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Negative-muon capture ratios for alloys and alkali halide solid solutions

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Negative-muon capture ratios have been determined at three different concentrations for each of the following solid solutions: Cu-Al, Ag-Zn, NaCl-NaBr, KCl-KBr. Over a concentration ratio range of approximately 50 for the alloy systems and 400 for the alkali halide solutions, the data show that the per-atom capture ratios are essentially independent of atomic concentration, in agreement with some recent theoretical models.

I. INTRODUCTION

The Coulomb capture of negative muons in mixtures of elements or compounds dispersed at the atomic or molecular level, that is, in components of a *single-phase solution*, has been of recent experimental and theoretical interest. In the earliest theoretical investigation of this problem, Fermi and Teller[1] proposed the well-known "Z rule," intended to be valid for metallic alloy systems. Under the model assumption that muon capture takes place at essentially zero muon energy by interaction with a degenerate Fermi-Thomas electron distribution, the Z rule predicts that muons are captured in the individual elements in proportion to their atomic numbers (Z's) and concentrations. Thus the per-atom capture ratio

$$A(Z_X/Z_Y) \equiv (X_{\rm capt}/X_{\rm atom})/(Y_{\rm capt}/Y_{\rm atom})$$

should be independent of composition.

If, in contrast to the Fermi-Teller picture, one assumes that muon capture can take place over a range of positive energies, that is, up to $\simeq 100$ eV, one may construct situations where composition invariance of the per-atom capture ratio will not obtain. Then the reduced ratio $A(Z_X/Z_Y)$ may be expressed in terms of integrals for each element evaluated over the capture energy range. Each integral involves an elemental capture cross section $\sigma_i^c(E)$ and the effective muon flux energy distribution (slowing-down spectrum) $P(E)=d^3N_\mu/dE\,dt\,dA$ appropriate to a particular single-phase solution. Here E is the energy, t is the time, and A is the area. Thus

$$A(Z_X/Z_Y)=\int_0^{E_{\rm max}} P(E)\sigma_X^c(E)\,dE \bigg/ \int_0^{E_{\rm max}} P(E)\sigma_Y^c(E)\,dE .$$

We consider, for example, a case where the muons lose only small amounts of energy in individual atomic encounters. If the product $c_X\sigma_X^c$ of concentration and capture cross section for species X is essentially zero above some muon energy E_X, while $c_Y\sigma_Y^c$ remains significantly large above E_X up to E_Y, then most of the muons would be captured into species Y in the energy range $E_X<E<E_Y$. Thus in the slowing-down process the muons could be extinguished by Y before reaching an energy where they could be captured by X, or in the previous expression $P(E)$ would be severely reduced below E_X. These circumstances then lead to composition dependence for $A(Z_X/Z_Y)$.

Semiclassical models of muon energy loss and capture in individual atoms have been described in several recent theoretical papers.[2-6] Vogel *et al.*,[2] calculating negative-muon capture ratios for mixtures of monatomic gases Ne-Ar, Ne-Xe, and Ar-Xe, concluded that the shape of the effective muon energy distribution $P(E)$ in the capture energy range was essentially independent of composition for each binary mixture. Accordingly, they predicted a constant per-atom capture ratio for these model solutions.

In another analysis of the Coulomb capture by atoms in the condensed state, Daniel[3] concluded that a muon encountering an atom of a given atomic number loses a specific, constant amount of energy independent of muon energy. With this result, $P(E)$ for muons slowing in a pure or mixed sample of atoms is independent of energy from zero muon energy to beyond the individual capture thresholds of the constituent atoms.[7] Thus Daniel's model results in a "white" muon flux energy spectrum in the capture range, with a shape that is unaffected by relative concentration of the constituents.

Recently, Leon[8] has generalized Daniel's results by showing that if the shape of the muon energy loss distribution in an atomic encounter is independent of muon energy, and if all muons after capture can have only negative total energy, then the *arrival* rate $R(E)=P(E)\sigma_{\text{inel}}(E)$ for muons slowing in a mixture is independent of energy for any composition. By considering the centrifugal barrier, he relaxed somewhat the condition that the muon energy be negative after capture. By also incorporating the energy dependence of the atomic capture cross section expected from his earlier investigations, Leon concluded that the reduced capture ratio still should remain essentially independent of composition over the muon capture energy range.

