Fusion yields for carbon-cluster impact on CD_2 targets

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Nuclear fusion yields for the secondary reaction D+D have been measured for carbon-atom anions and carbon-cluster anions incident on a deuterated polyethylene target for atom and cluster energies up to 324 keV. Comparison of the cluster- and atom-anion fusion yields per carbon at the same bombarding energy per carbon show no evidence of a collective enhancement for clusters as large as C_{19}. The absolute yields can be reproduced by a knock-on model. An upper limit to the fusion yield for $C_7D_7SO_3^-$ bombardment has also been determined.

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I. INTRODUCTION

In 1989 Beuhler, Friedlander, and Friedman [1] reported observing D+D fusion events where accelerated heavy-water cluster ions bombarded a deuterated target at very low deuteron velocities. The fusion yield was many orders of magnitude larger than expected on the basis of the extrapolation of measured D+D fusion yields to lower energies, and it was suggested that there was a enormous enhancement due to the collective effects associated with the other atoms in the projectile. A followup time-of-flight experiment [2] seemed to support the cluster origin of the fusion events, and an independent experiment by Bae, Lorents, and Young [3] confirmed the fusion yield for a 100-molecule cluster. Both Beuhler *et al.* [4] and Bae, Lorents, and Young [3] reported also seeing fusion events with normal-water cluster projectiles, although with an appreciably lower yield (~5%) than for deutrated clusters. This suggested an important role for the heavy atoms in producing anomalous fusion yields, as does also the failure to observe enhanced fusion with pure deuterium clusters [5]. These observations motivated the present investigation with pure carbon clusters. We wished to explore the fusion yields due to a heavy-atom cluster impact without the complications of other kinds of atoms in the projectile.

Since this work was initiated several developments have called into question some of the early results on cluster-fusion enhancement. In their report claiming to confirm the results of Beuhler *et al.* for $n = 100$ clusters, Bae, Lorents, and Young [3] also reported an enhancement for very small clusters. Work in our laboratory [6] has not confirmed this claim. For clusters of two to four molecules we observed one to two orders of magnitude less yield than observed by Bae, Lorents, and Young and our yields were consistent with the yields expected for single deuterons of the cluster velocity [6]. Very recently Beuhler *et al.* have published [7] an Erratum to their original work in which they report that their original cluster-impact fusion rates were overestimated by at least two orders of magnitude. The evidence for enhanced cluster-impact fusion is thus not very solid at this time. Nevertheless, we feel that the present results with carbon clusters define an important contributing mechanism which must be taken into account when attempting to characterize the origin of cluster-impact fusion yields.

II. EXPERIMENTAL PROCEDURES

Many of the important features of the acceleration and detection systems used in this study have been described previously in connection with our earlier heavy-water cluster experiments. The major difference in the present experiment was the use of a sputter ion source rather than a direct-extraction source for making the anions. The sputter ion source is located on the same high-voltage platform as the ion source shown in Fig. 1 of our previous publication [6], and injected into the 90° analyzing magnet through a 45° magnet between the direct extraction source and the 90° magnet. The sputter ion source, obtained from General Ionex, produces negative ions by the bombardment of a substrate by a focused beam of Cs^+ ions. The energy of the Cs beam is typically 4–5 keV. We produced carbon atomic anions and cluster anions by bombarding a graphite pellet. Attempts to enhance the yield of larger clusters using a C_{60} sample, supplied through the courtesy of Beck [8] and Kappes

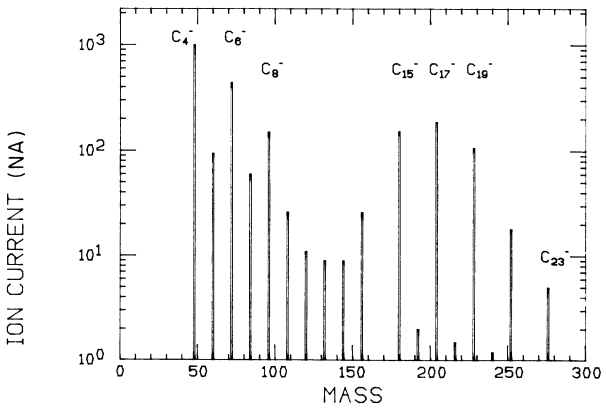

FIG. 1. Carbon-cluster mass spectrum obtained from sputter ion source.

