Influences of Na$_2$O and K$_2$O Additions on Electrical Conductivity of CaO-SiO$_2$-(Al$_2$O$_3$) Melts

Guo-Hua ZHANG,* Wei-Wei ZHENG, Shuqiang JIAO and Kuo-Chih CHOU

State Key Laboratory of Advanced Metallurgy, University of Science and Technology Beijing, Beijing, 100083 China.

(Received on May 18, 2017; accepted on August 8, 2017)

The present study investigated the influences of Na$_2$O and K$_2$O on the electrical conductivity of CaO-SiO$_2$-(Al$_2$O$_3$) melts by the four electrode method. From the experimental results, it was found that the temperature dependence of electrical conductivity obeys the Arrhenius law. As adding Na$_2$O or K$_2$O to CaO-SiO$_2$-(K$_2$O) melts, electrical conductivity monotonously decreases. Meanwhile, Na$_2$O bearing melt always has a larger value of electrical conductivity than K$_2$O bearing melt when the compositions are the same. By substituting K$_2$O for Na$_2$O, the mixed alkali effect occurs that electrical conductivity first decreases and then increases with the substitution amount of K$_2$O. Furthermore, the mixed alkali effect is more evident for melts with Al$_2$O$_3$ than that without Al$_2$O$_3$.

KEY WORDS: electrical conductivity; arrhenius law; mixed alkali effect.

1. Introduction

Electrical conductivity is one of the most important thermophysical properties of oxide melts. It only plays a prominent role in the design and optimization of electric smelting furnaces, but also develops base knowledge about the structures of oxide melts. For instance, the earliest evidence of the ionic structure of oxide melts resulted from electrical conductance measurements.$^{1-3}$ However, much less attention has been paid on electrical conductivity due to the difficulty of experimental measurements at high temperatures. Limited data not only couldn’t provide enough fundamental supports to the practical production, but also misleads the modeling work about electrical conductivity. When modeling electrical conductivity of molten slags, Thibodeau and Jung$^{4}$ stated that electrical conductivity monotonously increases when CaO is gradually replaced by Al$_2$O$_3$ at constant SiO$_2$ content in CaO–Al$_2$O$_3$–SiO$_2$ melts. However, the minimum values of electrical conductivity were found in both CaO–Al$_2$O$_3$–SiO$_2$ melts$^{5}$ and CaO–FeO$_x$–Al$_2$O$_3$–SiO$_2$ melts$^{6}$ with the change of CaO/Al$_2$O$_3$ molar ratio. Therefore, much more accurate data of electrical conductivity are urgently needed. CaO-(Al$_2$O$_3$)-SiO$_2$ system is a fundamental system in both glasses and metallurgical slags, and Na$_2$O and K$_2$O are widely used as additives. Therefore, in the present study, the influences of Na$_2$O and K$_2$O on the electrical conductivity of CaO-(Al$_2$O$_3$)-SiO$_2$ system will be investigated.

2. Materials and Experiments

The compositions of samples are shown in Table 1, in which the molar fraction of CaO to SiO$_2$ for all the compositions is 1.1. These samples are classified into five different groups: In groups A and B, contents of Na$_2$O and K$_2$O gradually increase, respectively; in group C, no Al$_2$O$_3$ was presented, and from B3, to C1, to C2 and to A3, K$_2$O was gradually replaced by Na$_2$O; in groups D and E, content of Al$_2$O$_3$ keeps constant, while the substitution amount of Na$_2$O for K$_2$O increases from D0 to D3, and E0 to E3,