The first experiments to examine the concentration dependence of the per-atom muon capture ratios in solution were reported by Knight *et al.*[9] Using alkali halide salts dissolved in water, they investigated reduced atomic capture ratios with respect to oxygen, $A(X^{\pm}/O)$, particularly for the dissociated aqueous ions of potassium and chlorine. Since water was always the major component in these solutions, this molecule dominated all of the muon interactions before capture, that is, the water determined the slowing-down spectrum. Thus the capture ratios relative to oxygen determined in these experiments do not test the effects of a major change in the moderating medium.

A wider range of concentrations was spanned in one set of recent experiments reported by Bergmann *et al.*[10] investigating muon capture in binary metallic alloys. From first results on the Nb-V solid solution system, reduced atomic capture ratios $A(\text{Nb}/\text{V})$ were reported as 1.76 ± 0.24, 1.07 ± 0.05, and 0.98 ± 0.17 for alloys containing 3.3, 50, and 95.4 atomic percent niobium, respectively. The authors state, however, that they do not consider these results as establishing a concentration dependence of $A(Z_X/Z_Y)$.

In the present work, we chose to investigate four sets of solid solutions, two involving a relatively electronegative metal alloyed with a relatively electropositive metal, and two involving alkali halide salts.

II. PREPARATION OF SOLID SOLUTIONS

The phase diagram for the Al-Cu system[11] shows an α phase isomorphic with pure copper extending between 0 and 9.5 weight percent aluminum and a narrowly defined θ phase at 46.5 weight percent, corresponding closely to the atomic ratio Al/Cu $=2/1$. The θ phase crystallizes directly from the melt. Accordingly, Cu-Al alloys of composition 33, 85, and 95 atomic percent copper were chosen for our experiments.

The phase diagram for the Ag-Zn system[11] shows an α phase isomorphic with pure silver extending between 0 and about 20 weight percent zinc and an ϵ phase extending between 56 and 81 weight percent zinc. Ag-Zn alloys of 25, 85, and 96 atomic percent silver were selected.

The metallic alloys were commerically[12] prepared by local arc melting of mixed powders of the two components. In order to establish the structure of the alloys, Debye-Scherrer powder x-ray diffraction patterns were taken of each alloy sample and checked against the tabulated structural patterns.[13] The targets were self-supporting slabs of dimensions $10\times10\times0.6$ cm. These, together with similarly sized targets of the pure metallic elements, were used to obtain the muonic x-ray spectra. At the conclusion of the experiments,

TABLE I. Muonic capture ratios in alloy samples.[a,b]

Capture ratio	Atomic concentration ratio	Reduced capture ratio
$R(\text{Cu}/\text{Al})$	$C(\text{Cu}/\text{Al})$	$A(\text{Cu}/\text{Al})$
1.75 (0.03)	0.492 (0.004)	3.56 (0.06)
20.4 (0.5)	5.85 (0.11)	3.49 (0.11)
53.2 (3.5)	21.1 (2.2)	2.52 (0.31)
$R(\text{Ag}/\text{Zn})$	$C(\text{Ag}/\text{Zn})$	$A(\text{Ag}/\text{Zn})$
0.316 (0.009)	0.327 (0.013)	0.97 (0.05)
6.23 (0.20)	5.77 (0.93)	1.08 (0.17)
20.9 (1.8)	21.5 (1.6)	0.97 (0.11)
	Predicted capture ratios	
	$A(\text{Cu}/\text{Al})$	$A(\text{Ag}/\text{Zn})$
Fermi-Teller (Ref. 1)	2.23	1.57
Daniel (Ref. 3)	1.83	1.35
Vogel *et al.* (Ref. 6)	2.17	1.49

[a] Numbers in parentheses are the estimated 1σ uncertainties.

[b] Does not include errors in relative x-ray counting efficiencies.

three samples were drilled from each of the six alloy slabs and analyzed chemically for the two elements present. The results are included in Table I.

