the transparent crystals. With larger crystal fragments of SiS_2 II, uniaxial interference figures were observed in agreement with the tetragonal structure. In general, crystal fragments were colorless.

With sulfur-selenium melts of different compositions, the immersion method indicated mean indices of refraction for SiS_2 II and GeS_2 II of 2.24 and 2.40, respectively.

All crystals of SiS_2 II, GeS_2 II, and mixed compositions essentially behaved as insulators having electrical resistivities in excess of 10^{10} to 10^{11} ohm cm.

C. T. PREWITT
H. S. YOUNG
Central Research Department, Experimental Station, E. I. du Pont de Nemours and Company, Wilmington, Delaware 19898

References and Notes

1. M. S. Silverman and J. R. Soulen, *Inorg. Chem.* **4**, 129 (1965).
2. W. Büssem, H. Fischer, E. Gruner, *Naturwissenschaften* **23**, 740 (1935); E. Zintl and K. Loosen, *Z. Physik. Chem. Leipzig A* **174**, 301 (1935).
3. W. H. Zachariasen, *Phys. Rev.* **49**, 884 (1936); *J. Chem. Phys.* **4**, 618 (1936).
4. R. W. G. Wyckoff, *Am. J. Sci.* **9**, 448 (1925); *Z. Krist.* **62**, 189 (1925).
5. T. F. W. Barth, *Am. J. Sci.* **23**, 350 (1932).
6. W. A. Dollase, *Z. Krist.*, in press.
7. G. E. R. Schulze, *Z. Physik. Chem. Leipzig B* **24**, 215 (1934).
8. F. Dachille and R. Roy, *Z. Krist.* **111**, 451 (1959).
9. E. C. Lloyd, U. O. Hutton, D. P. Johnson, *J. Res. Nat. Bur. Std. C* **63**, 59 (1959).
10. S. Block, C. W. Weir, G. J. Piermarini, *Science* **148**, 947 (1965).
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Polonium-210 Content of Mainstream Cigarette Smoke

Abstract. *When eleven brands of cigarettes were smoked in a standardized manner, differences in the polonium-210 content of various brands were found. The differences were not directly related to the presence of a filter or to the construction of the filter, but were related to the amount of particulate matter in the mainstream smoke.*

Polonium-210, a radioisotope that emits α-particles, is a natural contaminant of cigarette smoke. Radford and Hunt (*1*) suggested that Po^{210} is an important factor in the genesis of bronchial cancer in smokers. These authors measured the Po^{210} content of cigarette smoke, whole tobacco, and samples of bronchial tissue. The validity of their conclusions regarding both the quantities of Po^{210} found in the lung and the importance of the role of Po^{210} in tumor initiation has been discussed (*2*, *3*). Radford and Hunt's data further revealed a marked difference between the Po^{210} content of mainstream smoke from filter and nonfilter cigarettes. Michelson (*2*) pointed out that the difference was substantial and might be related to the action of cigarette filters in removing portions of the particulate phase of the smoke. It has also been suggested that Po^{210} might explain the increased incidence of bladder cancer in smokers (*4*).

Because Po^{210} may be a significant factor in the initiation of bronchial cancer in smokers, it is important to have more accurate and extensive information about the quantities of Po^{210} in mainstream smoke. Furthermore, if there are real differences in the Po^{210} content of smoke among cigarette brands, they would probably be due to the action of cigarette filters. The characteristic of cigarette filters which makes them effective against Po^{210} should also be investigated.

To resolve these questions, cigarettes were smoked in a standardized manner, the particulate phase removed and measured, and the Po^{210} content analyzed. It was then possible to make a direct comparison between the amounts of Po^{210} and particulate matter in mainstream smoke. Eleven popular brands of cigarettes were tested. These included one regular-sized nonfilter, one regular-sized filter, and one king-sized nonfilter cigarette, and eight king-sized filter cigarettes. Cigarettes were purchased on the open market and used from freshly opened packs. They were smoked to approximately equal butt lengths, which required fewer puffs for regular-sized than for king-sized cigarettes.

The particulate phase of mainstream smoke was trapped on Millipore type AP fiberglass prefilter discs held in a modified Unico filter-disc holder with reduced dead space. Efficiency of the filter was checked in the following manner. Smoke from test cigarettes was passed first through a fiberglass filter and then through a Millipore filter—type HA, of 0.45-μ pore-size—which is assumed to act as an absolute filter for smoke particles. When both filters were weighed separately, it was found that a minimum of 96 percent of the particulate phase was retained by the fiberglass filter. Cigarettes held by a latex diaphragm cemented to a small glass funnel were smoked in a horizontal position. The latex diaphragm provides a leak-proof, distortion-free method for holding cigarettes and does not affect smoke flow. A falling-water-column apparatus provided suction standardized in 35-ml puffs of 2-second duration at 58-second intervals. The criteria for puff frequency, duration, and size are those set forth by

Table 1. The Po^{210} content of mainstream smoke of various brands of cigarettes. For each analysis two cigarettes were used. Except where indicated, nine puffs were used for each cigarette.

