Chemical-bond dependence of Coulomb capture of pions in beryllium borides

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Atomic Coulomb capture ratios for pions have been measured for beryllium borides and mixtures of the constituents. The ratios for the mixtures are independent of the atomic fraction and are reproduced well by a quantum-mechanical calculation. However, the ratios for the compounds are less than half of those of the mixtures and decrease rapidly with increase in the atomic fraction of boron. These results for the borides are not predicted by any model, except when the ionicity parameter in the mesomolecular model is used as an adjustable parameter.

I. INTRODUCTION

The process of slowing down and Coulomb capture of negatively-charged pions or muons (we call them mesons for convenience in this text) in matter has been a subject of continuum interest for many experimental and theoretical groups since the pioneering work of Fermi and Teller.[1] Currently, the studies have been focused onto the molecular- or atomic-structure dependence of the atomic (per-atom) Coulomb capture ratio (ACR) of mesons in binary and more complex compounds and the intensity pattern of x rays produced following the cascade process.[2-12] So far, several semiclassical models[8,9,12] and a quantum-mechanical calculation[11] have been applied to the theoretical description of the capture process. There remains, however, the question of which atomic or molecular electrons play the important role in the observed chemical effects. That is, the experimental results for the capture ratios were reproduced to a certain extent by both the mesomolecular model of Ref. 9 and a quantum-mechanical calculation of Ref. 11. In the former model, Schneuwly, Pokrovsky, and Ponomarev assumed that valence electrons played a major role in the capture process, while in the latter, von Egidy, Jakubassa-Amundsen, and Hartmann assumed that mesons were captured through the interaction with individual electrons, regardless of valence electrons.

In very light elements a large fraction of the electrons act as valence electrons. Therefore, studies for binary compounds formed with these elements could make clear the relation between the distribution of valence electrons and the mechanism of Coulomb capture. However, only a few experiments have been carried out with these compounds;[2,4,6] they are BN, NO, CO, and CO_2. For the compound NO, the capture ratio was compared with that

resulting from the mixture of the constituents, and a large difference was found.[2]

In the present study the atomic Coulomb capture ratios have been measured systematically for four species of beryllium borides ($Be_k B_l$). For these light constituents, the ratios would be expected to depend strongly on the species, if the valence electrons are primarily responsible for Coulomb capture of mesons. A comparison of experimental results for the boride compounds and the mixtures of the constituents should tell how important is the role of the valence electrons in the process of slowing down and Coulomb capture of a meson.

II. EXPERIMENTAL PROCEDURE

Experimental arrangement

The experiments were carried out at a $\pi\mu$ channel of the National Laboratory for High Energy Physics (KEK) with 140 MeV/c negative pions. A momentum bite $\Delta p/p$ of $\pm 6\%$ and a beam duty factor of 20% at a frequency of 0.4 Hz were also applied. The experimental arrangement is shown in Fig. 1. Pions, which were focused onto a target, were degraded with an 8.2-cm thick carbon absorber and collimated with a 1-cm thick iron plate with a hole of 4×4 cm² at its center. Pions stopped in the target were counted with a conventional counter telescope composed of one 20×20 cm², two 4×4 cm², and one 20×20 cm² (veto) thin plastic scintillators. Targets used were beryllium boride compounds of Be_4B, Be_2B, BeB_2, and BeB_6 and mixtures of the same constituents in atomic fractions as in the compounds. The beryllium borides used were purchased from Cerac Inc.[13] and were typically 98% pure. Mixtures were prepared by mixing uniformly beryllium powder (100 μm diam particles) with metallic boron
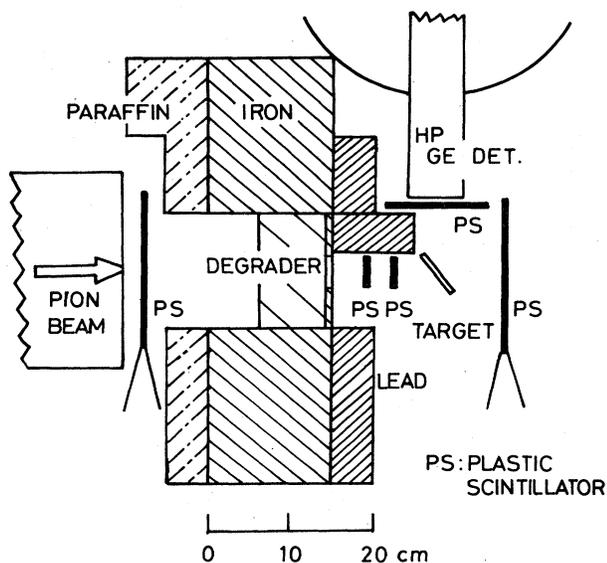
FIG. 1. Schematic drawing of experimental setup.