The alkali halide pairs NaCl-NaBr and KCl-KBr are each known to form continuous single-phase solid solutions.[14] The targets were prepared by mixing the required weights of reagent-grade powdered salts to produce 5, 50, and 95 mole-percent mixtures. The mixed salts were melted in Vycor beakers for five minutes and then powdered after cooling. The targets were next sintered for two days at 150 °C below the corresponding melting points and finally powered to form the solid solutions used in our experiments. Debye-Scherrer powder x-ray diffraction patterns recorded for these alkali halide solutions showed a single cubic phase with lattice parameters intermediate between those of the two pure salts. For recording muonic x-ray spectra the powdered samples were contained in Lucite holders with inner dimensions 17×17×1 cm and window thickness 0.25 mm.

III. EXPERIMENTAL OBSERVATIONS

The experiments were conducted at the stopped muon channel of the Clinton P. Anderson Meson Physics Facility (LAMPF) by means of 130-MeV/c incident muons. A scintillation-counter telescope and polyethylene degrader system previously described[9] were used to define stopping events in the targets. Since all muonic x rays of interest were at or above 250 keV ($K\alpha$ line of Na) a 70-cm^3 closed-end coaxial Ge(Li) detector of known efficiency was employed to detect the x rays accompanying muon stops. X-ray spectra were accumulated in multichannel analyzers operated in prompt and delayed coincidence with the muon stopping signal; the latter were recorded to evaluate coincidence efficiencies and to correct for nuclear γ rays. The intensities of the muonic x rays were evaluated by using the photo-peak fitting program SAMPO.[15] The spectra of the ten metallic samples were also evaluated by direct summation and graphical estimates of the background. Self-absorption corrections were calculated using tabulated x-ray cross sections for the individual elements[16] and a Los Alamos computer code accounting for both target-detector geometry and muon spatial stopping distribution in the target. Assuming the total K mesic x-ray yield per capture event is constant for all elements considered here (calculations using the Akylas-Vogel code[17] indicate 0.963 and 0.974 K x rays for aluminum and silver, respectively), the capture ratios were calculated from the sums of the corrected Lyman intensities of the two elements of interest. Experimental errors were assigned to the intensities of the individual transitions by taking into account: (a) peak-area determination including background evaluation; (b) electronic efficiency of the counter-telescope–Ge(Li) coincidence system; (c) photon-detection efficiency of the Ge(Li) detector, including extended source geometry; and (d) photon self-absorption by the sample. The errors in the measured chemical compositions of the alloy samples have been taken into account.

Results for the Cu-Al and Ag-Zn alloys are summarized in Table I. Because of the low intensity of the higher members of the aluminum K series for the most dilute aluminum alloy, a $K_{\text{total}}/K\alpha$ intensity ratio of 1.28 (obtained from observations on pure aluminum metal) was assumed in order to determine the total intensity of the aluminum K series from the measured $K\alpha$ intensity. Similarly, in the case of the most dilute zinc alloy, a $K_{\text{total}}/K\alpha$ intensity ratio of 1.22, obtained from obtained from observations of pure zinc metal, was used.

In a previous report[18] concerning our measurements of the Cu-Al alloy system, a possible increase in the aluminum $K\beta/K\alpha$ intensity ratio was indicated at the lower aluminum concentrations. We now note that this apparent effect results from the close coincidence between the energies of the aluminum $K\beta$ muonic x-ray (412.1 keV) and the copper $L\beta$ muonic transitions $4p_{1/2} \rightarrow 2s_{1/2}$ (412.1

TABLE II. Copper muonic Lyman intensity patterns.[a]

(Cu/Al)	0.492	5.85	21.1	100% Cu	Average alloy[b]
$K\beta/K\alpha$	0.100 (0.003)	0.092 (0.002)	0.088 (0.003)	0.093 (0.003)	0.094 (0.006)
$K\gamma/K\alpha$	0.025 (0.002)	0.027 (0.002)	0.026 (0.002)	0.027 (0.002)	0.026 (0.001)
$K\delta/K\alpha$	0.022 (0.002)	0.016 (0.001)	0.015 (0.001)	0.017 (0.002)	0.018 (0.004)
$K\epsilon/K\alpha$	0.017 (0.002)	0.015 (0.001)	0.015 (0.001)	0.012 (0.002)	0.016 (0.001)
$K\zeta/K\alpha$	0.017 (0.001)	0.016 (0.001)	0.016 (0.001)	0.013 (0.002)	0.016 (0.001)
$K_{>\eta}/K\alpha$	0.113 (0.006)	0.016 (0.003)	0.016 (0.006)	0.097 (0.004)	0.108 (0.004)

[a] Numbers in parentheses are the estimated 1σ uncertainties.