| Compositions | CaO | SiO$_2$ | Al$_2$O$_3$ | Na$_2$O | K$_2$O | CaO/ SiO$_2$ |
|--------------|-----|--------|-------------|--------|------|--------------|
| A0           | 0.524 | 0.476 | 0 | 0 | 0 | 1.1 |
| A1           | 0.513 | 0.467 | 0.02 (0.018) | 0 | 0 | 1.1 |
| A2           | 0.503 | 0.457 | 0.04 (0.037) | 0 | 0 | 1.1 |
| A3           | 0.492 | 0.448 | 0.06 (0.058) | 0 | 0 | 1.1 |
| B1           | 0.513 | 0.467 | 0 | 0.02 (0.019) | 1.1 |
| B2           | 0.503 | 0.457 | 0.04 (0.037) | 0 | 1.1 |
| B3           | 0.492 | 0.448 | 0.06 (0.057) | 0 | 1.1 |
| C1           | 0.492 | 0.448 | 0.04 (0.037) | 0.02 (0.019) | 1.1 |
| C2           | 0.492 | 0.448 | 0.02 (0.018) | 0.04 (0.039) | 1.1 |
| D0           | 0.478 | 0.434 | 0.028 | 0.06 (0.057) | 0 | 1.1 |
| D1           | 0.478 | 0.434 | 0.028 | 0.04 (0.037) | 0.02 (0.018) | 1.1 |
| D2           | 0.478 | 0.434 | 0.028 | 0.03 (0.019) | 0.04 (0.037) | 1.1 |
| D3           | 0.478 | 0.434 | 0.028 | 0.06 (0.058) | 0 | 1.1 |
| E0           | 0.452 | 0.41 | 0.078 | 0.06 (0.058) | 0 | 1.1 |
| E1           | 0.452 | 0.41 | 0.078 | 0.04 (0.037) | 0.02 (0.019) | 1.1 |
| E2           | 0.452 | 0.41 | 0.078 | 0.02 (0.018) | 0.04 (0.039) | 1.1 |
| E3           | 0.452 | 0.41 | 0.078 | 0.06 (0.059) | 0 | 1.1 |

* Corresponding author: E-mail: ghzhang_ustb@163.com
DOI: http://dx.doi.org/10.2355/isijinternational.ISIJINT-2017-285
respectively. Slag samples were prepared using reagent grade CaCO₃, SiO₂, Al₂O₃, Na₂CO₃, and K₂CO₃ powders. SiO₂, Al₂O₃, and CaCO₃ were calcined at 1 273 K for 10 h in a muffle furnace, while Na₂CO₃ and K₂CO₃ were calcined at 873 K for 10 h. Then the reagents were precisely weighted according to the compositions shown in Table 1, and mixed in an agate mortar thoroughly. The mixtures were kept for the following experiments.

The schematic diagram of the experimental apparatus is shown in Fig. 1. Before measuring resistance of slags, the standard aqueous KCl (1.0 Dimal) solution was used for cell calibration at temperatures from 293 K to 295 K. After obtaining the cell constant, the prepared slag sample was packed into a platinum crucible and then placed in the constant temperature zone of the furnace with the heating elements of MoSi₂. After the temperature measured by a type B (Pt-6 pct Rh/Pt-30 pct Rh) thermocouple reached to 1 873 K and held for 2 h (which is enough for the decomposition of carbonate and escape of CO₂ as well as the uniformity of melts based on our preliminary experiments), the electrodes were lowered slowly until touching the surface of the melt. During this process, the resistance was continuously monitored. When the tips of the electrodes contacted with the surface of melt, the resistance significantly decreased. From this critical point, the electrodes were lowered 3 mm which is the same as that during the cell calibration. The resistance measurement was carried out at every 50 K interval from 1 873 K to 1 673 K. At each temperature before measuring, the melt was kept for 30 min first to ensure the equilibrium. All the measurements were carried out at the frequency of 20 kHz. It was concluded by Kim⁷) that when the content of Na₂O or K₂O in aluminosilicate melts was 25 mol%, the vaporization of Na₂O or K₂O at high temperature was less than 4%. In our previously study, it was also found that the vaporazation of K₂O was very small when its content was 5 mol%.⁸) The composition analyses of samples after experiments were also carried out by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES), results of which are shown in the brackets of Table 1. It can be concluded that the vaporization of Na₂O and K₂O can be neglected under the present experimental condition.

3. Results

In the literatures, the electrical conductivity of CaO–SiO₂ slags with CaO/SiO₂ molar ratio of 1.1 are collected and shown in Table 2. From Table 2, it can be seen that there are not so large differences between the present measured electrical conductivity data and these from the literatures. In Table 2, electrical conductivity from the work of Bockris et al., Mori et al., Keller⁹) were measured by two electrode method, while that from Berryman¹⁰) was measured by central electrode. In order to get accurate electrical conductivity values, it is necessary to exclude the resistances of lead wires and electrodes from the total measured resistance. If the current electrode is used in common for the potential one, contribution of interfacial resistance is usually too large to be disregarded. The four electrode technique can avoid those difficulties. Therefore, the present results should be more reliable.

