

## Analysis of Energy-Loss Data for 0.2–0.5 MeV/amu $p$ , $\alpha$ , and $N$ in Se

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The analysis of experimental energy-loss data for 0.2–0.5 MeV/amu  $p$ ,  $\alpha$ , and  $N$  in Se has been made to obtain certain quantities. Data of standard media are also shown for comparison. The results tell us that the effective-charge parameter for  $N$  is quite different from unity. The variation of the relative mean excitation energy measured by  $\alpha$  particles is not linear in terms of  $Z_2$ . The parameter  $\xi$  for  $N$  is slightly larger than the theoretical value ( $Z_1^{1/6}$ ). The electronic stopping cross section for  $N$ , which lies between two theoretical values, is not exactly proportional to the projectile velocity, suggesting that nitrogen as a projectile shows a peculiar behavior particularly unlike other light-heavy ions.

### I. INTRODUCTION

The mechanism of energy loss of low-energy heavy ions has been the subject of theoretical and experimental studies in the field of atomic stopping since Bohr<sup>1</sup> laid the theoretical foundation for it.

In the region of  $E_A = 0.1$ – $0.5$  MeV/amu, it is considered that the electronic contribution is still primarily responsible for discrepancies in range or energy-loss quantities between theory and experiment. These discrepancies are partly related to an effective charge of the ions in media when theories contain no ionization effects.

For small  $Z_1$  and high  $E_A$  projectiles in isolated atoms, the Bethe-Bloch treatment<sup>2</sup> applies to the electronic stopping region. Thus, it can be reconsidered only for the estimate of ionization.

The relativistic Bethe-Bloch formula for the stopping power is given by

$$\frac{-dE}{dX} = 4\pi e^4 Z_1^2 N_0 Z_2 m^{-1} v^{-2} M_2^{-1} B, \quad (1)$$

where

$$B = \ln(2mv^2 I^{-1}) - \beta^2 - \ln(1 - \beta^2) - C Z_2^{-1} - 0.5\delta.$$

The quantities  $e$  and  $m$  are the charge and mass of the electron,  $v = c\beta$  the velocity of the incident ion, and  $N_0$  is Avogadro's number.  $I$  and  $C/Z_2$  denote mean excitation energy and shell correction of the medium, respectively. The factor  $C/Z_2$  is effective at very low velocities; the quantity  $\delta$  expresses the density correction term which is of importance at very high velocities.<sup>3</sup>

Equation (1) is often applied to evaluate the stopping power of heavy ions, merely by replacing the full nuclear charge of the ion by an effective charge  $Z_{\text{eff}} = \gamma Z_1$ , which depends on the ion velocity. In terms of the effective charge, Eq. (1) may be rewritten in the form

$$\frac{-dE}{dX} = 3.072 \times 10^{-4} Z_{\text{eff}}^2 Z_2 \beta^{-2} M_2^{-1} B, \quad (2)$$

where  $-dE/dX$  and  $M_2$  are expressed in MeV cm<sup>2</sup>

mg<sup>-1</sup> and amu, respectively. In the case of ions penetrating the same medium at the same velocity, one obtains

$$\frac{-dE}{dX} = \text{const} \times Z_{\text{eff}}^2. \quad (3)$$

### II. EXPERIMENT ON $p$ IN Se

A deuteron beam of 200 keV, 100  $\mu\text{A} \times 20\%$  extracted from the Kyoto University Cockcroft-Walton-type accelerator bombarded a deuteron target built in Zr (backed by Cu) to yield a nuclear reaction  $D(d, p)T$ . Use was made of these protons as projectiles to absorber targets. Various kinds of proton energies were selected by varying the scattering angle or degrading the initial energy with Al foils. Signals from a solid-state detector were analyzed by a 256-channel pulse-height analyzer. The gain and zero point of the detector-amplifier-analyzer system were monitored by inserting test pulses from a mercury pulser into the preamplifier. The main sources of error are location of peak in profile, drift in pulse-height analyzer, and uncertainty in foil thickness.

Table I shows the experimental results  $-dE/dX$  (MeV cm<sup>2</sup> mg<sup>-1</sup>) of  $p$  in Al, Se, and Ag. The results for  $p$  in Al and Ag are successfully compared with others.<sup>4–10</sup> The atomic weight averaged over the abundance ratio was used. The Se foils employed in the experiment were transparent and light red.