[b] Average alloy indicates average and standard deviation calculated from alloy data alone.

TABLE III. Muonic capture ratios for sodium halide solid solutions.[a,b]

Sample	100% NaCl	95% NaCl 5% NaBr	50% NaCl 50% NaBr	5% NaCl 95% NaBr	100% NaBr
R (Na/Cl)	0.692 (0.012)	0.725 (0.016)	1.48 (0.04)	13.1 (1.3)	
C (Na/Cl)	1.000	1.053	2.000	20.00	
A (Na/Cl)	0.692 (0.012)	0.689 (0.015)	0.741 (0.020)	0.655 (0.066)	
R (Na/Br)		8.22 (0.22)	0.860 (0.040)	0.403 (0.010)	0.372 (0.010)
C (Na/Br)		20.00	2.000	1.053	1.000
A (Na/Br)		0.411 (0.011)	0.430 (0.020)	0.383 (0.009)	0.372 (0.010)
R (Cl/Br)		11.3 (0.27)	0.580 (0.013)	0.0307 (0.0031)	
C (Cl/Br)		19.00	1.000	0.0526	
A (Cl/Br)		0.596 (0.014)	0.580 (0.013)	0.584 (0.058)	

	Predicted capture ratio A(Na/Cl)	 A(Na/Br)	 $\frac{A(\text{Na/Br})}{A(\text{Na/Cl})}$
Fermi-Teller (Ref. 1)	0.65	0.31	$\frac{0.372}{0.692} = 0.538\ (0.017)$[c]
Daniel (Ref. 3)	0.70	0.42	
Vogel *et al.* (Ref. 6)	0.70	0.37	

[a] Numbers in parentheses are the estimated 1σ uncertainties.

[b] Does not include errors in relative x-ray counting efficiencies.

[c] From experimental values listed above.

keV) and $4p_{3/2} \rightarrow 2s_{1/2}$ (413.0 keV). Table I shows the observed capture ratios $R(Z_X/Z_Y)$, the atomic concentration ratio $C(Z_X/Z_Y)$, and the per-atom capture ratios $A(Z_X/Z_Y)$.

If the characteristics of the muon Coulomb capture process, particularly the muon angular momentum distributions during the cascade, are independent of the composition of the solutions, then the intensity patterns of the x-ray series observed for the component elements should also be invariant with concentration. Sufficient members of the copper muonic K series were observed in the Cu-Al alloys to test this invariance. Table II shows that, within experimental error, no variation of the copper K series intensity pattern was observed.

The results for the NaCl-NaBr and KCl-KBr solid solutions are shown in Tables III and IV. In the case of the chlorine muonic K x-ray series

TABLE IV. Muonic capture ratios for potassium halide solid solutions.[a,b]

Sample	100% KCl	95% KCl 5% KBr	50% KCl 50% KBr	5% KCl 95% KBr	100% KBr
R(K/Cl)	1.14 (0.02)	1.21 (0.02)	2.35 (0.04)	27.4 (4.1)	
C(K/Cl)	1.000	1.053	2.000	20.000	
A(K/Cl)	1.14 (0.02)	1.15 (0.02)	1.17 (0.02)	1.37 (0.21)	
R(K/Br)		10.96 (0.24)	1.18 (0.02)	0.559 (0.015)	0.583 (0.008)
C(K/Br)		20.000	2.000	1.053	1.000
A(K/Br)		0.548 (0.012)	0.591 (0.010)	0.531 (0.014)	0.583 (0.008)
R(Cl/Br)		9.08 (0.19)	0.503 (0.008)	0.0204 (0.0031)	
C(Cl/Br)		19.000	1.000	0.0526	
A(Cl/Br)		0.478 (0.010)	0.503 (0.008)	0.388 (0.058)	

	Predicted capture ratios A(K/Cl)	 A(K/Br)	 $\frac{A(\text{K/Br})}{A(\text{K/Cl})}$
Fermi-Teller (Ref. 1)	1.12	0.54	$\frac{0.583}{1.14} = 0.511\ (0.011)$[c]
Daniel (Ref. 3)	1.09	0.65	
Vogel *et al.* (Ref. 6)		0.70	

[a] Numbers in parentheses are the estimated 1σ uncertainties.

