quantum mechanical analog because according to the latter view, "a molecule is something new and different from an additive combination of atoms."[14] In this respect, partial ionic character has approximately the same usefulness and validity as the concept of atomic radii.

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Selectivity Effects in Recoil Labeling of Methyl Iodide by Fission-Produced Iodine

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Recoil labeling of methyl iodide by fission-produced iodine isotopes was observed to be of higher efficiency with independently produced iodine than of the same isotopes recoiling after β-decay.

A study is being made of the formation of radioactive methyl iodide by the recoil of fission-produced iodine isotopes into methane, methyl iodide, and mixtures of methyl iodide with different gases. We have measured the isotope composition of the iodine in labeled methyl iodide formed in the reactions

$$I^* + CH_4 \longrightarrow CH_3I^* \quad (I)$$

$$I^* + CH_3I + CH_4 \longrightarrow CH_3I^* \quad (II)$$

$$I^* + CH_3I + He \longrightarrow CH_3I^* \quad (III)$$

Plastic irradiation containers ("rabbits") in which the interior was coated with a layer of $\sim$2 mg./cm.^{2} uranium were filled with the corresponding gases and irradiated in the neutron flux of the Israel Research Reactor IRR-1. The gas, in which a substantial quantity of the recoiling fission products of the uranium was stopped, was then extracted from the container and the methyl iodide was separated from the other gases by gas chromatography. Various gas chromatography columns were used and found adequate, the best being a 100-cm. column of 25% silicone fluid No. 702 on Sil-o-Cel firebrick of 50–70 mesh at 70°, with a flow rate of 80 ml./min. The CH_3I peak appeared after 1.5 min. The radioactivity of the methyl iodide fraction was analyzed with a scintillation spectrometer connected to a multichannel pulse height analyzer.

The γ-ray spectra of the purified fractions of methyl iodide from different sources disclosed differences in the relative yields of the radioactive iodine isotopes. Table I gives representative results for the ratio of I^{135} to I^{133}, as obtained from recoil labeling of a few gas mixtures, compared with the ratio of the same isotopes extracted from uranium irradiated and separated under the same time conditions.

TABLE I

RATIOS OF I^{135}/I^{133} FROM DIFFERENT SOURCES

Reaction	I^{135}/I^{133}[b]
Iodine from U^{235}(n,f)	1.0
$I^* + CH_4$ (760)[a]	1.5–2.0
$I^* + CH_3I$ (300) + CH_4 (460)	2.5–5.0[c]
$I^* + CH_3I$ (300) + He (460)	$\sim$5

[a] Indicated pressures are partial, in mm. [b] The error in each measurement of CH_3I is $\pm$25%. [c] For short irradiations (1 min. or less) the ratios are close to 5, and for longer irradiations ($\geq$10 min.) they are closer to 2.5.

It should be noted that I^{135} is formed in fission with a greater independent fission yield (2.37%) than I^{133} (<0.33%).[2] The ratio of the independent yields of I^{135} and I^{133} is then $\geq$7.2. If the ratio of the cumulative yields of these isotopes is taken as unity (it is actually 0.92), as presented in Table I, the ratio of the independent yields is $\geq$7.8.

The results shown in the table point to the occurrence of a process in which the formation of labeled methyl iodide by recoil is of higher efficiency when the recoiling iodine is produced independently, *viz.*, as a fission fragment, rather than when the iodine results from β-decay of its precursors.

A recent experiment[3] in which fission recoils were stopped in methane showed marked differences in the distribution of iodine isotopes in the gas as compared with the distribution on a negatively charged collector. It was indicated that the abundance of I^{135} compared to I^{133} was greater in the gaseous phase in which the radioactive iodine is believed to be mainly in the form of CH_3I.

Since all recoils resulting from β-decay are charged,[4] while primary fission fragments near the end of their range are not,[5] electric collection will be mainly effective with β-decay products, and it is thus to be expected that a fraction of fission products extracted electrostatically from the stopping gas will be enriched to a certain extent with β-decay products, or rather, depleted with respect to primary fission fragments. Hence, the effect observed in ref. 3, although very similar to that reported in this paper, can be attributed to *electrostatic fractionation* which is expected *a priori*, rather than to *selective labeling* of the gas. In addition, the form of the labeled gaseous compound has not been investigated experimentally.

Thus, to demonstrate unambiguously that selective labeling in methyl iodide does take place, it is a prerequisite to isolate the CH_3I from the stopping gas and compare the distribution of iodine isotopes in it with an *undisturbed* pattern of cumulative yields of iodine isotopes from fission.

The indication of the occurrence of selective labeling is in general agreement with the model[6] pointing at kinetic energy considerations as criteria for discrimination and selectivity in replacement reactions, like the ones studied in the present work.

(1) Part of a study leading to a Ph.D. degree.

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