powder (100 μm diam particles). Other targets were also prepared with 0.8—1.2 μm diam particles of amorphous boron powder for checking the effect of particle size on the atomic Coulomb capture ratio. Powder samples were contained in rectangular-shaped target holders composed of styroform frames with an interior cross section of 4 cm $\times$ 8 cm and with top and bottom windows of 6-μm thick Mylar film. All targets were 0.65 g/cm^2 in areal thickness and were positioned 10 cm behind the collimator at an angle of 45° to the beam direction.

The detector used was an Ortec intrinsic pure germanium detector for low-energy photon spectrometry (LEPS) 2.5 cm in diameter and 0.5-cm thick. The energy resolution of the detector was 0.4 keV for 5.9-keV photons. The detector was set 8 cm from the target and was shielded by paraffin, iron, and lead collimators to reduce the background from photons, neutrons, and scattered charged particles.

The pionic x rays, detected with the LEPS operated in coincidence with stopped-pion events, were analyzed with an Ortec 800 analog-to-digital converter (ADC) and recorded onto a floppy disk for subsequent spectrum analysis. The detection-efficiency curve was obtained with a mixed standard of ^{60}Co, ^{137}Cs, and ^{241}Am activities of known intensities and also with ^{152}Eu of known activity. Each of the two sources was mixed homogeneously in boron powder and put in the styroform container. Because the standards were as thick as the targets, the self-absorption of low-energy photons was compensated for automatically. The efficiency of the fast coincidence circuit was determined to be 0.69$\pm$0.02 by comparing the coincidence events with the singles events for the pionic 2→1 x rays from pure beryllium and boron samples.

In a typical run, 6.7$\times$10^4 pions and 9.1$\times$10^3 muons per accelerator beam pulse entered the target, and 8.1$\times$10^3 pions and 2.4$\times$10^2 muons stopped it in every pulse. The total counting rate in the analyzer was about 200 counts/pulse. Each spectrum was accumulated for

approximately 2 h. Delayed coincidence spectra and singles spectra were taken to identify random coincidence events.

III. RESULTS

A. Pionic x-ray spectra

The pionic x-ray spectra were observed for the Be$_4$B, Be$_2$B, BeB$_2$, and BeB$_6$ compounds and for the mixtures of atomic fractions [Be/B] of $\frac{4}{1}$, $\frac{2}{1}$, $\frac{1}{2}$, and $\frac{1}{6}$. Spectra were also taken for the separate elements of beryllium and boron. Typical spectra are shown in Fig. 2 for the BeB$_2$ compound and the corresponding mixture. The pionic x-ray peaks of the 2→1 transitions of Be and B are observed at energies of 42.4 and 65.0 keV, respectively. The other peaks were in the background and were attributed to carbon and oxygen from surface oxide and water in the target, the target container, the plastic scintillators with their light-tight vinyl covering, etc. Muonic x-ray peaks are also present for Be and B and overlap the respective main pionic 2→1 peaks. Therefore, special attention was taken to evaluate the areas of the 2→1 peaks, especially referring to the data of the shape of x-ray peaks.[14] Data for a blank sample holder, carbon plates, and water were also used for the critical evaluation of background subtraction. For all the observed spectra, Be peaks are less populated in the compounds than in the mixtures and vice versa for B.