The measured data of electrical conductivities for different compositions in Table 1 at different temperatures are given in Table 3. According to the values in Table 3, the logarithm of electrical conductivity for groups A, B, C, D and E as functions of the reciprocal of temperature are displayed in Figs. 2, 3, 4, 5, and 6. From these Figures it

![Fig. 1. Schematic diagram of the experimental apparatus.](image)

Table 2. Comparison with data from literatures for CaO–SiO₂ system (CaO/SiO₂=1.1).

| Source      | Bockris et al. | Mori et al. | Keller | Berryman | Present study |
|-------------|----------------|-------------|--------|----------|--------------|
| Ref. [7]    | 0.41           | 0.25        | 0.3    | 0.4      | 0.317        |
| 1 823 K     | 0.5            | 0.32        | 0.42   | 0.5      | 0.392        |
| 1 873 K     | 0.5            | 0.32        | 0.42   | 0.5      | 0.392        |

Table 3. Electrical conductivity for different compositions, Ω⁻¹cm⁻¹.

| Compositions | 1 873 K | 1 823 K | 1 773 K | 1 723 K | 1 673 K |
|--------------|---------|---------|---------|---------|---------|
| A0           | 0.392   | 0.319   | 0.251   | 0.195   | 0.146   |
| A1           | 0.491   | 0.408   | 0.325   | 0.257   | 0.195   |
| A2           | 0.529   | 0.430   | 0.355   | 0.287   | 0.227   |
| A3           | 0.589   | 0.509   | 0.432   | 0.365   | 0.299   |
| B1           | 0.427   | 0.356   | 0.282   | 0.226   | 0.175   |
| B2           | 0.502   | 0.421   | 0.335   | 0.268   | 0.209   |
| B3           | 0.556   | 0.488   | 0.416   | 0.344   | 0.268   |
| C1           | 0.545   | 0.452   | 0.371   | 0.303   | 0.241   |
| C2           | 0.517   | 0.427   | 0.353   | 0.288   | 0.229   |
| D0           | 0.418   | 0.309   | 0.242   | 0.194   | 0.149   |
| D1           | 0.200   | 0.170   | 0.138   | 0.107   | 0.085   |
| D2           | 0.228   | 0.196   | 0.158   | 0.127   | 0.096   |
| D3           | 0.245   | 0.209   | 0.168   | 0.134   | 0.100   |
| E0           | 0.259   | 0.217   | 0.191   | 0.159   | 0.139   |
| E1           | 0.166   | 0.146   | 0.126   | 0.106   | 0.085   |
| E2           | 0.193   | 0.171   | 0.144   | 0.119   | 0.093   |
| E3           | 0.212   | 0.188   | 0.161   | 0.136   | 0.109   |
can be concluded that there is always a linear relationship between the logarithm of conductivity and the reciprocal of temperature. In other words, the temperature dependence of conductivity obeys the Arrhenius law as follows,

\[ \ln \sigma = \ln \sigma_0 - \frac{E}{RT} \] .......................... (1)

where \( \sigma \) is the electrical conductivity, \( \Omega^{-1} \cdot \text{cm}^{-1} \); \( \sigma_0 \) is the pre-exponent factor, \( \Omega^{-1} \cdot \text{cm}^{-1} \); \( E \) is the activation energy of electrical conductivity, J/mol; \( R \) is the gas constant, 8.314 J/(mol·K); \( T \) is the absolute temperature, K.

From Figs. 2 and 3, it can be concluded that as gradually increasing Na\(_2\)O and K\(_2\)O contents, while keeping the molar ratio of CaO to SiO\(_2\) constant, both the electrical conductivity of CaO–SiO\(_2\)–Na\(_2\)O and CaO–SiO\(_2\)–K\(_2\)O melts increase.

From Figs. 4, 5 and 6, as substituting Na\(_2\)O for K\(_2\)O, the electrical conductivity first decreases and then decreases. Or, there is a minimum value of electrical conductivity with the change of K\(_2\)O/R\(_2\)O (R=Na, K).