[9], were not very successful. An example of a cluster-anion intensity distribution for graphite is shown in Fig. 1. The favoring of even-n clusters through $n=8$ has been observed previously [10,11], as has the enhancement of odd-n clusters for $n \geq 11$. The even-n anions through $n=8$ are attributed to linear chain structures and the larger odd-n anions are attributed to cyclic structures on the basis of molecular-orbital-theory calculations [12,13]. Our yields for ~5-keV Cs^+ ions fall less steeply with n and show a more prominent enhancement for $n=15,17,19$ than do those obtained with 14.5-keV Cs^+ ions [11].

We also report here an experiment with a deuterated organic molecule, p-toluene sulfonic acid. We synthesized this compound from deuterated toluene and deuterated sulfuric acid according to the procedure of Fieser [14]. The acid anion of this compound was produced in the sputter ion source. Only a few nanoamperes of current were obtained and source pellet lifetimes were short even under gentle conditions (low Cs boiler temperature and Cs-ion current).

The CD_2 targets were prepared by dissolving 98% enriched deuterated polyethylene [15] in deuterated xylene and letting the solution spread over a 0.000 35-in.-thick Al foil. Targets of about 1 mg/cm^2 thickness were obtained by this procedure. This CD_2 thickness plus the Al was sufficiently small that the 3-MeV protons from the D+D fusion reaction could be transmitted to the detector with modest energy degradation. The thickness and uniformity of the targets was checked by examination of the energy spectrum of 6.05-MeV particles transmitted through the foils. A troublesome problem during the course of the measurement was target deterioration (carbonization) of the front layer of the target with loss of target deuterons and a falloff of fusion yield with time. This falloff became noticeable after an integrated particle flux of about 10^{17} particles/cm^2. Frequent target changes during the course of the measurements were necessary, and care was taken to make measurements at a reference energy before and after a series of measurements with a given target. Nevertheless, uncertainties of up to 30% in our absolute yields arise from this problem. The proton energy spectra typically had a full width at half maximum of 10%.

III. RESULTS

The proton yield has been deduced from the number of proton counts and the detector geometry assuming an isotropic distribution. The results, expressed as protons per incident carbon, are plotted as a function of carbon atom energy in Fig. 2. On such a plot a collective enhancement of the fusion yield would reveal itself as a larger yield at a given carbon atom energy when that atom was part of a cluster. One observes that we have no evidence for cluster enhancement of the fusion yield for clusters up to $n=19$.

An extrapolation of our C^- yield to 1 MeV gives a yield in reasonable agreement with that reported by Mizota *et al.* [16]. The latter authors have also looked for but not observed cluster enhancement with O_2 and O_3 clusters.

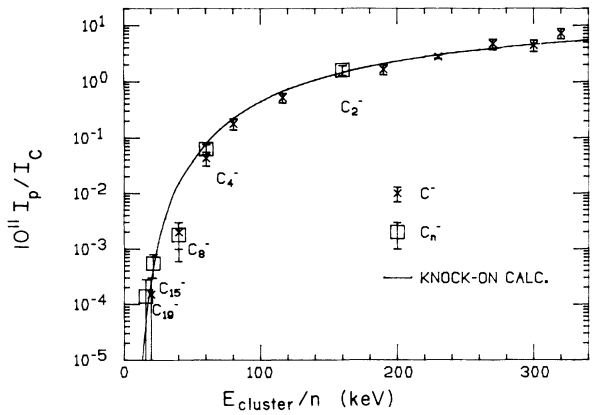

FIG. 2. Comparison of fusion yields for carbon clusters with those for single carbon atoms. Collective effects would be revealed on this plot by a larger yield for clusters compared to single atoms at the same value of energy per atom. I_p/I_C is the number of protons obtained per incident carbon atom.

We also looked for fusion with a beam of p-toluene sulfonic acid anion, $C_7D_7SO_3^-$. We observed two candidate events, which we treat as an upper limit, giving a proton-to-deuteron yield ratio of $\leq 1\times10^{-13}$ protons per deuteron at a bombarding energy of 324 keV.

IV. CALCULATION OF THICK-TARGET KNOCK-ON YIELD

The leading-order process by which one might expect D+D nuclear fusion for a projectile which does not contain deuterons is scattering of the projectile atoms off of target deuterons, leading to recoil deuterons with sufficient energy to react with other stationary deuterons in the target. This mechanism was one of several considered by Carraro *et al.* [17] in their early exploration of processes which could account for the fusion yields resulting from the deuterated water cluster bombardments of Beuhler, Friedlander, and Friedman [1]. Carraro *et al.* calculated the knock-on contribution of the projectile oxygen atoms. In their calculation this contribution generally exceeded that from the projectile deutrons.