Our previous experimental data for  $\alpha$  particles and nitrogen ions<sup>11,12</sup> in Al, Ni, Se, Ag, and Au were analyzed extensively.

### III. EFFECTIVE-CHARGE PARAMETER OF $N$ IN Se

The relative effective charge responsible for the electronic part of the energy-loss process is deduced from the comparison of  $-dE/dX$  of different ions in the same medium at the same velocity by using Eq. (1). The capture and loss of electrons and the use of a full nuclear charge in place of the effective charge in Eq. (2) is less valid.

TABLE I. Stopping powers (MeV cm<sup>2</sup> mg<sup>-1</sup>) of *p* in Al, Se, and Ag as a function of proton energy (MeV).

<i>p</i> in	$E_{\text{proton}}$	$-dE/dX$
Al	0.992 ± 0.015	0.171 ± 0.007
	1.381 ± 0.015	0.092 ± 0.020
	0.947 ± 0.015	0.115 ± 0.020
Ag	0.707 ± 0.015	0.138 ± 0.020
	1.366 ± 0.015	0.064 ± 0.015
	1.141 ± 0.015	0.077 ± 0.015

From our *p*,  $\alpha$ , and *N* experiments, the effective-charge parameter of nitrogen ions

$$\gamma_N = \frac{\gamma_p}{7} \left[ \left( \frac{-dE}{dX} \right)_N \left/ \left( \frac{-dE}{dX} \right)_p \right. \right]^{1/2}, \quad (4)$$

$$\gamma_N = \frac{2\gamma_\alpha}{7} \left[ \left( \frac{-dE}{dX} \right)_N \left/ \left( \frac{-dE}{dX} \right)_\alpha \right. \right]^{1/2} \quad (5)$$

was obtained by noting that the factor *B* remains unchanged.

The quantity  $-dE/dX$  for *p* or  $\alpha$  decreases with increasing  $E_A$ , but that for *N* increases up to the maximum and decreases in this  $E_A$  region.

Table II shows the experimental results of the effective-charge parameter  $\gamma_N$  for *N*. It is clear that  $\gamma_N$  increases with energy.  $\gamma_N$  from Eq. (4) is lower than that from Eq. (5) since the parameter  $\gamma_\alpha$  for  $\alpha$  particles is lower than  $\gamma_p$  for protons at the same velocity. In terms of  $Z_2$  dependence, it is almost constant.

#### IV. MEAN EXCITATION ENERGY OF Se

The value  $-dE/dX$  is sensitive to the quantities *I* and  $C/Z_2$  which indicate the characteristics of the electrons of the medium atom; when the effective charge is approximated to be a nuclear charge, experimental errors for *I* become large. For these reasons, most experiments concern high-energy protons, deuterons, or  $\alpha$  particles so that the effective charge may be assumed to be equal to a full nuclear charge.<sup>13</sup>

The factor *B* in Eq. (2) can be approximated as a nonrelativistic form:

$$B \approx \ln(2mv^2/I^*)$$

or

$$I^* \approx 2mv^2 e^{-B}. \quad (6)$$

The quantity  $I^*$  contains the mean excitation energy, shell corrections, etc. Considering  $Z_{\text{eff}}$ , high-energy experiments are more valid than lower-energy ones. However, this nonrelativistic approach is valid in the lower-energy region.

Table III shows the experimental results of  $I^*$  (eV) and  $I^*/Z_2$  for  $\alpha$  particles as a function of  $E_A$ ,

where  $\ln I^* = \ln I + C/Z_2$  or  $I^* = e^{C/Z_2}$  and  $\gamma_\alpha = 1$ , and those for protons where  $\gamma_p = 1$ .

The value  $I^*$  increases with increasing  $E_A$  and  $Z_2$ ; the absolute value cannot be believed to be accurate because of the factor  $e^{C/Z_2}$ . This increase is not linear in terms of  $E_A$  because the factor *B* does not vary monotonically with *v*. At least the factor *B* contains the variation due to  $(-dE/dX)\beta^2$ . In the present velocity region, the quantity  $-dE/dX$  decreases with increasing *v*.

