# THE EFFECTS OF SALT TYPE AND SALINITY ON FORMATION WATER VISCOSITY AND NMR RESPONSES

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## ABSTRACT

Reservoir water salinity and viscosity are important parameters for formation evaluation and production. For log analysts, salinity is critical to resistivity-based saturation estimation. It is well understood that water salinity affects its viscosity and diffusivity, which, in turn, affects Nuclear Magnetic Resonance (NMR) relaxation time estimation and NMR-based log interpretation. For production engineers, the ability to measure the formation water viscosity is important for enhanced oil recovery because water viscosity affects the  $CO_2$  injection via the effective permeability. The formation water viscosity in sedimentary rocks can vary by one order of magnitude for different types and concentrations of salts.

Previous studies on formation water properties have focused on NaCl and KCl, the two most common brines in connate water and in water-based drilling mud. The common practice in formation evaluation is to treat the physical properties of non-NaCl brines by a phenomelogical NaCl-equivalent quantity. While this approach may be reasonable for the estimation of brine resistivity, the viscosities of NaCl and KCl follow a different salinity trend. In fact, viscosity versus salinity behaves differently for different saline types. Three factors contribute to the viscosity of the ionic solution: Brownian motion, Debye-Hückel interaction (electrostatic potential due to all other ions surrounded), and structural temperature effect (the structural tightening or loosening due to hydrated or unhydrated ions, respectively). The Brownian motion and the Debye-Hückel interaction always contribute positively to the viscosity of any brine. The structural temperature effect, however, alters the viscosity of brine either positively or negatively depending on the type of salts. Until now, this effect has not been recognized for brines of interest within the formation evaluation community.

We found the solute composition of formation water plays an important role in determining its viscosity and diffusivity with varying salt concentration. By considering the structural temperature effect of brine components and using our experimental data on various brines, we are able to predict the viscosity and diffusivity of a large suite of brine types over a selected temperature range that more accurately compares with the previous correlations and does not consider the structural temperature effect. Furthermore, our

experimental studies establish better NMR correlations with brine types and salinity, which are beneficial to improving NMR log interpretation.

## **INTRODUCTION**

Nuclear Magnetic Resonance (NMR) well logging techniques have gained recognition for being one of the most valuable formation characterization tools. Among various NMR logging applications, estimating the viscosity of formation fluids from the transverse relaxation time ( $T_2$ ) or diffusion measurements is a unique NMR capability not accomplishable by conventional logging techniques.

Although reservoir water properties are not directly affected by hydrocarbons as they are immiscible, water properties are important for formation evaluation such as resistivity based saturation analysis and they affect fluid transport and mobility in formation rocks. It has been a common practice to estimate the viscosity of any formation water from that of an NaCl solution [1]. In other words, when the viscosity of formation water is considered, all ionic solutes are treated as if they were NaCl. This oversimplification could result in inaccurate NMR data interpretation or production rate estimation due to a wrong viscosity estimation.

Various factors affect the viscosity of brine, such as temperature, salinity, and composition. It is obvious all brines become thinner as the temperature increases. The brines with higher salinity, however, do not always result in higher viscosity. The different viscosity responses to the salinity changes are mainly due to the sign differences of the structural temperature coefficients of each salt [2]. That is to say, the composition of brine affects the relationship between its viscosity and salinity.

In this paper, the theoretical overview and experimental data for the viscosity of formation water with a different temperature, salinity, and composition are presented and applied to improving the quality of the NMR relaxation time,  $T_2$ , and the diffusivity estimation of formation water.

### Viscosity Mechanism for Water

The physical origin of viscosity can be treated by considering three mechanisms [2]. Firstly, each ion is treated as an independent particle carrying momentum from one part of the liquid to another by its Brownian motion. Secondly, the effect of the ionic interaction is accounted for by the Debye-Hückel theory [3] that a certain resistance to shear relates directly to viscosity. Thus, the contributions to the viscosity of any ionic solution by the Brownian motion and Debye-Hückel interaction are always positive. Finally, the addition of ions affects the water structure, either increasing or decreasing the strength of the bonds. This last factor contributes to the viscosity of the ionic solution either positively or negatively depending on the type of salts. Bernal and Fowler expressed the contributions of the above three factors to the viscosity of the ionic solution of the salt, AX, as follows:

