ELECTRON EMISSION FROM CONDENSED PHASE MATERIAL INDUCED BY FAST PROTONS[†]

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Monte Carlo track simulation has become an important tool in radiobiology. Monte Carlo transport codes commonly rely on elastic and inelastic electron scattering cross sections determined using theoretical methods supplemented with gas-phase data; experimental condensed phase data are often unavailable or infeasible. The largest uncertainties in the theoretical methods exist for low-energy electrons, which are important for simulating electron track ends. To test the reliability of these codes to deal with low-energy electron transport, yields of low-energy secondary electrons ejected from thin foils have been measured following passage of fast protons. Fast ions, where interaction cross sections are well known, provide the initial spectrum of low-energy electrons that subsequently undergo elastic and inelastic scattering in the material before exiting the foil surface and being detected. These data, measured as a function of the energy and angle of the emerging electrons, can provide tests of the physics of electron transport. Initial measurements from amorphous solid water frozen to a copper substrate indicated substantial disagreement with MC simulation, although questions remained because of target charging. More recent studies, using different freezing techniques, do not exhibit charging, but confirm the disagreement seen earlier between theory and experiment. One now has additional data on the *absolute* differential electron yields from copper, aluminum and gold, as well as for thin films of frozen hydrocarbons. Representative data are presented.

INTRODUCTION

Monte Carlo track simulation codes have been highly successful in many areas of radiation biology, from predicting radiation quality effects on DNA fragmentation spectra to aiding in the description of chromosome damage. Still questions remain regarding the accuracy of low-energy electron transport where theoretical methods are highly uncertain and whether these simulation techniques can accurately describe clustered damage occurring at electron track ends. For the past several years, authors have been studying electron transport phenomena in condensed material by analysis of the energy and emission angle of electrons ejected from the surface of thin films following their production within the film by fast heavy ion impact and their ultimate transport and emission from the film surface^(1, 2). These data should provide insight into the electron transport process in condensed phase material of interest in radiobiology and provide sensitive tests of Monte Carlo transport codes used in assessing effects of charged particle track structure.

Experimental technique

Our measurements have focused on electron transport following electron production by fast protons

and fluorine ions by studying the energy and angular distributions of electrons ejected from thin metal foils and from multilayers of molecules frozen to the metal surfaces. The electron energy spectra are obtained by time-of-flight analysis, using a pulsed ion beam, which provides optimum reliability for low-energy electrons. The relative yields are placed on an absolute scale by comparison of total electron yields for clean copper foils, obtained by integrating the relative yields over emission energy and angle, to published values for total electron yields^(3, 4). Condensed foils of amorphous solid water (ASW) and the hydrocarbons propane and butene were prepared by freezing vapour on sputter cleaned and cryogenically cooled (~50 K) copper foil substrates. The foils are held in ultra-high vacuum (1.33×10^{-7} Pa) and foil surfaces are sputter cleaned with low-energy neon ions both prior and between freezing cycles as needed to assure initial cleanliness.

Data for electrons ejected from ASW have been published recently⁽⁵⁾, so this brief report will focus on data from frozen hydrocarbons and differences observed between the different materials that have been studied. In these studies, the molecular target of interest is introduced into the ultra-high-vacuum chamber through a precision leak valve and frozen to a thin (1 μm) copper foil held at approximately 50 K by a closed-cycle helium refrigerator. The frozen foil thicknesses are determined by extrapolation of the usual assumption that one monolayer

[†]Manuscript for the poster presented in Verona by L.H. Toburen *et al.*, entitled 'Electron transport in condensed phase material', Abstract ID: 90.

(ML) of condensate is produced per second at a pressure of 1.33×10^{-4} Pa⁽⁶⁾. The pressure in the target chamber was measured with an ionisation gauge and recorded at one-second intervals as the target gas was leaked into the chamber and frozen to the Cu foil. The integral of the pressure over time was then used to determine the condensate thickness. Correction factors for the response of the ionisation gauge to different gases were provided by the manufacturer of the ion gauge: an ionisation constant of 1.0 was used for pressure measurements of water vapour, 4.0 for propane and 4.2 for butene. All of the frozen condensate films were prepared by so-called ‘background’ dosing, where the vapour is diffuse and the molecules impinge on the surface isotropically.