Since the value  $I^*$  refers to a medium atom, it should be the same for the same medium at the same velocity whether the incident particle is a proton or an  $\alpha$  particle. Nevertheless, these results are not the same, partly due to the difference of the effective-charge parameter between protons and  $\alpha$  particles.

The shell corrections are energy dependent, unlike the mean excitation energy. If the ion velocity is much larger than the orbital velocity of electrons, the term  $C/Z_2$  tends to zero. It may not be negligible in heavy elements since the orbital velocity is high. It is reported that the value  $C/Z_2$  extends from 0.1 for light elements to 0.3 for heavy ones. Therefore the term  $e^{C/Z_2}$  will vary from 1.15 to 1.35.

Recently it has been suggested that the quantity  $I/Z_2$  does not increase monotonically with  $Z_2$ , but oscillates probably because of the shell structure of the medium atom.<sup>14</sup> More precise measurements in the intermediate  $Z_2$  region are required for the recognition of this oscillatory behavior of  $I/Z_2$ .

#### V. STOPPING CROSS SECTIONS OF *p*, $\alpha$ , AND *N* IN Se

Accurate measurements of  $-dE/dX$  values for light particles in the region  $E_A = 0.1-0.5$  MeV/amu are important, particularly to discuss the  $Z_2$  oscillation of the electronic stopping cross section  $S_{e1}$ .<sup>8,13</sup>

Chu and Powers<sup>15</sup> have found that  $S_{e1}$  for  $\alpha$  particles increases with  $Z_2$ , except in the region of  $Z_2 = 22-29$ , where a zigzag exists, and where it decreases with increasing  $Z_2$  except at  $Z_2 = 26$ , in contrast with the increase predicted by theories

TABLE II. Effective-charge parameters of *N* in Al, Ni, Se, and Au obtained from Eq. (4) or (5), where  $\gamma_p = \gamma_\alpha = 1$ ; experimental errors are 5% at most.

$E_A$	Al	Ni	Se	Se	Au
	Eq. (5)	Eq. (5)	Eq. (4)	Eq. (5)	Eq. (5)
0.20	0.51	0.59	0.56	0.61	0.57
0.25	0.56	0.62	0.60	0.66	0.61
0.30	0.61	0.65	0.63	0.70	0.65
0.35	0.65	0.68	0.66	0.73	0.68
0.40	0.69	0.70	0.68	0.76	0.71
0.45	0.73	0.71	0.70	0.77	0.73
0.50	0.75	0.73	0.72	0.79	0.75

TABLE III. Mean excitation energies (eV) of Al, Ni, Se, Ag, and Au measured by  $\alpha$  particles.  $I^*$  means the value of Eq. (6), where  $\gamma_\alpha=1$ ; experimental errors are less than 5%. The last column indicates those of Se measured by protons, where  $\gamma_p=1$ .

$E_A$	Al		Ni		Se		Ag		Au		Se (measured by $p$ )	
	$I^*$	$I^*/Z$	$I^*$	$I^*/Z$	$I^*$	$I^*/Z$	$I^*$	$I^*/Z$	$I^*$	$I^*/Z$	$I^*$	$I^*/Z$
0.20	113	8.7	239	8.5	256	7.5	260	5.5	310	3.9	...	...
0.30	120	9.3	271	9.7	316	9.3	331	7.0	406	5.2	...	...
0.40	137	10.5	287	10.2	356	10.5	385	8.2	481	6.1	...	...
0.50	158	12.1	293	10.5	384	11.3	428	9.1	540	6.8	305	9.0
0.60	169	13.0	300	10.7	408	12.0	461	9.8	587	7.4	315	9.3
0.70	184	14.2	304	10.9	423	12.4	486	10.3	624	7.9	323	9.5
0.80	191	14.7	308	11.0	438	12.9	503	10.7	653	8.3	334	9.8
0.90	194	14.9	310	11.1	445	13.1	512	10.9	677	8.6	343	10.1
1.00	197	15.2	315	11.3	452	13.3	515	11.0	688	8.7	356	10.5

which do not include the effect of the periodic variation in radial electron density.<sup>16</sup> They have also found a zigzag for protons in  $Z_2=23-29$  at lower energies, probably due to the existence of a 4s electron.