$$\eta(AX, C, T) = \eta(aq, T) - f_{(a)}(M_A, M_X)CT - f_{(b)}(C, z)e^{-\alpha/T} - f_{(c)}(r_A, r_X, z)C,$$
(1)

where  $\eta(AX, C, T)$  is the viscosity of AX with concentration C and temperature T;  $\eta(aq, T)$ , the viscosity of pure water at temperature T;  $f_{(a)}(M_A, M_X)$ , the Brownian coefficient that depends on  $M_A$  (the mass of  $A^+$  ion) and  $M_X$  (the mass of  $X^-$  ion);  $f_{(b)}(C, z)$ , the Debye-Hückel coefficient with concentration C and charge z; and  $f_{(c)}(r_A, r_X, z)$ , the structural temperature function with the ionic radii  $r_A$ ,  $r_X$  and the charge z [2].

The structural temperature effect has been known since 1933, when the work of Bernal and Fowler was published, whereby the addition of ions changes the structure of water [2]. This phenomenon can be observed in the viscosities of the dilute solutions of alkali halide salts. Solutions of KF in water are more viscous than pure water, while solutions of KI in water are less viscous than pure water. The term 'structural temperature' was also suggested in ref. [2]. The F<sup>-</sup> ion strengthens the structure of water, an analogy to the effect of lowering the temperature, whereas the I<sup>-</sup> ion weakens the structure making' ions, or *kosmotropes*, whereas ions such as I<sup>-</sup> are called 'structure breaking' ions, or *chaotropes* [2, 4].

The structural temperature classification of ions is also possible with the Jones-Dole viscosity B coefficient [5]. Jones and Dole, while studying dilute aqueous solutions, found their results could be written in the form,

(2) 
$$\eta/\eta_{w} = 1 + Ac^{1/2} + Bc,$$

where  $\eta_w$  is the viscosity of the pure water,  $\eta$  is that of aqueous salt solution, *c* is the concentration of the solute, and *A* and *B* are constants characteristic of the solute. *A* is an electrostatic term that can be neglected at moderate concentrations, and *B* is a measure of ion-water interactions. Thus, the ions can be divided into *kosmotropes* or *chaotropes* by the sign of the Jones-Dole viscosity *B* coefficient, which is positive and negative, respectively [2, 4]. Table 1 lists the viscosity coefficient *B* for a number of ions and is adopted from the earlier reference [5]. The viscosity of brine with mixtures of various ions can be derived from Eq. (2), since the viscosity of the ionic solution is proportional to the electrostatic forces between the positively and the negatively charged ion, and to the number of ions of that species present in unit volume [6].

The experimental measurements of viscosity,  $T_2$ , and diffusivity of the selected brines from both group (*kosmotropes* and *chaotropes*) at various temperatures and salinities confirm the importance of structural temperature effect for accurate formation water evaluation.

## PROCEDURES

All experiments were performed on a 2 MHz (<sup>1</sup>H) Maran Ultra bench top NMR spectrometer. The magnitude of the gradient used for the diffusivity measurement was 15.3 Gauss/cm. The high-temperature, high-pressure (HTHP) NMR experiment was performed by circulating a pressurized fluid heated to the desired temperature through the sample probe. The viscosity measurements were conducted with a Cannon-Fenske routine viscometer and a temperature-controlled oil bath with a  $\pm 0.1^{\circ}$ C precision. The timer was a CMS 219-519 stopwatch reading to the nearest 0.2 sec.

The ionic solution of LiCl, NaCl, KCl, and CsCl were chosen to study the salinity effects of various brines. The selected alkali chloride solutes (purity > 99%) are shelf-products available from Sigma-Aldrich.

### **RESULTS AND DISCUSSION**

#### Method for measuring viscosity

The two most frequently used methods for measuring viscosity are kinematic viscosity  $\eta_k$ , or dynamic (absolute) viscosity,  $\eta_d$ . The majority of oil analysis labs today utilize the kinematic methodology. Kinematic viscosity measures fluid flowing characteristics resulting from the effect of gravity on its mass and is generally described in centistokes (cSt). Dynamic viscosity measurements, however, determine the force required to shear a fluid and are in centipoises (cP). The two measures are related by

$$\eta_k = \eta_d / \rho,$$

(3)

where  $\rho$  is the fluid density. Usually, dynamic viscosity measurements assess a fluid's resistance to shear under a high shear rate, or high force, conditions. It is an adequate method of choice to measure the viscosity of non-Newtonian fluids in which the viscosity changes with the applied shear force. Measurement of kinematic viscosity addresses fluids resistance to flow under a low shear rate, or low force, conditions. Kinematic viscosity is the preferred unit when the shear stress and shear rate of the fluid is influenced by the density.