4. Discussion

4.1. Influence of R\(_2\)O Addition on Electrical Conductivity of Groups A and B

Since there is no transition metal oxide in all the studied compositions in Table 1, the electronic conductance can be neglected, and the charge transport is mainly completed by ions. Generally, contributions of Si\(^{4+}\) and Al\(^{3+}\) ions to
the charge conductance are very small due to their large valences and small ionic radii which lead to large interactions with the nearby ions.\textsuperscript{13) For the case of oxygen ion, there are three types of oxygen ions in oxide melts based on the classification of Fincham and Richardson:\textsuperscript{13)} bridging oxygen, bonded with two cations from acidic oxides, e.g. Si\textsuperscript{4+}, or Al\textsuperscript{3+} in [AlO\textsubscript{4}] tetrahedron after being charge balanced; non-bridging oxygen, bonded with one cation from acidic oxide and one cation from basic oxide; free oxygen, bonded with two cations from basic oxides. The mobile ability of bridging oxygen or non-bridging oxygen are very weak since they are bonded with Si\textsuperscript{4+} ion or Al\textsuperscript{3+} ion and form the covalent bond dominated chemical bond. Whereas, the mobile ability of the free oxygen ion is large since it forms the ionic bond dominated chemical bond with metal cation from the basic oxide. However, concentration of free oxygen ion for all compositions shown in Table 1 should be very small because of the low basicity. Thereby, contribution of oxygen ion to the charge conductance can also be neglected. Therefore, in the present study, only the contributions from Ca\textsuperscript{2+}, Na\textsuperscript{+} and K\textsuperscript{+} ions are considered.

Generally, the ionic conductance of oxide melt is determined by the concentration of mobile ion and the degree of polymerization.\textsuperscript{13) The larger the concentration of mobile ion and the lower the degree of polymerization, the larger the conductivity will be. For the CaO–SiO\textsubscript{2}–R\textsubscript{2}O (R = Na, K) systems as shown in group A and group B, it can be seen from Figs. 2 and 3 that the electrical conductivity monotonously increases as increasing R\textsubscript{2}O content. The reason for this is that the addition of R\textsubscript{2}O increases the concentration of mobile ions (Ca\textsuperscript{2+} and R\textsuperscript{+}) ions. Furthermore, the degree of polymerization also decreases due to the increase of concentration of basic oxides (CaO and R\textsubscript{2}O) which form more non-bridging oxygen and destroy the network of melt.

4.2. Different Influences of Na\textsubscript{2}O and K\textsubscript{2}O on Electrical Conductivity

From the composition shown in Table 1, it can be seen that for compositions A1 and B1, A2 and B2, A3 and B3, D0 and D3, as well as E0 and E3, the contents of CaO, SiO\textsubscript{2}, Al\textsubscript{2}O\textsubscript{3} and R\textsubscript{2}O are the same, respectively. So, the electrical conductivity data of these compositions can be compared to distinguish the different influences of Na\textsubscript{2}O and K\textsubscript{2}O. The logarithm of electrical conductivity for these compositions as the function of the reciprocal of temperature for these compositions is plotted and shown in Fig. 7, from which it is concluded that the electrical conductivity of Na\textsubscript{2}O bearing melts are higher than K\textsubscript{2}O bearing melts. The reasons will be analyzed as follows.

Since the contents of Na\textsubscript{2}O and K\textsubscript{2}O are the same for different groups shown in Fig. 7, the concentrations of mobile ions (Ca\textsuperscript{2+}, Na\textsuperscript{+} and K\textsuperscript{+}) are approximately the same. Herein, the structure variations of Na\textsubscript{2}O bearing and K\textsubscript{2}O bearing melts will be discussed. It has been proved that when there are several basic oxides in the melts containing Al\textsubscript{2}O\textsubscript{3}, there is a strict order for which cations carry out the charge-compensation of the Al\textsuperscript{3+} ions: K\textsuperscript{+} > Na\textsuperscript{+} > Ca\textsuperscript{2+}.\textsuperscript{16–18) Strictly, even if there are enough basic oxides, not all the Al\textsuperscript{3+} ions exist as [AlO\textsubscript{4}], and some Al\textsuperscript{3+} ions still exist with a high oxygen coordination, such as [AlO\textsubscript{5}] or [AlO\textsubscript{6}].\textsuperscript{19) Furthermore, the stronger the compensation ability of R\textsuperscript{+} ion, the less the concentration of high oxygen coordination [AlO\textsubscript{5}] and [AlO\textsubscript{6}] will be, which will result in a higher degree of polymerization. Therefore, K\textsubscript{2}O will make the concentration of [AlO\textsubscript{5}] or [AlO\textsubscript{6}] much less or the degree of polymerization much higher. The similar conclusion was also proved by Sukenaga \textit{et al.}\textsuperscript{20) that the [AlO\textsubscript{5}] content in the CaO–Al\textsubscript{2}O\textsubscript{3}–SiO\textsubscript{2}–R\textsubscript{2}O (R = Li, Na, K) systems followed the order: CAS > CASL > CASN > CASK. Therefore, the K\textsubscript{2}O bearing aluminosilicate melts will have a large degree of polymerization than Na\textsubscript{2}O bearing melts, provided the contents of other components are the same.