B. Pionic x-ray yields per pion stop

Pionic x-ray yields per pion stop were obtained for the 2→1 and 3→1 peaks in the spectra of the separate ele-
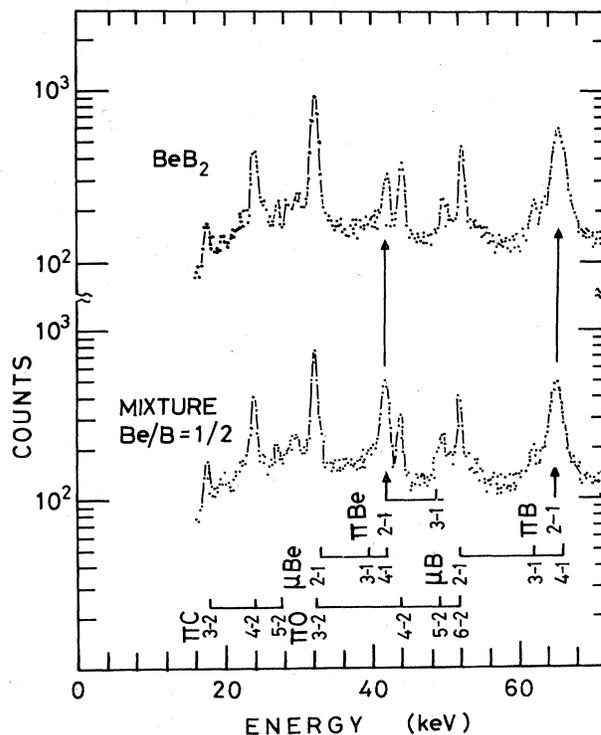
FIG. 2. Pionic x-ray spectra of a boride (BeB$_2$) and the corresponding mixture target (Be + 2B).

TABLE I. Intensities of pionic x rays per stopped pion [$I(x)$].

	Transition	This work	Other work
Beryllium	2→1	15.0±0.9	15.0±1.6[a]
			10.5±1.4[b]
	3→1	2.7±0.3	
Boron	2→1	8.6±0.5	10.9±1.4[a]
			12.5±1.0[b]
	3→1	1.2±0.2	

<table>
<tr><td colspan="4" style="text-align:center">$I(x)$ (%)</td></tr>
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[a]Reference 14.
[b]Reference 15.

ments of beryllium and boron. Here, the peak areas in the spectra were corrected for detection efficiencies and the fast-coincidence efficiency. The number of stopped pions were obtained by correcting the total of stopped events for the contribution of the fraction (3%) of muons stopped in the targets. The results are shown in Table I along with previous data.[15,16] Our data agree with the results of Ref. 15 within the quoted errors.

C. Atomic Coulomb capture ratio

By applying the corrections of detection efficiency and the x-ray yield per pion stop to the observed intensities of the relevant peaks, we have obtained atomic Coulomb capture ratios, A(Be,B), between Be and B. The results are shown in Table II. Each result is generally the mean value of three runs, where agreement is within their statistical errors. The sample of BeB$_6$ contains a large error. This was caused by the spectrum analysis of a small peak of the Be 2→1 x ray disturbed by the pionic 4→2 line in oxygen. For the mixture samples the particle-size effect on ACR was checked carefully with two different particle sizes of boron powder (0.8–1.2 μm diam and 100 μm diam particles), but no effect was found outside of the statistical uncertainty. This is because the stopping power of both elements are very similar and no difference is expected according to the equations of Daniel.[17]

In Fig. 3, we have plotted ACR against the atomic fractions F_B of the number of boron atoms to the total number of beryllium and boron atoms for beryllium borides and mixtures. The value of ACR for the mixture did not vary when we changed the atomic fraction. However, for
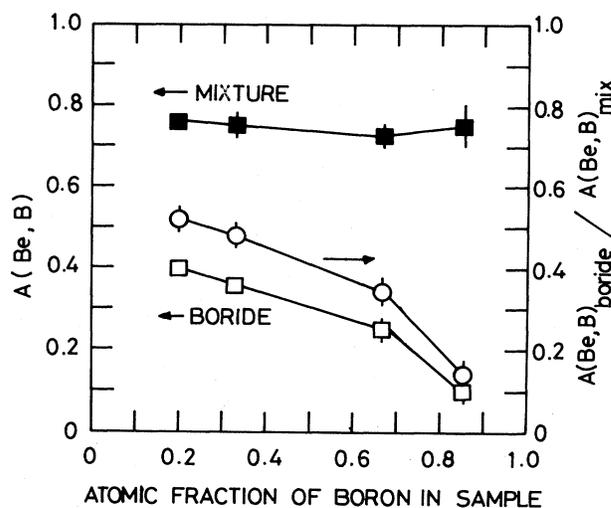
FIG. 3. Atomic (per-atom) Coulomb capture ratios for borides and mixtures, denoted with open and closed squares, respectively. The ratios of these ratios of borides to mixtures as a function of the atomic fraction of boron (open circles).

the compounds, ACR was significantly lower, at most 52% for Be$_4$B, and decreased drastically with increase in the fraction of boron in the sample, as shown in the bottom line of Table II and Fig. 3.