According to Fig. 1, the electronic stopping cross section  $S_{e1}$  (in units of  $10^{-14}$  eV cm<sup>2</sup>/atom) for  $\alpha$  in Se is as large as that for  $\alpha$  in Ni. Hence it may be said that it does not increase monotonically with  $Z_2$  between  $Z_2=28$  and  $Z_2=34$ .

In order to compare the stopping cross section between theory and experiment for  $N$  in Se,

$$S_F = 5.15 \times 10^{-15} (Z_1 + Z_2) v v_0^{-1} \text{ eV cm}^2/\text{atom} \quad (7)$$

or

$$S_{e1} = \xi 8\pi e^2 a_0 Z_1 Z_2 (Z_1^{2/3} + Z_2^{2/3})^{-3/2} v v_0^{-1},$$

$$\xi = Z_1^{1/6}, \quad v_0 = e^2/\hbar, \quad a_0 = \hbar^2/m_e^2, \quad v_0 Z_1^{2/3} > v \quad (8)$$

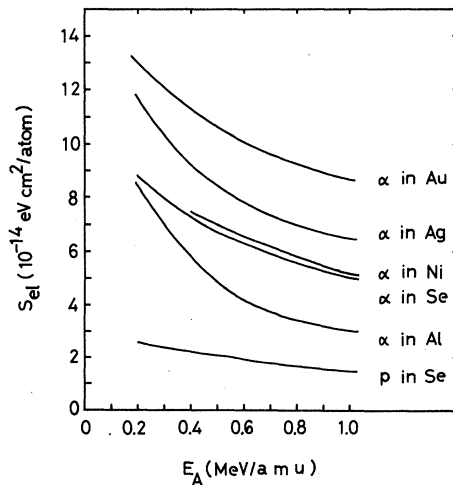


FIG. 1. Smoothed curves of experimental stopping cross sections ( $10^{-14}$  eV cm<sup>2</sup>/atom) for  $p$  or  $\alpha$  in Al, Ni, Se, Ag, and Au; experimental errors are 5% at most, as shown in Ref. 12.

was quoted as the theoretical electronic stopping cross section per atom.<sup>17-19</sup>

The observed stopping cross section per atom  $S_0$  is given by

$$S_0 = \left( \frac{-dE}{dX} \right)_{\text{expt}} N_u^{-1}, \quad (9)$$

where  $N_u$  is the number of stopping atoms per unit volume;  $(-dE/dX)_{\text{expt}}$  was calculated by differentiating the energy-thickness relation obtained from our previous experiment.

The nuclear stopping cross section per atom calculated by

$$S_{nu} = \xi_{nu} \frac{1}{2} \pi^2 e^2 a Z_1 Z_2 M_1 (M_1 + M_2)^{-1},$$

$$\xi_{nu} = 2/(2.718 \times 0.8853),$$

$$a = 0.8853 a_0 (Z_1^{2/3} + Z_2^{2/3})^{-1/2} \quad (10)$$

is about 0.2-0.3 in units of  $10^{-13}$  eV cm<sup>2</sup>/atom for  $N$  in Al, Ni, Se, etc., suggesting that the nuclear contribution is negligible in  $S_0$ .

Table IV shows the results of calculation by Eqs. (7) and (8) compared with experimental results of Eq. (9) in units of  $10^{-13}$  eV cm<sup>2</sup>/atom. It is seen that  $S_{e1} < S_0 < S_F$  in every case.

When  $S_0$  was plotted in log-log paper as a function of energy to examine if it was proportional to  $v$ , the exponent  $p$  of the equation

$$S_0 = cE^p \quad (11)$$

was found to be about 0.40, where  $c$  is a constant. Consequently, it is recognized that  $S_0$  for  $Z_1=7$  is not in proportion to  $v$ , but is entirely in agreement with the result of Hvelplund and Fastrup.<sup>20</sup>

The present analyses indicate that  $(-dE/dX)_{\text{expt}}$  and  $S_0$  deviate from the respective theoretical values systematically. It is said that the relative amplitude of the periodical  $Z_1$  oscillation of  $S_{e1}$  decreases for fixed  $Z_2$  with increasing  $v$  and it is more dominant in the case of low  $Z_1$ . Our results support the fact that nitrogen itself behaves pecu-

TABLE IV. Stopping cross sections  $S_0$ ,  $S_{el}$ , and  $S_F(10^{13} \text{ eV cm}^2/\text{atom})$  for  $N$  in Al, Ni, Se, Ag, and Au; experimental errors are less than 5%. Column  $A$  shows the observed stopping power, while columns  $B$ ,  $C$ , and  $D$  the theoretical ones ( $\text{MeV cm}^2 \text{mg}^{-1}$ ); experimental errors are less than 3%. Column  $E$  shows the  $\xi$  value derived from  $(-dE/dX)_{\text{expt}}$ .

