) \quad (3)$$

$$a_2 = 4 \gamma_{\pm,2}^3 m_2^2 (m_1 + m_2) \quad (4)$$

Inserting equations (2)-(4) into equation (1), we obtain equation (5).

$$(m_1 + m_2) d\phi + (\phi - 1) d m_1 + (\phi - 1) d m_2 - m_1 d \ln \gamma_{\pm,1} = m_2 d \ln \gamma_{\pm,2} \quad (5)$$

when $m_2 = m_2^\circ$ (constant), then $d m_2 = 0$. Here m_2° denotes the molality of Na_2SO_4 (or Li_2SO_4) in a binary Na_2SO_4 (or Li_2SO_4)- H_2O system.

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The integration of equation (5) at a constant m_2° value yields equation (6).

$$2.303 m_2^\circ \log(\tau_{\pm,2}/\tau_{\pm,2}^\circ) = \int_0^{m_1} (m_1 + m_2^\circ) d\phi + \int_0^{m_1} (\phi - 1) dm_1 - 2.303 \int_0^{m_1} m_1 d \log \tau_{\pm,1} \quad (6)$$

where $\tau_{\pm,2}^\circ$ is a mean activity coefficient of Na_2SO_4 (or Li_2SO_4) at its concentration of m_2° in a binary Na_2SO_4 (or Li_2SO_4)- H_2O system.

For aqueous H_2SO_4 - MgSO_4 system, equations (2)-(6) can be replaced by equations (2')-(6'), respectively.

$$55.51 \ln a_w = -(3m_1 + 2m_2)\phi \quad (2')$$

$$a_1 = 4\tau_{\pm,1}^3 m_1^2 (m_1 + m_2) \quad (3')$$

$$a_2 = \tau_{\pm,2}^2 m_2 (m_1 + m_2) \quad (4')$$

$$(3m_1 + 2m_2)d\phi + 3(\phi - 1)dm_1 + 2(\phi - 1)dm_2 - 3m_1 d \ln \tau_{\pm,1} = 2m_2 d \ln \tau_{\pm,2} \quad (5')$$

$$4.606 m_2^\circ \log(\tau_{\pm,2}/\tau_{\pm,2}^\circ) = \int_0^{m_1} (3m_1 + 2m_2) d\phi + 3 \int_0^{m_1} (\phi - 1) dm_1 - 6.909 \int_0^{m_1} m_1 d \log \tau_{\pm,1} \quad (6')$$

The graphic integration of each term on the right hand side of equations (6) and (6') from $m_1=0$ to $m_1=m_1$ under the condition of $m_2=m_2^\circ$ enables us to determine the activity coefficient of metal sulfate, M_nSO_4 , for aqueous H_2SO_4 - M_nSO_4 system.

3 Data Used for the Determination of the Mean Activity Coefficients of Metal Sulfates

As mentioned before, the authors and their coworkers determined the activities of H_2SO_4

and H_2O in aqueous H_2SO_4 - Na_2SO_4 , H_2SO_4 - Li_2SO_4 and H_2SO_4 - MgSO_4 systems in separate studies⁴⁻⁶. However, these data cannot be used directly in the present calculation since the osmotic coefficients, ϕ and the mean activity coefficients of H_2SO_4 , $\tau_{\pm,1}$, at a constant metal sulfate concentration $m_2=m_2^\circ$ are required for this calculation. In this study, the necessary values of ϕ and $\tau_{\pm,1}$ were determined by means of both interpolation and extrapolation based on the activity values determined in our laboratory and on the thermodynamic data previously published⁷.

Tables I to III show the values of ϕ and $\tau_{\pm,1}$ thus determined at $m_2=m_2^\circ$. These values are used for the calculation of $\tau_{\pm,2}$ in aqueous H_2SO_4 - M_nSO_4 system.

4 Results and Discussion

A typical example of graphic integration is shown in Fig. 1. In this particular example, thermodynamic data for aqueous H_2SO_4 - Li_2SO_4 system at 298 K were used. Figures 1(a), 1(b) and 1(c) correspond to the first term, second term and third term, respectively, on the right hand side of equation (6). By integrating graphically from $m_1=0$ to $m_1=m_1$, we can determine $\tau_{\pm,2}$ at a constant level of $m_2=m_2^\circ$ from equation (6).

Similarly the values of $\tau_{\pm}$ of Na_2SO_4 and MgSO_4 in aqueous H_2SO_4 - Na_2SO_4 and H_2SO_4 - MgSO_4 systems, respectively, can be determined by graphic integration.