In Figure 1, the measured viscosity of LiCl, NaCl, KCl, and CsCl solutions at an ambient condition (30°C, 1atm) are plotted with two different types of viscosity, kinematic viscosity and dynamic viscosity, as a function of concentration. It is obvious that measuring the viscosity of brines with the kinematic viscosity method (Figure 1(a)) represents the flow of brines under gravity better than the viscosity measured by the dynamic viscosity method (Figure 1(b)). Thus, for brines, which are Newtonian fluids, we adapt the unit of kinematic viscosity, cSt, for the current study. In the remainder of the paper, we will simply use  $\eta$  instead of  $\eta_k$  for kinematic viscosity unless specified.

#### Viscosity, T<sub>2,LM</sub>, and Diffusivity at ambient conditions

The experimental values of  $\eta/\eta_w$ , as a function of the salinity of selected brines at ambient conditions (30°C, 1atm) are plotted as a function of concentration in Figure 2(a). Figure 2(b) and (c) shows the percentage error when considering the viscosities of brines are same as those of pure water or NaCl solution, respectively.

Figure 2(a) shows the plot of  $\eta/\eta_w$ , where  $\eta$  is the viscosity of brine and  $\eta_w$  is that of pure water, with the concentration of brine varying from ~3 to ~335 kppm. Two distinctive observations can be made from Figure 2(a). It clearly shows the structural temperature effect mentioned previously. In other words, LiCl and NaCl strengthen the structure of water, whereas KCl and CsCl weaken the structure. Thus, LiCl and NaCl are found to cause solutions more viscous (*kosmotropes*); and KCl and CsCl solutions, less viscous (*chaotropes*), than pure water. Interestingly, the KCl viscosity at an ambient condition is not much different from water itself even in the high-concentration range. In general, this structural temperature effect is much more prominent at higher ionic solute concentrations as seen from the last term of Eq. (1).

In order to estimate the error that might occur should one have treated any brine viscosity by the viscosity of pure water, we plotted in Figure 2(b) the percentage errors of  $\eta/\eta_w$ values of brines by considering them as those of pure water. Firstly, we observe that the error percentage depends on the solute concentration. At the salt saturation level, we see the error percentage of the most common formation brine, NaCl, from Figure 2(b) is about 40%.

In Figure 2(c), the error percentage plots of the  $\eta/\eta_w$  values of various brines by considering them as those of the NaCl solution as a function of concentration. The viscosity of CsCl solution at 30°C and 1atm can be overestimated as large as twice of its actual value by treating it as an NaCl solution.

In the logging industry, NMR relaxation times of oil and water are separated through a specific logging pulse scheme such as multiple- $G \cdot TE$  and/or multiple-TW [7-9]. Then, NMR-based hydrocarbon viscosity estimates are derived from the correlation between viscosity and relaxation time or diffusivity [10]. For formation water, the relaxation time variation in response to different salt types and salinity has not received much attention. The logarithmic mean of the translational relaxation time,  $T_{2,LM}$ , and NMR-derived diffusivity vs. the solute concentration of selected brines are plotted in Figure 3(a) and (b), respectively. The values of  $T_{2,LM}$  multiplied by viscosity are plotted as a function of salinity in Figure 3(c). In agreement with the measured viscosity, the experimental values of  $T_{2,LM}$  and diffusivity also show either positive or negative structural temperature effects from selected brines.