Besides the concentration of mobile ion and the degree of polymerization, ion radius also affects the electrical conductivity. Generally, for cations with the same valence, the smaller the radius, the greater the mobility will be. However, when the cation has a high valence, the polarization of the cation should also be considered. In this case, the smaller cation has a larger polarizing ability, which leads to a stronger interaction with the nearby anion ions, thus a great resistance will be. For the alkali metal ion, the polarization ability is normally weak due to its monovalence. Therefore, it is easier for small cation to transport. By measuring the electrical conductivity of PbO–SiO\textsubscript{2} melts as adding various oxides, Sugimoto \textit{et al.}\textsuperscript{21)} also found that in the case of alkali oxide addition, the electrical conductivity decreases with increasing ionic radius of cation, whereas the opposite tendency was found in the case of alkali earth oxide addition. Therefore, in the present study, K\textsuperscript{+} ion with a larger ion radius than Na\textsuperscript{+} ion (1.33 Å v.s 0.97 Å\textsuperscript{22}) could block or hinder the pathways of ion conduct and lead to the further deterioration of conductance.

According to the above analyses, it can be concluded that both the larger degree of polymerization and smaller mobile ability of K\textsuperscript{+} ion can result in a smaller electrical conductivity of K\textsubscript{2}O bearing melts relative to Na\textsubscript{2}O bearing melts.

4.3. The Mixed Alkali Effect in Groups C, D and E

From Figs. 4–6, it can be seen that as gradually substituting Na\textsubscript{2}O for K\textsubscript{2}O while keeping the contents of other components constant, electrical conductivity first decreases and then increases. In order to clearly see this tendency, the change of electrical conductivity as a function of K\textsubscript{2}O/2R\textsubscript{2}O
for groups C, D and E are displayed in Fig. 8. It is evident that there is a minimum value of electrical conductivity with the change of K$_2$O/ΣR$_2$O ratio. This extreme departure from linearity is called the “mixed alkali” effect, which is very common in glasses. As stated by Swenson et al., the alkali ions tend to preserve their local structural environment from the single glasses regardless of the glass composition. The distribution of the two types of cations in the structure is predominantly random. A large mismatch exists between the different alkali ion sites, and there are effectively less sites available for ionic motion. There, in the present study, this mismatch between the local potential of Na$^+$ and K$^+$ sites may lead to a high activation energy for ionic jumps to dissimilar sites, which results in the presence of minimum value of electrical conductivity (analysis about the activation energy will be given in the following section). Furthermore, by comparing Figs. 8(b) and 8(c) with Fig. 8(a), it can be seen that the mixed alkali effect is much more evident for the Al$_2$O$_3$ bearing melts than melts without Al$_2$O$_3$. The complex structure changes for the Al$_2$O$_3$ bearing melts should be the inherent cause, even if the detailed reason is still unclear.

4.4. The Activation Energy of Electrical Conductivity

According to Eq. (1) and the experimental data shown in Table 3, the activation energies of electrical conductivity for different compositions can be calculated and shown in Fig. 9. Figure 9(a) exhibits the change of activation energy with R$_2$O content for group A and group B. It can be con-

---

**Fig. 8.** Change of electrical conductivity with the substitution amount of K$_2$O for melts with (a) 0% Al$_2$O$_3$; (b) 0.028 mol% Al$_2$O$_3$; (c) 0.078 mol% Al$_2$O$_3$.

**Fig. 9.** Activation energy of electrical conductivity for different groups.
cluded that for melts without Al\textsubscript{2}O\textsubscript{3}, the activation energy monotonously decreases as increasing R\textsubscript{2}O content, since in this case R\textsubscript{2}O only acts as the network modifier and decreases the degree of polymerization of melts. Thus, there will be a decrease of activation energy.