[b] Does not include errors in relative x-ray counting efficiencies.

[c] From experimental values listed above.

TABLE V. Potassium muonic Lyman intensity patterns.[a]

Composition	100% KCl	95% KCl 5% KBr	50% KCl 50% KBr	5% KCl 95% KBr	100% KBr	Average solution[b]
$K\beta/K\alpha$	0.080 (0.003)	0.081 (0.003)	0.079 (0.002)	0.072 (0.002)	0.081 (0.003)	0.077 (0.004)
$K\gamma/K\alpha$	0.031 (0.002)	0.029 (0.001)	0.027 (0.001)	0.030 (0.001)	0.030 (0.001)	0.029 (0.001)
$K\delta/K\alpha$	0.028 (0.001)	0.029 (0.001)	0.027 (0.001)	0.023 (0.001)	0.029 (0.002)	0.027 (0.003)
$K\epsilon/K\alpha$	0.021 (0.001)	0.027 (0.002)	0.024 (0.001)	0.026 (0.001)	0.027 (0.002)	0.025 (0.001)
$K\zeta/K\alpha$	0.016 (0.001)	0.017 (0.001)	0.015 (0.001)	0.016 (0.001)	0.019 (0.002)	0.016 (0.001)
$K\eta/K\alpha$	0.0076 (0.001)	0.0069 (0.001)		0.0058 (0.001)		
$K\theta/K\alpha$		0.0098 (0.001)		0.011 (0.001)		

[a] Numbers in parentheses are the estimated 1σ uncertainties.

[b] Average solution indicates average and standard deviation calculated from solution data alone.

an interference between the $K\beta$ member at 692.5 keV and a 695-keV nuclear γ transition (^{78}Se $2+' \rightarrow 2+$) following muon capture in bromine, $^{79}\mathrm{Br}(\mu^-, n)^{78}\mathrm{Se}$, occurred for all the solutions. Accordingly, we adopted the chlorine constant intensity ratio $K_{\mathrm{total}}/K\alpha = 1.18$ determined from pure sodium chloride and pure potassium chloride. Similarly, another interference between the sodium $K\gamma$ transition (313.7 keV) and the bromine $M\delta$ line (313.5 keV) required us to adopt a sodium $K_{\mathrm{total}}/K\alpha$ intensity ratio of 1.24 (as determined from pure sodium chloride) for all solutions containing sodium and bromine.

In the case of potassium, sufficient members of the muonic K x-ray series were observed for both pure compounds, potassium chloride and potassium bromide, as well as their solid solutions, so that it was possible to search for variations of the intensity pattern with concentration. Table V shows that, within experimental errors, no such variation was observed.

Tables II and V compare the copper and potassium chloride or bromide with those from the solid solutions. The close similarity that can be noted from these tables makes reasonable the adoption of standard $K_{\mathrm{total}}/K\alpha$ intensity ratios derived from pure components in those cases where interferences occur.

IV. DISCUSSION

In order to set limits for a possible concentration dependence of the per-atom muon capture ratios $A(X/Y)$ measured in these experiments, we have assumed a dependence on the atom ratio $C(X/Y)$ of the form

$$A(X/Y) = A_0(X/Y) + m \ln C(X/Y) .$$

This form is convenient for examination of $A(X/Y)$ over a wide variation in the concentration ratio. If, for example, the value of m were 0.01 or 0.1, then for a tenfold change in atom ratio C the corresponding absolute change in A would be 0.023 or 0.23, respectively. For all the experimental capture ratios A included in Tables I, III, and IV, linear regressions were carried out to yield values of A_0 and m, together with the respective errors in the quantities. Weightings appropriate to the uncertainties in the experimental values of A were included. The value of $A_0(X/Y)$ so obtained corresponds to the capture ratio expected in a solution, where the relative atomic concentrations of X and Y are equal. Results of these linear regressions are shown in Table VI.