Although quite large differences are observed in  $T_2$  between pure water and various brines in high solute concentrations, they are less significant (except for the exceptions to be discussed later) in terms of interpreting NMR logs. The reason is water, as a wetting phase, is dominated by the surface relaxation mechanism rather than the bulk relaxation,

thus the effect due to the change of brine  $T_2$  from the water  $T_2$  lessens when considering the stronger effect of surface relaxation. Exceptions to the above argument are for cases in which the surface relaxation influence is weak. The first scenario is formations containing large vugs. Some carbonate reservoirs contain large size vugs with a  $T_2$ relaxation time less affected by a surface relaxation mechanism. For salt-saturated carbonate reservoirs, often found in Middle East, the use of brine  $T_2$  instead of water  $T_2$ for the bulk relaxation time may be important for accurately evaluating these formations. The second scenario is for oil wet rock formations. For oil wet rock, the formation water is a non-wetting phase; thus the relaxation time of the water phase is not dominated by surface relaxation. On the other hands, some new NMR log interpretation techniques require the forward modeling of fluid properties. Inaccurate inputs of water properties (e.g.,  $T_2$ , viscosity, and diffusivity) may result in not only the erroneous estimation of water  $T_2$ , but also hydrocarbon saturation because the NMR inversion model is nonorthogonal. Furthermore, the recently developed 2DNMR technique [11] improves the sensitivity of diffusion and the estimation of intrinsic relaxation time. This improved technique provides knowledge of viscosity (and, thus, diffusivity) variation with salt contents that helps to correctly discern the restricted diffusion effect from salt-induced diffusivity changes.

The diffusivity plot illustrates we can treat most diffusivities of *chaotrope* brines as pure water diffusivity. With the *kosmotrope* brines, however, it would be desirable to use the correct brine diffusivity when interpreting NMR log data. This is especially important for NMR-based fluid-typing applications such as discerning very light oil and water. Because the light oil and water diffusivities are close, eliminating the uncertainty of water diffusivity is important [12].

Figure 3(c) shows the plot of  $T_{2,LM} \times \eta$  (msec·cSt) as a function of salinity of selected brines. As can be seen from ref. 11, the value of  $T_{2,LM} \times \eta$  has been treated as a constant at fixed temperature whether the fluid is brine or oil. Our study reveals the value of  $T_{2,LM}$  $\times \eta$  is not a constant at a fixed temperature over different salinity. This different behavior of *kosmotrope* brines and *chaotrope* brines relative to water is clearly shown. The observed characteristics of *kosmotrope* and *chaotrope* brines from Figure 3(c) agree well with the viscosity measurements from Figure 2.

#### Viscosity, T<sub>2,LM</sub>, and Diffusivity at various temperature

The roles of temperature on viscosity,  $T_{2,LM}$  and diffusivity of brines were also investigated. The following three figures (4, 5, and 6) present the same experimental data differently to assist with a better understanding of the temperature effects on the viscosity. The viscosity measurements of brines at 3 different temperatures are shown in Figure 4.

Unlike brines with low salinity, the increase of temperature alters the viscosity characteristics of *chaotrope* brines with high salinity. This phenomenon can be observed easily by investigating the general appearance of brine viscosity relative to pure water

viscosity (red solid line) in Figure 4. Investigation of the  $\eta/\eta_w$  value of all brines with high salinity over different temperatures can be used as confirmation. The  $\eta/\eta_w$  values of brines with concentration of 250 kppm at 3 different temperatures from Figure 4 are listed in Table 2.

For all temperatures, the viscosity of an LiCl and NaCl (*kosmotropes*) solution does not change relative to that of pure water. The viscosity of KCl and CsCl (*chaotropes*) solution, however, increases as the temperature increases relative to that of pure water. The KCl solution, especially, becomes thicker than pure water above 50°C. As the temperature increases, the principal term that determines the viscosity of KCl and CsCl (*chaotropes*) solution changes from the structural temperature to Brownian motion from Eq. (1). In other words, for high temperature, the addition of *chaotropes*, such as KCl and CsCl, strengthen the water structure just like *kosmotropes*. The Brownian motion term of Eq. (1) is proportional to concentration and temperature so that it becomes a more dominant term at high concentration and temperature as shown from Figure 4 and Table 2.

Figure 5 plots the normalized viscosity vs. salinity for four selected brines at various temperatures. Figure 5(a) and (b) shows the values of  $\eta/\eta_w$  of LiCl and NaCl are independent of temperature. The viscosity of KCl and CsCl, however, shows considerable temperature dependence (Figure 5(c) and (d)). The transition of the dominant term from the structural temperature to the Brownian motion determines the viscosity of the ionic solution can be seen clearly from Figure 5(c) and (d).