Figure 9(b) shows the activation energies of electrical conductivity for different compositions in groups C, D and E. It can be seen that as increasing the substitution content of K\textsubscript{2}O, the activation energy first increases but then decreases. Or, there is a maximum value of activation energy. By combining Fig. 8 with Fig. 9(b), it can be concluded that the electrical conductivity and the activation energy have opposite variation tendencies. Furthermore, the maximum value of activation energy and the minimum value of electrical conductivity almost occur at the same composition. The change of melt structure resulting from the mixed alkali effect forms a large energy barrier at a certain composition, which retards the transfer of mobile ion and leads to the minimum of electrical conductivity.

5. Conclusions

The present study investigated the influences of Na\textsubscript{2}O and K\textsubscript{2}O on the electrical conductivity of CaO-SiO\textsubscript{2}-(Al\textsubscript{2}O\textsubscript{3}) melts by the four electrode method. The following conclusions could be drawn.

(1) The temperature dependence of electrical conductivity obeys the Arrhenius law.

(2) Both additions of Na\textsubscript{2}O and K\textsubscript{2}O could increase the electrical conductivity of CaO–SiO\textsubscript{2} melts.

(3) The electrical conductivity of Na\textsubscript{2}O bearing melts are higher than that of the K\textsubscript{2}O bearing melts if the contents of other components are the same.

(4) As gradually substituting Na\textsubscript{2}O for K\textsubscript{2}O, there is a minimum value of electrical conductivity. Futhermore, existence of Al\textsubscript{2}O\textsubscript{3} could increase this mixed alkali effect.

Acknowledgement

Thanks are given to the financial supports from the National Natural Science Foundation of China (51474141 and 51474020).

REFERENCES

1) P. Harasymenko: Trans. Faraday Soc., 34 (1938), 1245.
2) A. E. Martin and G. Derge: Trans. AIME, 154 (1943), 104.
3) J. O’M. Bockris and J. W. Tomlinson: Research, 2 (1949), 162.
4) E. Thibodeau and J. H. Jung: Metall. Mater. Trans. B, 47 (2016), 355.
5) J. H. Liu, G. H. Zhang and K. C. Chou: Can. Metall. Q., 54 (2015), 170.
6) J. H. Liu, G. H. Zhang and K. C. Chou: ISIJ Int., 55 (2015), 2325.
7) K. D. Kim: J. Am. Ceram. Soc., 79 (1996), 2422.
8) G. H. Zhang and K. C. Chou: Metall. Mater. Trans. B, 43 (2012), 841.
9) J. O’M. Bockris, J. A. Kitchener, S. Ignatowicz and J. W. Tomlinson: Trans. Faraday Soc., 48 (1952), 75.
10) K. Mori and Y. Matsushita: Tetsu-to-Hagané, 38 (1952), 283.
11) H. Keller, K. Schwerdtfeger and K. Hennesen: Metall. Trans. B, 10 (1979), 67.
12) R. A. Berryman: Doctoral dissertation, University of Toronto, (1989).
13) G. H. Zhang and K. C. Chou: J. Iron Steel Res. Int., 18 (2011), 13.
14) C. J. B. Fincham and F. D. Richardson: Proc. R. Soc. Lond. A, 223 (1954), 40.
15) G. H. Zhang, B. J. Yan, K. C. Chou and F. S. Li: Metall. Mater. Trans. B, 42 (2011), 261.
16) A. Navrotsky, G. Peraudeau, P. Mcmillan and J. P. Coutures: Geochim. Cosmochim. Acta, 46 (1982), 2039.
17) F. Domine and B. Piriou: Am. Mineral., 71 (1986), 38.
18) G. H. Zhang, K. C. Chou and K. Mills: Metall. Mater. Trans. B, 45 (2014), 598.
19) J. F. Stibbins and Z. Xu: Nature, 390 (1997), 60.
20) S. Sukenaga, T. Nagahisa, K. Kanehashi, N. Saito and K. Nakashima: ISIJ Int., 51 (2011), 333.
21) Y. Suginozawa, T. Yanagase and H. Ito: Trans. Jpn. Inst. Met., 3 (1962), 227.
22) R. D. Shannon: Acta Crystallogr., 32 (1976), 175.
23) J. Swenson, A. Matic, C. Karlsson, L. Borjesson, C. Meneghini and W. S. Howells: Phys. Rev. B, 63 (2001), 132202.