The values of $\tau_{\pm,2}$ for aqueous H_2SO_4 - Na_2SO_4 , H_2SO_4 - Li_2SO_4 and H_2SO_4 - MgSO_4 systems calculated in this manner at 298 K are listed in the last column of Tables I to III.

Fig. 1 A typical example of graphic integration at $m(\text{Li}_2\text{SO}_4)=0.5 \text{ mol} \cdot \text{kg}^{-1}$ (Example employed: aqueous H_2SO_4 - Li_2SO_4 system)

Table 1 Thermodynamic data used for the calculation of mean activity coefficients of Na_2SO_4 in aqueous H_2SO_4 - Na_2SO_4 system at 298 K

$m(\text{Na}_2\text{SO}_4)$ mol·kg ⁻¹	$m(\text{H}_2\text{SO}_4)$ mol·kg ⁻¹	$a(\text{H}_2\text{O})$	ϕ	$\log \gamma(\pm, \text{H}_2\text{SO}_4)$	$\log \gamma(\pm, \text{Na}_2\text{SO}_4)$
0.1	0		0.787*	-0.556**	-0.344***
	0.1	0.9929	0.659	-0.688	-0.462
	0.2	0.9893	0.664	-0.769	-0.491
	0.3	0.9860	0.652	-0.800	-0.581
	0.4	0.9822	0.665	-0.824	-0.625
	0.5	0.9788	0.661	-0.842	-0.699
	0.6	0.9750	0.669	-0.860	-0.719
	0.7	0.9711	0.678	-0.876	-0.726
	0.8	0.9672	0.686	-0.889	-0.738
	0.9	0.9630	0.698	-0.899	-0.733
	1.0	0.9588	0.708	-0.907	-0.750
	1.1	0.9543	0.721	-0.912	-0.750
	1.2	0.9497	0.735	-0.914	-0.777
	1.3	0.9451	0.746	-0.914	-0.818
0.2	0		0.748*	-0.717**	-0.431***
	0.1	0.9895	0.651	-0.792	-0.533
	0.2	0.9859	0.657	-0.832	-0.573
	0.3	0.9827	0.646	-0.851	-0.638
	0.4	0.9791	0.651	-0.864	-0.684
	0.5	0.9756	0.653	-0.876	-0.733
	0.6	0.9718	0.662	-0.888	-0.759
	0.7	0.9679	0.671	-0.899	-0.783
	0.8	0.9640	0.678	-0.908	-0.801
	0.9	0.9598	0.690	-0.915	-0.812
	1.0	0.9556	0.700	-0.920	-0.830
	1.1	0.9512	0.712	-0.925	-0.824
	1.2	0.9465	0.727	-0.928	-0.824
	1.3	0.9423	0.733	-0.929	-0.857
0.5	0		0.693*	-1.053**	-0.569***
	0.1	0.9792	0.648	-0.986	-0.625
	0.2	0.9760	0.642	-0.965	-0.666
	0.3	0.9731	0.631	-0.961	-0.706
	0.4	0.9697	0.633	-0.958	-0.740
	0.5	0.9661	0.638	-0.957	-0.767
	0.6	0.9623	0.646	-0.957	-0.790
	0.7	0.9583	0.657	-0.957	-0.810
	0.8	0.9545	0.663	-0.957	-0.833
	0.9	0.9501	0.677	-0.957	-0.845
	1.0	0.9458	0.687	-0.957	-0.860
	1.1	0.9414	0.698	-0.956	-0.873
1.0	0		0.644*	-1.218**	-0.690***
	0.1	0.9630	0.634	-1.147	-0.714
	0.2	0.9600	0.630	-1.098	-0.740
	0.3	0.9570	0.626	-1.079	-0.762
	0.4	0.9538	0.625	-1.069	-0.783
	0.5	0.9503	0.629	-1.060	-0.799
	0.6	0.9466	0.635	-1.048	-0.818
	0.7	0.9429	0.640	-1.033	-0.839
	0.8	0.9385	0.653	-1.020	-0.854

$m(\text{Li}_2\text{SO}_4)$ mol·kg ⁻¹	$m(\text{H}_2\text{SO}_4)$ mol·kg ⁻¹	$a(\text{H}_2\text{O})$	ϕ	$\log \gamma(\pm, \text{H}_2\text{SO}_4)$	$\log \gamma(\pm, \text{Li}_2\text{SO}_4)$
1.5	0		0.621*	-1.253**	-0.764***
	0.1	0.9470	0.630	-1.200	-0.772
	0.2	0.9443	0.624	-1.168	-0.790
	0.3	0.9413	0.622	-1.152	-0.804
	0.4	0.9381	0.622	-1.142	-0.818
	0.5	0.9346	0.626	-1.135	-0.830

* calculated from mean activity coefficients of Na_2SO_4 in Na_2SO_4 - H_2O binary system by using Gibbs-Duhem equation.