$N$ in	$E_A$	$A$	$B$	$C$	$D$	$E$	$S_0$	$S_{el}$	$S_F$
Al	0.05	3.55	2.73	2.82	3.36	1.79	1.59	1.22	1.46
	0.10	4.55	3.88	3.93	4.52	1.62	2.04	1.74	2.07
	0.15	5.38	4.75	4.79	5.38	1.56	2.41	2.13	2.53
	0.20	6.06	5.49	5.52	6.12	1.53	2.72	2.46	2.93
	0.25	6.61	6.14	6.16	6.77	1.49	2.96	2.75	3.27
	0.30	7.04	6.72	6.74	7.36	1.45	3.15	3.01	3.59
Ni	0.05	2.04	1.63	1.70	1.93	1.73	1.99	1.59	2.55
	0.10	2.63	2.31	2.36	2.61	1.57	2.56	2.26	3.62
	0.15	3.13	2.83	2.86	3.13	1.53	3.05	2.76	4.43
	0.20	3.55	3.27	3.30	3.57	1.50	3.46	3.19	5.12
	0.25	3.90	3.66	3.68	3.96	1.47	3.80	3.57	5.73
	0.30	4.18	4.01	4.03	4.31	1.44	4.07	3.91	6.28
Se	0.05	1.76	1.28	1.33	1.48	1.90	2.30	1.67	2.99
	0.10	2.26	1.81	1.85	2.02	1.73	2.96	2.38	4.24
	0.15	2.68	2.22	2.24	2.42	1.66	3.52	2.91	5.20
	0.20	3.04	2.57	2.58	2.77	1.63	3.99	3.36	6.00
	0.25	3.34	2.87	2.88	3.07	1.61	4.38	3.76	6.71
	0.30	3.58	3.14	3.16	3.34	1.58	4.69	4.12	7.35
Ag	0.05	1.74	1.01	1.06	1.15	2.38	3.11	1.81	3.93
	0.10	2.22	1.43	1.46	1.58	2.13	3.98	2.57	5.59
	0.15	2.63	1.76	1.78	1.90	2.07	4.71	3.15	6.84
	0.20	2.97	2.03	2.05	2.17	2.02	5.32	3.64	7.91
	0.25	3.25	2.27	2.28	2.41	1.98	5.81	4.06	8.84
	0.30	3.46	2.48	2.50	2.63	1.92	6.20	4.45	9.68
Au	0.05	0.89	0.61	0.65	0.68	2.01	2.91	2.00	6.26
	0.10	1.15	0.87	0.89	0.93	1.83	3.77	2.84	8.90
	0.15	1.38	1.06	1.08	1.13	1.79	4.51	3.48	10.9
	0.20	1.58	1.23	1.24	1.29	1.78	5.15	4.02	12.6
	0.25	1.75	1.37	1.38	1.44	1.76	5.71	4.49	14.1
	0.30	1.89	1.50	1.51	1.57	1.73	6.18	4.92	15.4

liarily as a projectile, as expected from the phenomenon that  $Z_1=7$  is positioned just on one of the peaks.

#### VI. $\xi$ FOR $N$ IN Se OBTAINED FROM $(-dE/dX)_{\text{expt}}$

The discrepancy between theoretical and experimental energy-loss data is expected to be due to the difference between theoretical and experimental  $\xi$  values. For this reason, the  $\xi$  value calculated from stopping powers was compared with theory.

A  $^{14}\text{N}$  ion with a velocity  $v_0$  has an energy of 350 keV (i. e.,  $E_A = 0.05$ ), and that of energy 4.63 MeV (i. e.,  $E_A = 0.33$ ) has a velocity of  $v_0 Z_1^{2/3} = 8.0 \times 10^8$  cm/sec. Because of a small contribution of  $(-dE/dX)_{\text{nu}}$  in that energy interval,  $(-dE/dX)_{\text{el}}$  should be obtained with little error by differentiating the energy-thickness relation.