Figure 6 shows the plot of the normalized viscosity of brines as a function of temperature for four different salinities of four selected brines. From Figure 6, it becomes obvious the value of  $\eta/\eta_w$  is independent of the temperature in the case of the solution with *kosmotropes* (LiCl and NaCl). In other words, the viscosity values of LiCl and NaCl solutions increase with the concentration, but the value of  $\eta/\eta_w$  is constant over the range of temperatures. Unlike *kosmotropes*, the viscosity of *chaotropes* (KCl and CsCl) decreases with the concentration at a low temperature and increases with the concentration at a high temperature. In addition, the  $\eta/\eta_w$  values of the brines with *chaotropes* increase slightly with the temperature.

## CONCLUSION

Our experimental study involving a series of viscosity,  $T_2$ , and diffusivity measurements demonstrates the composition of brine should be addressed if an accurate evaluation of formation water is to be reached. The deviated behavior of a non-NaCl solution from an NaCl solution is more apparent at a low temperature and high concentration. At 30°C and 1atm, the viscosity of a 335-kppm KCl and CsCl can be overestimated by ~70% and ~100%, respectively, of the actual value by treating them identical to NaCl.

In addition, we have observed two interesting aspects about KCl. First, the viscosity, thus the  $T_2$  and the diffusivity of KCl, does not differ much from that of water. Second, KCl

changes its role on changing the viscosity of brine at  $\sim 50^{\circ}$ C. The KCl raises the structural temperature of brine below  $\sim 50^{\circ}$ C and lowers it above  $\sim 50^{\circ}$ C.

We found an error up to 100% may occur if we use the known viscosity and NMR  $T_2$  or diffusivity correlations, such as  $T_2$  (sec) =  $3T_k/298 \eta_d$  or D (cm<sup>2</sup>/sec) =  $(2T_k/298 \eta_d) \times 10^{-5}$  (ref. 10) for different types of brines and solute concentrations. Thus, using the viscosity of pure water for all brines is not recommended. Our study further illustrates considering the concentration term and temperature term with brine specific coefficients improves the correlation between  $T_2$  and the viscosity of brines. The selected four solute types and the broad concentration ranges covered almost the full spectrum of the brine occurring in the earth formation and drilling mud. The results then can be used for improving the prediction of viscosity in other concentrations, solute mixtures, and for log interpretation. The knowledge of the brine viscosity change also can be helpful for designing drilling mud, providing correct inputs to reservoir simulation, and computing reservoir fluid mobility and relative and effective permeability estimation.

### NOMENCLATURE

- *A* = Jones-Dole viscosity A coefficient
- B = Jones-Dole viscosity B coefficient
- C = concentration
- $D = diffusivity (cm^2/sec)$
- $f_{(a)}$  = Brownian coefficient
- $f_{(b)}$  = Debye-Hückel coefficient
- $f_{(c)}$  = Structural Temperature coefficient
- G = magnetic field gradient
- $M_A$  = Mass of A<sup>+</sup> ion
- $M_X$  = Mass of X<sup>-</sup> ion
- $r_A$  = ionic radii of A<sup>+</sup> ion
- $r_X$  = ionic radii of X<sup>-</sup> ion
- T = Temperature
- $T_2$  = apparent transverse relaxation time
- $T_{2,LM}$  = logarithmic mean of transverse relaxation time
- $T_k$  = absolute temperature (Kelvin)
- TE = inter-echo time
- *TW* = wait time, recycling time
- z = charge of ion
- $\eta$  = fluid viscosity
- $\eta_w$  = viscosity of pure water
- $\eta_k$  = kinematic viscosity (cSt)
- $\eta_d$  = dynamic viscosity (cP)
- $\rho$  = fluid density (g/ml)

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## TABLES

Cations	В	Anions	В
Mg <sup>2+</sup>	0.385	PO4 <sup>3-</sup>	0.590
Ca <sup>2+</sup>	0.285	CH <sub>3</sub> CO <sub>2</sub> <sup>-</sup>	0.250
Ba <sup>2+</sup>	0.22	$SO_42^-$	0.208
$\mathrm{Li}^{+}$	0.150	F	0.10
Na <sup>+</sup>	0.086	HCO <sub>2</sub> -	0.052
$\mathbf{K}^{+}$	-0.007	Cľ	-0.007
$\mathrm{NH_4}^+$	-0.007	Br⁻	-0.032
Rb⁺	-0.030	NO <sub>3</sub> -	-0.046
Cs <sup>+</sup>	-0.045	ClO <sub>4</sub> -	-0.061
		ľ	-0.068
		SCN <sup>-</sup>	-0.103

 Table 1.
 Jones-Dole viscosity B coefficients [5].