** extrapolated from mean activity coefficients of H_2SO_4 as determined in the present study.

*** after Dobos⁷⁾.

Figure 2 shows the effect of H_2SO_4 concentration on $\gamma_{\pm}$ of Na_2SO_4 at a constant Na_2SO_4 concentration. As may be seen in this figure, $\gamma_{\pm}$ of Na_2SO_4 at a low Na_2SO_4 level, such as 0.1 mol·kg⁻¹, decreases sharply with an increase in H_2SO_4 concentration. On the contrary, the effect of H_2SO_4 concentration on $\gamma_{\pm}$ of Na_2SO_4 in 1.0 and 1.5 mol·kg⁻¹ Na_2SO_4 solutions was less than that in dilute solutions. The values of $\gamma_{\pm}$ of Na_2SO_4 in concentrated Na_2SO_4 solutions also decrease with an increase in H_2SO_4 concentration.

Figure 3 depicts the effect of H_2SO_4 concen-

Fig. 2 Mean activity coefficients of Na_2SO_4 in aqueous H_2SO_4 - Na_2SO_4 system at 298 K

Table 2 Thermodynamic data used for the calculation of mean activity coefficients of Li_2SO_4 in aqueous H_2SO_4 - Li_2SO_4 system at 298 K

$\frac{m(\text{Li}_2\text{SO}_4)}{\text{mol} \cdot \text{kg}^{-1}}$	$\frac{m(\text{H}_2\text{SO}_4)}{\text{mol} \cdot \text{kg}^{-1}}$	$a(\text{H}_2\text{O})$	ϕ	$\log \gamma(\pm, \text{H}_2\text{SO}_4)$	$\log \gamma(\pm, \text{Li}_2\text{SO}_4)$
0.1	0		0.798*	-0.671**	-0.321***
	0.1	0.9923	0.715	-0.723	-0.460
	0.2	0.9889	0.689	-0.766	-0.553
	0.4	0.9814	0.695	-0.806	-0.708
	0.5	0.9779	0.689	-0.826	-0.735
	0.7	0.9703	0.697	-0.863	-0.759
	1.5	0.9338	0.792	-0.897	-0.900
0.2	0		0.778*	-0.736**	-0.391***
	0.1	0.9883	0.726	-0.794	-0.462
	0.2	0.9850	0.699	-0.817	-0.529
	0.3	0.9816	0.687	-0.828	-0.592
	0.4	0.9779	0.689	-0.840	-0.636
	0.5	0.9740	0.696	-0.852	-0.674
	0.7	0.9661	0.709	-0.880	-0.693
	1.2	0.9435	0.769	-0.906	-0.728
0.5	1.5	0.9287	0.805	-0.904	-0.757
	0		0.761*	-0.913**	-0.487***
	0.1	0.9764	0.737	-0.920	-0.520
	0.2	0.9733	0.715	-0.923	-0.554
	0.3	0.9691	0.726	-0.922	-0.580
	0.4	0.9654	0.724	-0.919	-0.602
	0.5	0.9616	0.725	-0.916	-0.623
	0.7	0.9530	0.742	-0.921	-0.648
	1.0	0.9390	0.776	-0.932	-0.652
	1.2	0.9293	0.798	-0.926	-0.670
1.0	1.5	0.9137	0.835	-0.913	-0.701
	0		0.783*	-1.023**	-0.548***
	0.1	0.9546	0.782	-1.013	-0.559
	0.2	0.9510	0.775	-1.006	-0.572
	0.3	0.9470	0.775	-0.999	-0.585
	0.4	0.9428	0.779	-0.992	-0.597
	0.5	0.9389	0.778	-0.985	-0.607
	0.7	0.9301	0.789	-0.974	-0.611
	1.0	0.9159	0.813	-0.962	-0.642
1.5	1.2	0.9059	0.831	-0.949	-0.654
	0		0.816*	-1.073**	-0.565***
	0.1	0.9323	0.811	-1.059	-0.573
	0.2	0.9286	0.806	-1.047	-0.582
	0.3	0.9246	0.806	-1.040	-0.590
	0.4	0.9206	0.806	-1.034	-0.597
	0.5	0.9163	0.809	-1.026	-0.604

* calculated from mean activity coefficients of Li_2SO_4 in Li_2SO_4 - H_2O binary system by using Gibbs-Dühem equation.

