

Electrotransport of ^{22}Na and of ^{195}Au in Liquid Sodium

T. Persson and A. Lodding

Physics Department, Chalmers University of Technology,
Gothenburg, Sweden

(Z. Naturforsch. **28 a**, 1971–1972 [1973];
received October 22, 1973)

The mobilities of radioactive Na and Au tracers under electric current in liquid Na have been studied. In both cases the tracer is enriched at the anode, but the mobility of the impurity is by about two orders of magnitude greater than that of the Na tracer. The Haeffner-effect, for the first time studied with a radioactive isotope, yielded an "isotope factor" $\alpha \cong 0.2$ at 180 °C, of the same order of magnitude as in K and Rb. The effective charge number Z_{exp}^* for the electrotransport of Au in Na decreases from about -12 at 100 °C to -7.5 at 270 °C.

Isotope electrotransport in liquid metals (Haeffner-effect^{1,2}) has hitherto only been measured in such metals where the existence of several stable isotopes permits mass spectrometric analysis. Quantitative results have been obtained i. a. for Rb and K. We decided to study the effect also in Na, and for the first time utilize a radioactive isotope, i. e. by measuring the migration velocity of ^{22}Na at infinite dilution, relative to the matrix atoms with the atom mass 23. The relative mass difference (1:23) is here of the same order of magnitude as when using the stable isotopes of K (1:20), and although the available radioactive counting technique was not expected to give as good accuracy as the well established mass spectrometric analysis, at least semi-quantitative results were envisaged.

A capillary arrangement was used, of a type described in several earlier publications¹⁻³. The duration of each run was chosen so as not to approach the steady state in the 20 cm long capillary; however, in the runs with the maximal current density (9 A in 0.6 mm i. d. capillaries) a correction for convection effects had to be made. The need of this was realized when discovering that the specific activity far away from the anode did not, as theoretically expected, remain unchanged, but showed a certain depletion of the tracer. The correction, based on the difference in specific activity between the depleted region and reference samples, amounted to some 25% and introduced an extra uncertainty margin of about 15% in these runs.

The velocity of the tracer relative to the bulk was determined³ from

$$\Delta v = (l/t) (\Delta A/A_0), \quad (1)$$

where t is the duration of the run, l is the length along the capillary from the anode to a point P chosen where the tracer activity remained unchanged throughout the experiment; A_0 is the total tracer activity in that capillary portion before, and $A_0 + \Delta A$ after the experiment. The Haeffner-effect is then expressed by the magnitude of the "mass factor"^{1,2},

$$\alpha = (\Delta v/v_e) / (\Delta M/M), \quad (2)$$

where ΔM is the isotope mass difference, M the atom mass of the mixture (here that of the matrix), and

$$v_e = (D/kT) E e, \quad (3)$$

e being the electronic charge, k Boltzmann's constant, D the diffusion coefficient⁴, T abs. temperature and E the field strength.

The results are shown in Table 1. The accuracy was limited by the fluctuations in the counting sensitivity of the hollow-crystal single channel analyzer and by the uncertainty (particularly in the samples exhibiting convection effects, see above) concerning the original specific activity. In spite of the wide margins of error, it can be concluded from Table 1 that the mass factor is of the order of 0.5 at the m.p. and decreasing with rising temperature. In Table 1, a comparison is made with Haeffner-effect data of the other two alkali metals, taken from Ref.² and corrected according to the latest measurements of D in these metals⁴.

Metal	Temperature, °C	α
Na	137	0.40 ± 0.21
Na	162	0.26 ± 0.11
Na	164	0.19 ± 0.09
Na	224	0.16 ± 0.09
Na	225	0.13 ± 0.06
Na	226	0.15 ± 0.08
K	100	0.31
K	200	0.29
Rb	100	0.49
Rb	200	0.41

Table 1.
Isotope Factor
of Haeffner Effect
in Liquid
Alkali Metals.

As a part of a systematic investigation of impurity atom transport in liquid alkali metals³⁻⁵ we also studied the electrotransport of ^{195}Au in liquid sodium, employing the same experimental set-up as recently³ for ^{22}Na in K and for the Haeffner-effect study reported above. Again Δv was obtained via Equation (1). However, as the velocities of the impurity relative to the matrix here were of the order of 1 cm/hour, which is more than a hundred times faster than for ^{22}Na in Na, the results could be ob-

Reprint requests to Dr. A. Lodding, Arkitektvägen 6 B, S-43080 Hovås, Schweden.

tained much more accurately. Some of the runs required a correction for convection effects, similar to that mentioned above, but the extra error introduced was less than $\pm 5\%$. The results are listed in Table 2. The "experimental charge number" is defined as

$$Z_{\text{exp}}^* = -\Delta v/v_e \quad (4)$$

(i. e. negative as the tracer moves along with the electrons). The diffusion coefficients of Au in Na have been measured recently⁶.

The magnitudes as well as the temperature dependence of Z_{exp}^* are very similar to the behaviour³ of Na in K, where the experimental charge number ranges from about -12 at the m.p. to -5 at 310°C . A discussion along similar lines as in Ref.³ leads to the conclusion that the effective excess resistivity ρ_{eff}^* for Au in Na is close to $1.2 \mu\Omega \text{ cm}/\%$ impurity

Table 2. Experimental Charge Numbers of Electrotransport, ^{195}Au in Liquid Na.

Temperature, $^\circ\text{C}$	$-Z_{\text{exp}}^*$	Temperature, $^\circ\text{C}$	$-Z_{\text{exp}}^*$
106	11.8 ± 0.6	192	9.3 ± 0.4
119	10.7 ± 0.9	210	9.6 ± 0.7
129	10.4 ± 0.8	236	8.2 ± 0.6
142	11.0 ± 0.8	253	8.2 ± 0.3
155	10.2 ± 0.6	262	9.5 ± 0.6
172	10.7 ± 0.9	272	7.4 ± 0.4
185	9.4 ± 0.7		

(cf. 1.3 for Na in K), with practically no temperature dependence.

This work was supported by the Swedish Board of Technical Development. We thank Fil. lic. S. J. Larsson and Fil. mag. S. E. Eriksson for stimulating discussions.

¹ A. Lodding, Gothenburg Stud. Phys. **1** [1961].

² A. Lodding, J. Phys. Chem. Solids **28**, 557 [1967].

³ C. Roxbergh, T. Persson, and A. Lodding, Phys. Chem. Liq. **4**, 1 [1973].

⁴ S. J. Larsson, C. Roxbergh, and A. Lodding, Phys. Chem. Liq. **3**, 137 [1972].

⁵ T. Persson and S. J. Larsson, Z. Naturforsch. **28a**, 117 [1973].

⁶ T. Persson, S. J. Larsson, and A. Lodding, to be published in Phys. and Chem. Liquids.