The fusion yield from the knocked-on deuterons is given by

$$Y_{KO}=n_D n\int_0^E \frac{dE}{\left|\frac{dE}{dx}\right|}\int_0^{U_{max}(E)} Y_{TT}(U)\frac{d\sigma(U,E)}{dU}dU\ ,$$

where n_D is the deuteron number density in the target, n is the number of atoms per cluster, E_0 is the initial laboratory energy of each atom in the cluster, dE/dx is the stopping power of each projectile atom in the target, $U_{max}(E)=4m_1m_2E/(m_1+m_2)^2$ is the maximum energy that a projectile atom can transfer to a struck deuteron, $Y_{TT}(U)$ is the thick-target fusion yield per deuteron, and $d\sigma/dU$ is the differential energy cross section for the heavy projectile atom-deuteron collisions. Carraro *et al.* used a universal cross-section parametrization for

$d\sigma(U,E)/dU$ which, as they point out, is better suited for larger clusters than we are concerned with. We have found that a better choice in our energy and cluster-size domain is to assume Rutherford scattering, i.e., to neglect screening effects. Below roughly 20 keV the true scattering becomes less than Rutherford (Townsend, Kelly, and Hartley [18]) and our use of Rutherford scattering will lead to an overestimate of the knock-on yield. However, this overestimation is only significant for measurements at very low energy, due to the fast decrease in the fusion yield $Y_{TT}(U)$.

It is also necessary to parametrize the stopping of the projectile atoms in the CD_2 target. Although electronic stopping dominates at most energies, nuclear stopping begins to play an important role at the lowest energies. In our case we need to know the stopping of carbon in both carbon and deuterium. The former has been measured by Ormand and Duckworth [19] for carbon energies between 15 and 140 keV. We have parametrized the high-energy part of these data and added the nuclear stopping according to Lindhard, Scharff, and Schiott [20] to obtain a representation of the stopping over the needed dynamic range. For carbon stopping in deuterium the experimental C+C data in the region dominated by electronic stopping was scaled according to the Z scaling given by Fastrup, Hvelplund, and Sautter [21] and the nuclear stopping according to Lindhard, Scharff, and Schiott was added.

The thick-target D+D yield $Y_{TT}(U)$ was calculated in a manner similar to that of Carraro *et al.* except that the calculation was performed for a CD_2 target [6]. The knock-on yield calculated as described above is shown by the continuous curve in Fig. 2. The calculation provides a reasonable account of the energy dependence of the observed fusion yield, which varies more than four orders of magnitude over the energy range studied.

We have performed a similar calculation for 324-keV $C_7D_7SO_3^-$ ions. The dominant contribution to the calculated fusion yield comes from sulfur knock-on. We assumed Rutherford scattering and did not include nuclear stopping in this estimate, so the calculated result of 0.6×10^{-14} protons per deuteron should be an upper limit. This is well below our experimental upper limit.

V. CONCLUSIONS

We have found that useable beams of carbon cluster anions can be produced by Cs^+ bombardment of graphite. These beams can be used to induce D+D nuclear fusion even though there are no D's in the projectile. The absolute magnitude and the energy dependence of the yields for single carbon atom projectiles are well reproduced by a knock-on model calculation using available information on energy transfer, cross-section stopping power, and D+D fusion yields. A scaling procedure has been used to compare cluster yields with single-atom-projectile yields and no evidence for collective effects is seen for clusters as large as 19 atoms. Or to say it another way, the cluster yields per incident carbon are only determined by the velocity of the carbon atoms and are independent of whether there are other atoms in the projectile. It should be remarked that the magnitude of any collective-enhancement may depend on the geometry of the cluster. It is not clear at this time whether nearly spherical or linear structures would be most effective. A linear structure can have more atoms in a row but only for a narrow range of orientations. At the present time there is no definitive evidence for collective effects for any cluster geometry. For bombarding energies used in this and in the Beuhler *et al.* and Bae, Lorents, and Young studies it will be difficult to observe fusion for larger clusters unless a significant collective enhancement develops. This is because of the rapidly decreasing yield with increasing cluster size (and decreasing cluster velocity) and the inability to compensate with higher beam currents or longer runs due to target deterioration. The calculations performed for the $C_7D_7SO_3^-$ beam show that for heavy complex projectiles the knock-on contributions of the heavier atoms can dominate over the direct D+D fusion induced by deuterons in the projectile. This means that in interpreting results from studies with nonelemental clusters such as D_2O and CD_4 it will be necessary to carefully evaluate the noncollective contributions from both direct projectile deuteron-induced fusion and projectile heavy-atom knock-on induced fusion. The results reported here provide a calibration of calculational procedures for evaluating the latter contributions.

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