** extrapolated from mean activity coefficients of H_2SO_4 as determined in the present study.

*** after Dobos²⁾.

Table 3 Thermodynamic data used for the calculation of mean activity coefficients of MgSO_4 in aqueous H_2SO_4 - MgSO_4 system at 298 K

$\frac{m(\text{MgSO}_4)}{\text{mol} \cdot \text{kg}^{-1}}$	$\frac{m(\text{H}_2\text{SO}_4)}{\text{mol} \cdot \text{kg}^{-1}}$	$a(\text{H}_2\text{O})$	ϕ	$\log \gamma(\pm, \text{H}_2\text{SO}_4)$	$\log \gamma(\pm, \text{MgSO}_4)$
0.1	0		0.565*	-0.646**	-0.745***
	0.1	0.9943	0.631	-0.707	-0.922
	0.2	0.9907	0.645	-0.767	-0.991
	0.4	0.9836	0.655	-0.818	-1.201
	0.6	0.9763	0.666	-0.850	-1.362
	0.8	0.9686	0.681	-0.873	-1.479
	1.0	0.9605	0.699	-0.887	-1.595
	1.2	0.9518	0.722	-0.893	-1.728
	1.4	0.9425	0.747	-0.895	-1.799
0.2	1.5	0.9376	0.761	-0.897	-1.793
	0		0.501*	-0.764**	-0.959***
	0.1	0.9926	0.586	-0.790	-1.049
	0.2	0.9890	0.613	-0.812	-1.133
	0.4	0.9818	0.639	-0.845	-1.269
	0.6	0.9743	0.658	-0.870	-1.362
	0.8	0.9664	0.677	-0.885	-1.444
	1.0	0.9583	0.696	-0.893	-1.521
	1.2	0.9492	0.723	-0.896	-1.587
0.5	1.4	0.9395	0.753	-0.898	-1.606
	1.5	0.9342	0.771	-0.999	-1.585
	0		0.484*	-0.925**	-1.167***
	0.2	0.9838	0.567	-0.914	-1.246
	0.4	0.9760	0.613	-0.910	-1.321
	0.6	0.9677	0.650	-0.909	-1.379
	0.8	0.9589	0.686	-0.912	-1.412
	1.0	0.9490	0.726	-0.915	-1.408
	1.2	0.9399	0.748	-0.912	-1.445
1.0	1.4	0.9305	0.769	-0.906	-1.491
	1.5	0.9237	0.801	-0.903	-1.449
	0		0.500*	-1.017**	-1.310***
	0.2	0.9732	0.581	-1.000	-1.331
	0.4	0.9642	0.633	-0.965	-1.367
	0.6	0.9547	0.677	-0.944	-1.393
	0.8	0.9445	0.721	-0.935	-1.402
1.5	1.0	0.9334	0.765	-0.924	-1.406
	1.2	0.9216	0.809	-0.910	-1.405
	0		0.560*	-1.052**	-1.372***
	0.2	0.9592	0.642	-1.032	-1.368
	0.4	0.9485	0.699	-0.989	-1.381
1.5	0.6	0.9374	0.747	-0.959	-1.366
	0.8	0.9259	0.791	-0.938	-1.388
	1.0	0.9143	0.829	-0.919	-1.381

* calculated from mean activity coefficients of MgSO_4 in MgSO_4 - H_2O binary system by using Gibbs-Dühem equation.

** extrapolated from mean activity coefficients of H_2SO_4 as determined in the present study.

*** after Dobos²⁾.

Fig. 3 Mean activity coefficients of Li_2SO_4 in aqueous H_2SO_4 - Li_2SO_4 system at 298 K

Fig. 5 A comparison of the effects of H_2SO_4 concentration on the mean activity coefficients of metal sulfates whose concentration are $0.5 \text{ mol} \cdot \text{kg}^{-1}$

Fig. 4 Mean activity coefficients of MgSO_4 in aqueous H_2SO_4 - MgSO_4 system at 298 K

tration on $\gamma_{\pm}$ of Li_2SO_4 at a constant Li_2SO_4 level. Figure 4 shows a similar illustration for aqueous H_2SO_4 - MgSO_4 system. The effect of H_2SO_4 concentration on $\gamma_{\pm}$ of Li_2SO_4 , as well as on $\gamma_{\pm}$ of MgSO_4 , appears similar to that on $\gamma_{\pm}$ of Na_2SO_4 .