Table 2.	The $\eta/\eta_{w}$	values of brines with salt concentration of 250 kppm at 30°C, 50°C, and 70°C.

	30°C	50°C	70°C
LiCl	1.84	1.84	1.85
NaCl	1.41	1.41	1.40
KCl	0.94	1.01	1.05
CsCl	0.83	0.88	0.90

#### **FIGURES**



Figure 1. The plot of the viscosity of brine/viscosity of water  $(\eta/\eta_w)$  vs. concentration of ionic solutes (5, 50, 150, and 250 kppm) for selected brines in a unit of kinematic viscosity, cSt, (a), and dynamic viscosity, cP, (b). The selected brines are LiCl ( $\blacklozenge$ ), NaCl ( $\blacksquare$ ), KCl ( $\blacktriangle$ ), and CsCl ( $\circlearrowright$ ). The red solid line represents water and was drawn as a reference. All measured data are fitted with a 2<sup>nd</sup> order polynomial to help visualize the trends.



Figure 2. (a) The plot of viscosity of brine/viscosity of water  $(\eta/\eta_w)$  vs. concentration of ionic solutes in selected brines. (b) The plot of difference % of  $\eta/\eta_w$  for selected brines from that for pure water. (c) The plot of difference % of  $\eta/\eta_w$  for selected brines from that for NaCl. The selected brines are LiCl ( $\blacklozenge$ ), NaCl ( $\blacksquare$ ), KCl ( $\blacktriangle$ ), and CsCl ( $\blacklozenge$ ). The red solid lines were drawn as a reference ((a) and (b) : pure water, (c) : NaCl solution). All measured data are fitted with a 2<sup>nd</sup> order polynomial to help visualize the trends.



Figure 3. (a) The plot of  $T_{2,LM}$  vs. concentration of brines. (b) The plot of Diffusivity vs. concentration of brines. (c) The plot of  $T_{2,LM} \times \eta$  vs. concentration of brines. All experiments are conducted at ambient condition (30°C and 1 atm). The selected brines are LiCl ( $\blacklozenge$ ), NaCl ( $\blacksquare$ ), KCl ( $\blacktriangle$ ), and CsCl ( $\bullet$ ). The  $T_{2,LM}$ , Diffusivity, and  $T_{2,LM} \times \eta$  of pure water are plotted as a red solid line for all figures as a reference. All measured data are fitted with a 2<sup>nd</sup> order polynomial to help visualize the trends.



Figure 4. The plot of  $\eta/\eta_w$  vs. concentration of ionic solutes in selected brines at (a) 30°C, (b) 50°C, and (c) 70°C with 1atm. The selected brines are LiCl ( $\blacklozenge$ ), NaCl ( $\blacksquare$ ), KCl ( $\blacktriangle$ ), and CsCl ( $\blacklozenge$ ). The red solid lines represent pure water. All measured data are fitted with a 2<sup>nd</sup> order polynomial to help visualize the trends.



Figure 5. The plot of  $\eta/\eta_w$  vs. the concentration of (a) LiCl, (b) NaCl, (c) KCl, and (d) CsCl solution at 5 different temperatures and the pressure of 1 atm. The symbol represents different temperatures (30°C ( $\blacklozenge$ ), 40°C ( $\blacksquare$ ), 50°C ( $\blacktriangle$ ), 60°C ( $\circlearrowright$ ), and 70°C ( $\circledast$ ). In (a) and (b), all data points are fit together. All measured data are fitted linearly to help visualize the trends.



Figure 6. The plot of  $\eta/\eta_w$  vs. temperature of brines with the concentration of (a) 30 kppm, (b) 150 kppm, (c) 250 kppm, and (d) 335 kppm at 1 atm. The selected brines are LiCl ( $\blacklozenge$ ), NaCl ( $\blacksquare$ ), KCl ( $\blacktriangle$ ), and CsCl ( $\bullet$ ).