IV. DISCUSSION

A. Atomic Coulomb capture ratios

As shown in Table II and Fig. 3, when elements form a molecular compound, the atomic Coulomb capture ratio is substantially lower than the value for the mixtures. This tendency has already been observed by Knight *et al.* for a NO gas sample.[2] The second finding for the borides, which may be more important, is that atomic Coulomb capture ratio depends strongly on the atomic fraction of the constituent elements or on the structure of the molecules. In the following discussion, we first compare the experimental data for the mixtures with the theoretical predictions, and then we proceed to the compound molecules.

As shown in Fig. 3, ACR in the mixtures is independent of the atomic fraction of the constituents beryllium and boron. This independence was already confirmed for other condensed mixtures.[18,19] This is explained by the

TABLE II. Experimental atomic (per-atom) Coulomb capture ratios, A(Be,B), for compounds, mixtures, and compounds/mixtures.

Mixture	4Be + B	2Be + B	Be + 2B	Be + 6B
A_m(Be,B)	0.76±0.02	0.75±0.03	0.73±0.02	0.75±0.05
Compound	Be$_4$B	Be$_2$B	BeB$_2$	BeB$_6$
A_c(Be,B)	0.40±0.02	0.36±0.02	0.25±0.02	0.10±0.03
$\dfrac{A_c\text{Be,B})}{A_m\text{(Be,B)}}$	0.52±0.03	0.48±0.03	0.34±0.03	0.14±0.04

TABLE III. Predictions of atomic (per-atom) Coulomb capture ratios.

	Schneuwly et al.[a]	Daniel[b]	von Egidy et al.[c]
A(Be,B)	0.545	0.73	0.754

[a]Reference 9.
[b]Reference 8.
[c]Reference 11.

reason that the shape of the spectral flux density $n(W)$ of the mesons in the targets at energy W is not expected to depend strongly on the target material, because of the small size of the atoms, and hence, the steepness of the electronic potential.[20]

From Tables II and III, one can compare the experimental data for the mixtures of beryllium and boron with the theoretical predictions. Among the number of predictions proposed since the discovery of the periodicity dependence by Zinov et al.,[21] only the semiempirical models by Daniel[8] and by Schneuwly, Pokrovsky, and Ponomarev[9] and a quantum-mechanical description by von Egidy, Jakubassa-Amundsen, and Hartmann[11] accounted for the periodicity and were extensively compared with experiments. Therefore, only the calculated results based on these three models are presented in Table III. In the model by von Egidy et al. it is assumed that the atoms even in molecules are neutral, and mesons are captured through the interaction with individual electrons, and that the capture probabilities are determined by a general function involving the electron binding energy, the electron quantum numbers, and the atomic number. This model is successful when applied to heavy elements but fails for compounds composed of the very light elements Li, Be, and B. For compounds of these elements the chemical bond plays a decisive role and the distribution of electrons must be taken into account more carefully. Therefore, in the very light elements, the results of Ref. 11 may be applied only for mixtures. In fact, when comparing Tables II and III, the value cited from Ref. 11 reproduces very well the results of the mixture. On the other hand, as the model by Schneuwly et al., which originated from the mesomolecular model successfully applied for molecules containing hydrogen, makes the valence electrons play a major role, it is not appropriate for the mixtures.