Figure 5 shows a comparison of the effect of H_2SO_4 concentration on $\gamma_{\pm}$ of metal sulfates for aqueous H_2SO_4 - M_nSO_4 system whose M_nSO_4 concentration is $0.5 \text{ mol} \cdot \text{kg}^{-1}$. The effect of H_2SO_4 on mean activity coefficients of these metal sulfates was compared. It was found that the magnitude of $\gamma_{\pm}$ of these sulfates studied was in the order of $\text{Li}_2\text{SO}_4 > \text{Na}_2\text{SO}_4 > \text{MgSO}_4$ at the same molality.

5 Conclusions

Mean activity coefficients of Na_2SO_4 , Li_2SO_4 , and MgSO_4 were determined by means of a graphic integration method applying Gibbs-Duhem equation to aqueous H_2SO_4 - M_nSO_4 system. For this purpose, osmotic coefficients and mean activity coefficients of H_2SO_4 , determined by interpolation and extrapolation me-

thods from the thermodynamic data of the previous works of the present authors, were used

The mean activity coefficients of these sulfates decrease with an increase in the concentration of H_2SO_4 . The decreasing tendency was most distinct at low concentrations of these metal sulfates. The effect of H_2SO_4 concentration on the mean activity coefficients of these sulfates was compared. It was found that the magnitude of $\gamma_{\pm}$ of these sulfates studied was in the order of $Li_2SO_4 > Na_2SO_4 > MaSO_4$ at the same molarity.

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協会レポート

第6回溶液化学シンポジウム

第6回溶液化学シンポジウムは大阪市立大学理学部村上幸夫教授の肝煎で、昭和58年12月12日、13日の両日にわたり、近鉄上本町駅近くのなにわ会館で開催された。春や秋に殺到する他の討論会との重複を避けるために、このシンポジウムは12月頃に開かれることが多い。

これまでの溶液化学シンポジウムの短い歴史をふりかえてみると表1のようになる。

こうしてみると、はじめの2~3年は20~25件程度だった発表論文数が、その後約30数件に増え、さらに今年になって飛躍的に増加したことが示される。参加者もこれまではおよそ70~80人ぐらいであったが、今回は170人にも及ぶものとなり、もはや従来の1会場形式では納まらず、遂にやむを得ず2会場に拡張せざるを得なくなった。

今年は101才の長い生涯の最後まで溶液に関する研究に捧げられたHildebrand教授の逝去を悼んで特別セッションが設けられ、この中では特別講演2件を含む6件の論文が発表された。また折から来日中の韓国高等科学研究所の全武植 (Mu Shik Jhon) 博士が特別講演を行うなど、国際色も盛り込んだ多彩な企画がたてられ、最終日の最後の講演まで多数の参加者の足をひき止め、活発な討論が行われた。

溶液化学シンポジウムの特徴はまず発表論文の内容が

表 1

	期 日	会 場	発表論文数	備 考
第1回	53年4月5~6日	東工大 (長津田)	22	電気化学協会第45回大会と併催
第2回	54年12月12~13日	東工大 (長津田)	25	昭和54年度文部省科学研究費総合研究Bシンポジウムと併催
第3回	55年10月1~2日	東工大 (大岡山)	21	電気化学協会秋季大会と併催
第4回	56年11月11~13日	静岡大 (浜松)	39	日本熱測定学会と共催
第5回	57年12月8~10日	京大会館 (京都)	32	昭和57年度文部省科学研究費総合研究Bシンポジウムと併催
第6回	58年12月12~13日	なにわ会館 (大阪)	74	

実に多種多様で、さまざまな理論や測定法が適用されている点であろう。「溶液」というきわめて広範囲で漠然としている系を多方面からアプローチしようとする試みがなされ、ほとんどすべての研究発表に対して熱心な質疑応答が行われる。こういった熱気が長く継続するシンポジウムであって欲しいものである。

来年は東京地区で第7回の溶液化学シンポジウムが開催される予定である。

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