The theoretical electronic stopping, based on a model of a charged particle losing energy continuously in moving through a Fermi gas of charged particles, is<sup>21</sup>

$$\left(\frac{-dE}{dX}\right)_{\text{el}} = \frac{8\pi\xi N_u e^2 a_0 Z_1 Z_2}{(Z_1^{2/3} + Z_2^{2/3})^{3/2}} \frac{v}{v_0}, \quad v < v_0 Z_1^{2/3}. \quad (12)$$

For the estimation of  $(-dE/dX)_{\text{nu}}$  the following two relations were quoted<sup>1,18</sup>:

$$\left(\frac{-dE}{dX}\right)_{\text{nu}} = \frac{4\pi N_u Z_1^2 Z_2^2 e^4}{M_1 v} \ln\left(\frac{a M_1 M_2 v^2}{Z_1 Z_2 (M_1 + M_2) e^2}\right), \quad b/a < 1 \quad (13)$$

$$\left(\frac{-dE}{dX}\right)_{\text{nu}} = \frac{\pi^2 N_u a Z_1 Z_2 e^2 M_1}{2.718(M_1 + M_2)}, \quad b/a > 1 \quad (14)$$

where  $b$  is the classical distance of closest approach in a head-on collision of two unscreened atoms.

Equation (12) refers to the electronic stopping with  $\xi = Z_1^{1/6}$ . Equation (13) shows the nuclear stopping and is very small compared with Eq. (12). Equation (14) also gives the nuclear stopping and is independent of energy; it is 0.634, 0.299, 0.201, 0.143, and 0.0661 in units of  $\text{MeV cm}^2 \text{mg}^{-1}$  for  $N$  in Al, Ni, Se, Ag, and Au, respectively.

In Table IV is shown the comparison of  $-dE/dX$  between theory and experiment. Column *A* shows the experimental  $-dE/dX$  MeV cm<sup>2</sup>mg<sup>-1</sup> obtained from Refs. 11 and 12. Columns *B*, *C*, and *D* indicate the respective theoretical values (MeV cm<sup>2</sup>mg<sup>-1</sup>) obtained from Eqs. (12), (12)+(13), and (12)+(14), respectively. Column *E* gives the  $\xi_{\text{expt}}$  value obtained from the experimental  $-dE/dX$  value through Eq. (12).

According to the results,  $\xi_{\text{expt}}$  depends on  $E_A$  and the kind of a medium; it is always slightly larger than the theoretical value  $Z_1^{1/6} = 1.383$ . This phenomenon is consistent with the general tendency of the  $\xi$ -vs- $Z_1$  relation.<sup>19,22</sup>

As for some media and projectiles, it is known that the theoretical value is lower than the experimental one in the case of  $Z_1 = 5 - 9$ , but exceeds it in the case of  $Z_1 = 11 - 15$ ; the discrepancy vanishes at  $Z_1 = 10$ . This is the periodic dependence of  $S_{e1}$  on  $Z_1$ . Among these light-heavy ions, the discrepancy becomes maximum at  $Z_1 = 7$  or 13. This is consistent with the present results and it is not due to errors in the experiment, but due to the characteristic of nitrogen between theoretical and experimental stopping parameters.

The exact comparison of  $\xi$  values between two kinds of data is not comprehensible unless every parameter, i. e.,  $M_2/M_1$ , Lindhard's  $k$  parameter, reduced dimensionless energy  $\epsilon$ , etc., is identical in both cases. No one can do significant comparison between <sup>14</sup>N and <sup>15</sup>N, for example, though the theoretical electronic contribution remains the same in the assumption.

There are no other data of Se to be discussed in comparison with the present one.

## VII. CONCLUSIONS

(i) It is recognized that the effective-charge parameter for  $N$  in Se is sufficiently lower than unity, similarly with  $N$  in other media, so that this effect has to be taken into consideration in the region  $E_A = 0.1 - 0.5$  MeV/amu. (ii) The relative mean excitation energy does not linearly change with  $Z_2$ . (iii) The electronic stopping cross section is not in proportion to the velocity of a projectile. (iv) Particularly among some light-heavy ions, stopping data of nitrogen as a projectile are far from theoretical predictions. Those discrepancies result from the potential characteristic of nitrogen itself.

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