It is apparent that the largest value of m is indicated for the Cu-Al system [$A_0(\mathrm{Cu/Al}) = 3.508 \pm 0.053, m = -0.092 \pm 0.045$]. For this system, however, if the most dilute aluminum alloy (with the largest experimental uncertainties) is excluded from the analysis, the corresponding values of A_0 and m are 3.537 ± 0.053 and -0.032 ± 0.053. Reviewing this case and the other entries of Table VI, we conclude that for the metallic and ionic solutions investigated no significant evidence was obtained for a concentration dependence of the per-atom muon capture ratios. This feature of muon capture is of essential importance for the applications of muonic x-ray spectroscopy to the nondestructive analysis of chemical and biological

TABLE VI. Linear regression results.[a]

System (X/Y)	$A_0(X/Y)$	m
Cu/Al	3.508 ± 0.053	-0.092 ± 0.045
Ag/Zn	0.978 ± 0.044	0.005 ± 0.027
Na/Cl	0.696 ± 0.009	0.015 ± 0.019
Na/Br	0.381 ± 0.006	0.011 ± 0.004
Cl/Br	0.582 ± 0.012	0.005 ± 0.006
K/Cl	1.142 ± 0.014	0.052 ± 0.032
K/Br	0.578 ± 0.006	-0.008 ± 0.005
Cl/Br	0.499 ± 0.007	-0.006 ± 0.004

[a] Form $A(X/Y) = A_0(X/Y) + m \ln C(X/Y)$ assumed; see Sec. IV.

TABLE VII. Muonic capture ratios for alkali halides.

Salt	$A(M^+/X^-)$ from pure salt	$A_0(M^+/X^-)$ from solid solutions
NaCl	0.692 ± 0.019	0.696 ± 0.017
NaBr	0.372 ± 0.013	0.381 ± 0.011
KCl	1.14 ± 0.02	1.14 ± 0.01
KBr	0.583 ± 0.014	0.577 ± 0.023

systems.

From the data reported here one may deduce muon capture ratios $A(M/X)$ for the four pure salts NaCl, KCl, NaBr, and KBr. Here, although the γ-ray efficiency has been included in deriving these entries, the error arising from uncertainties in the relative detector efficiencies for the K x rays of the two elements must be included [$\sigma(\mathrm{Na/Cl}) = 2.0\%$, $\sigma(\mathrm{Na/Br}) = 2.5\%$, $\sigma(\mathrm{K/Cl}) = 0.3\%$, $\sigma(\mathrm{K/Br}) = 2.0\%$]. Taking these estimated additional errors in quadrature we obtain the capture ratios shown in Table VII.

Finally, it is of interest to note a comparison of derived capture ratios $A(\mathrm{Cl/Br})$ obtained from the pure sodium and potassium salts together with the corresponding experimental values for this ratio derived from the solid solutions. If the effective shape of the muon flux energy spectrum at capture, $P(E)$, were constant for all solid ionic systems, one would expect these ratios would be constant.[19] As shown in Tables III and IV one derives values of $A^*(\mathrm{Cl/Br}) = 0.538 \pm 0.017$ and 0.511 ± 0.011, respectively, from the experimental capture ratios determined for the pure sodium and potassium salts. This is to be compared with the values for this ratio $A_0(\mathrm{Cl/Br}) = 0.582 \pm 0.011$ and 0.499 ± 0.007 derived from the linear regression analysis shown in Table VII. We interpret these results as supporting the idea of shape constancy of $P(E)$ in ionic salts.

Note added in proof. In a recent article appearing in this Journal [Phys. Rev. A 20, 633 (1979)] Bergmann *et al.* report more extensive investigations of muon capture in the niobium-vanadium alloy system. These authors find no evidence for concentration dependence of the per-atom muon capture ratio $A(\mathrm{Nb/V})$ over a 40-fold change in atom ratio.

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