RESULTS

Spectra for low-energy electron emission at 45° from propane frozen on a copper substrate following the transmission of 6-MeV protons are shown in Figure 1 as a function of the thickness of the propane foil for a select number of the many thicknesses investigated. At the lowest electron energies measured, shown in the upper panel in Figure 1, it is observed that few MLs (approximately 50) are necessary to achieve saturation, i.e. additional thickness provides essentially no change in the spectrum. At the highest electron energies, shown in the lower panel of Figure 1, saturation occurs at approximately 100 ML. In the case of ASW, it was found that the electron emission spectrum saturated at 100–200 ML. At the saturation thickness, the condensate is ‘thick’ relative to the range of the electron, and the spectrum should be representative of the condensate and not the underlying substrate. At 200 ML, the ASW film is approximately 40 nm thick, which is approximately the range of a 1-keV electron.

An example of the full angular range of measurements of electrons from foils is shown in Figure 2 for 200 ML of ASW frozen to a copper substrate and bombarded by 6 MeV protons; these data are the average for two independent measurements. All spectra show a broad peak between 1 and 3 eV. The maximum in the peak increases from about 1 to 3 eV as the angle of emission increases from 15 to 60° in the forward direction and increases in the backward direction as the emission angle decreases from 155 to 115° (i.e. with increasing angle as measured from the normal to the surface). Similar data have been obtained for clean copper, gold and aluminium foils, as well as for the frozen hydrocarbons propane and butene. Data for copper targets and for ASW have been published^(3, 5); all of these data have been described in the doctoral dissertation of McLawhorn⁽²⁾.

Figure 1. Electron spectra observed for selected coverage of propane. Saturated coverage occurs for 50 ML at low energies and 100 ML at the highest energies. The yields approach those for the bare copper at the highest energies shown.

Figure 2. Electron emission from 200 ML thick ASW frozen on a 1-μm thick copper foil bombarded by 1-MeV $u^{-1}F^{4+}$ ions.

In Figure 3, a comparison of absolute yields for electrons ejected at 45 and 135° are shown for saturated coverage of ASW, propane and butene. All the molecular targets were prepared by background dosing with the respective vapours and frozen on the copper substrate at approximately 50 K. The data for the various targets in Figure 3 all show similarities within the experimental uncertainties of 20 to 30 % (representative error bars are shown as vertical lines at selected emission energies on the ASW spectra). The two hydrocarbon targets provide similar-shaped spectra but with the additional electrons of butene and propane leading to larger yields than ASW at the largest electron energies. In the forward direction, ASW has the largest yield at very

Figure 3. A comparison of the yield spectra for electrons emitted at 45 and 135° from saturated coverage of water, propane or butene frozen on a 1- μ m thick clean copper foil following passage of 6 MeV protons. The short vertical lines at selected electron energies are the estimated uncertainties in the water yields; comparable uncertainties can be expected for the hydrocarbon data.

low energies, but the yield drops more rapidly than the hydrocarbons as the electron energies increase past about 50 eV. The most important aspect of these results for modelling might be their similarities. Except for the structure in water at 10–15 eV, the similarities in spectral shape and absolute yields might allow modelling of low-energy electron transport with little differentiation between the target structure so long as the molecules do not differ greatly in atomic number, i.e. electron binding.

A major difference between the hydrocarbon spectra and that of ASW is the structure observed between about 10 and 15 eV in the ASW spectra. This structure has been studied extensively by Wilson *et al.*⁽⁷⁾ with electron impact excitation of ASW and is attributed to auto-ionisation decay of the $2a_1^{-1}4a_1$ excited state of water leading to a vacancy in the valence state. Although the electron impact velocity is somewhat less than that of the 6-MeV protons used in this study, the secondary electrons generated by proton impact have energies that range from below the threshold energy for auto-ionisation to well over the threshold for excitation of the $2a_1^{-1}4a_1$ state, and it is not surprising that the auto-ionisation structure was also seen in the obtained ASW spectra. In Figure 4, the spectra published by Wilson *et al.*⁽⁷⁾ are compared with the spectra obtained with fast proton excitation; here the data for electron impact has been normalised to the results obtained at 10 eV. Although the incident electron beam impinges on the target at 135°, whereas the proton beam hits the target at normal incidence and the ejected electron spectra is recorded at 135°, the peak shapes agree remarkably well. This

Figure 4. Electron emission yield from 6 MeV H^+ on ASW compared to results from 100 eV electron impact published by Wilson *et al.*⁽⁷⁾. The electron impact data were normalised to the proton data at 10 eV. Substantial differences in the geometry of the electron impact measurements exist, however the peak shape and size is remarkably similar.

may result because the majority of the excitation in the case of proton impact is actually produced by the secondary electrons that have undergone a great deal of direction and energy randomisation in the slowing down process, thus leading to results quite similar to scattered secondary electrons in the electron impact data.

FUNDING

This work is supported in part by the National Institutes of Health, National Cancer Institute Grant No. 2R01CA093351-04A1, and by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, US Department of Energy.

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