To study the effect of valence electrons on the atomic capture mechanism, the ACR has been measured for

many oxides with different valences,[5] but the observed differences were very small. This is not surprising because most of the previous experiments concentrated on heavy elements, and the role of the valence electrons is very small in these cases as known from the applicability of the model by von Egidy et al. But for the very light elements, the effect of the valence electrons is expected to be large. In fact, there are a few observations which support the importance of the valence electrons in the Coulomb capture process. That is, Knight et al. obtained different values of ACR between the molecular NO target and the simple $N_2 + O_2$ mixture.[2] Also, the reported values[2,4] for CO and CO_2 differ by a factor of 2 from each other. For BN, even the allotropic effect was observed.[6] Figure 3 shows the obvious effect of the valence electrons. The value of ACR decreases very rapidly with an increase in the boron fraction. The value ranges from 0.40 for Be_4B to 0.10 for BeB_6, which corresponds to 52% and 14% of the value of the mixture, respectively. This downward shift and the chemical-bond dependence of ACR in the borides are clear indications of the influence of the polarity of the bonds. Schneuwly et al. predicted a value of 0.545 for the ACR, independent of the species of the beryllium borides, when the ionicity σ of the $Z_1 - Z_2$ bond was estimated from Pauling's electron negativities.[22] The predicted value is much larger than the experimental results.

However, in previous work with the formula of Schneuwly et al.[9] the ionicity σ of the chemical bond was originally taken as a parameter independent of the structure of matter or the species of molecule but was later used as an adjustable parameter in order to achieve better agreement between the model predictions and the experimental values. Although the borides have rather complex crystal structures and, therefore, it would not seem appropriate to apply this model to the borides, we have used the same procedure in order to find a clue to the

TABLE IV. Values of the ionicity parameter σ_m in the model by Schneuwly et al. required to achieve the best agreement between predicted and experimental atomic (per-atom) Coulomb capture ratios, $A(Z_1,Z_2)$. Z_2 denotes the element with greater electron negativity.

	Be_4B	Be_2B	BeB_2	BeB_6	BN (cub)	BN (hex)	CO	CO_2	NO
$A(Z_1,Z_2)$	0.40 ±0.02	0.36 ±0.02	0.25 ±0.02	0.10 ±0.03	0.233[a] ±0.011	0.275[a] ±0.012	0.766[b] ±0.030	0.46[c] ±0.07	0.959[b] ±0.030
σ_m	−0.43 ±0.02	−0.06 ±0.03	0.67 ±0.03	0.95 ±0.03	0.20[a] ±0.02	0.10[a] ±0.02	−0.06 ±0.03	0.0 ±0.1	−0.10 ±0.02

[a]Reference 6.
[b]Reference 2.
[c]Reference 4.

chemical-bond effect observed in this experiment. The parameters we used, other than the ionicity parameter σ, are the values reported in Ref. 9. The resulting values of the ionicity parameter σ, which reproduce the experimental capture ratios, are presented in Table IV, along with the results from previous experiments.[2,4,6]

Knight *et al.* pointed out that for the cases of CO and NO molecules, bond ionicities are negative.[2] But for the case of the CO_2 molecule,[4] the bond ionicity was zero. Redistribution of the valence electrons was needed in order to explain the difference in ACR between the hexagonal and cubic boron nitrides.[6] In this experiment, it was found that the parameter σ for the borides increases rapidly from a negative to a positive value with an increase in the atomic fraction of boron, as shown in Table IV. This implies that in boron-rich borides, electrons transfer from the beryllium to the boron atom. This electron transfer agrees with the general understanding that in metal borides MB_2 and MB_6, the initial transfer of electrons from metal M to make the respective B_2^{2-} layer and B_6^{2-} cage, fill the bonding molecular orbitals, and this is no doubt accompanied by a stabilizing transfer of part of the electrons back from the boron layers and cages to the metal.[23]

Thus, some idea of the ionicity is deduced from the experimental ACR combined with the model by Schneuwly *et al.* However, further studies, both theoretical and experimental are needed on compounds composed of light elements in order to understand the relationship between the distribution of the valence electrons and the ACR value.

V. SUMMARY

Our principal results and conclusion from this investigation may be summarized as follows.

(1) The values of ACR for the mixtures are independent of the atomic fractions of the constituents beryllium and boron. The experimental results are reproduced by the model by von Egidy *et al.*

(2) The values for the boride compounds are less than half of those of the mixtures. This fact shows the influence of the polarity of the bonds.

(3) Furthermore, the ACR value depends strongly on the species of the boride, and it decreases rapidly with increase in the atomic fraction of boron.

(4) In order to achieve better agreement between the predictions with the model by Schneuwly *et al.* and the experimental results, an adjustable ionicity parameter was used. In the framework of the model, electrons transfer from beryllium to boron in the boron-rich borides.

ACKNOWLEDGMENTS

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