Brand	No. of analyses	Average butt length (mm)	Range	Radioactivity of mainstream smoke per cigarette (pc)
		Regular-sized filter cigarette		
A	9	32.2	(30–34)	.029 ± .0023*†
		Regular-sized nonfilter cigarette		
B	5	30.9	(29–32)	.043 ± .0032†
		King-sized filter cigarette		
C	9	32.1	(31–33)	.029 ± .0020
D	9	32.4	(31–34)	.032 ± .0023
E	6	34.4	(30–36)	.031 ± .0025
F	11	34.3	(33–35)	.033 ± .0021
G	8	32.2	(31–34)	.038 ± .0019
H	8	35.4	(34–37)	.038 ± .0023‡
I	6	34.0	(32–36)	.038 ± .0030
		King-sized nonfilter cigarette		
J	6	34.6	(32–36)	.042 ± .0021
		King-sized filter cigarette		
K	6	34.3	(33–35)	.044 ± .0033

* Plus or minus standard error. † Six puffs for each cigarette. ‡ The average number of puffs was 9.25; the range was nine to ten.

Table 2. Correlation between the Po^{210} content and particulate matter of cigarette smoke.

Brand	Radioactivity per smoked cigarette (10^3pc)	Particulate matter per cigarette (mg)	Ratio of radioactivity to amount of particulate matter
	Regular-sized filter cigarette		
A	29	10.5	2.76
	Regular-sized nonfilter cigarette		
B	43	14.9	2.88
	King-sized filter cigarette		
C	29	11.5	2.52
D	32	9.5	3.37
E	31	13.1	2.36
F	33	11.4	2.89
G	38	16.4	2.31
H	38	15.2	2.50
I	38	16.5	2.30
	King-sized nonfilter cigarette		
J	42	17.1	2.45
	King-sized filter cigarette		
K	44	16.5	2.66

the Analytical Methods Committee of the Tobacco Chemists Conference (*5*). After the smoking sequence, the inside surfaces of the funnel and the filter chamber were wiped with a small piece of filter disc, and the material thus obtained was combined in the plating procedure with that retained on the disc. The Po^{210} content of the tobacco from whole, unburned cigarettes was also determined. Tobacco and mainstream smoke retained by filter discs were prepared for counting in the same manner. Tobacco or filter discs were wet-ashed in concentrated HCl at 60°C and then transferred to a plating chamber constructed from a nursing bottle which held a 2.5-cm planchet cut from milled sheet fine-silver (*6*). The contents of the chamber were stirred for 4 hours while being heated in a boiling water bath. The silver planchets were rinsed with deionized water and air-dried. Samples were counted in a gas-flow proportional counter with a background of less than 0.8 count per hour.

There were no significant differences in the Po^{210} content of whole tobacco between the brands tested. The amounts of Po^{210} found were very similar to the amounts reported by Radford and Hunt (average for tobacco from regular-sized cigarettes, 0.39 pc).

In confirmation of the work of Radford and Hunt, Po^{210} was found in mainstream smoke (see Table 1). However, the quantities of Po^{210} were considerably lower, amounting to about three-eighths of the amounts reported by those authors. Differences in the smoking methods could account for this discrepancy (*7*). These differences are considerable and are as follows: gas-flow rate, 17.5 ml/sec as against 15 ml/sec; puff length, 2 seconds as against 2 to 3 seconds; puff frequency, 60 seconds as against 50 seconds; number of puffs (regular-sized cigarettes), 6 as against 8; butt length (regular-sized cigarettes), 33.4 mm as against 24.5 mm (estimated). When the smoking method and the puffing sequence of Radford and Hunt were simulated, larger amounts of Po^{210} were obtained in mainstream smoke than under the conditions described here, but the total amounts reported by those authors were never quite reached. The Po^{210} content of mainstream smoke apparently depends upon the manner of smoking.

It is generally accepted that the 35-ml-per-minute, 2-second-puff sequence imitates closely the dynamics of human smoking. Pfyl (*8*), in 1933, was the first to recognize the need for controlling puff-rate, volume, frequency, and time. Bradford *et al.* (*9*) made the first attempts to regulate those parameters to approximate the manner of human smoking in order to be able to make in vitro data valid for an in vivo situation. Bradford measured the smoking characteristics of a large number of smokers and found on the average that 35-ml puffs of 2-second duration were taken with a frequency of one puff per minute. Other workers reported similar average volumes of 32 and 40 ml (*8, 10*). Wolman (*11*) accepted the conditions described by Bradford (*9*). Schurr and Richards (*12*) also found similar averages in studying the smoking habits of 20 male subjects. Designers of smoking machines have been aware of the need to imitate the manner of human cigarette puffing (*12, 13*), and this is the method most widely used in smoking machines. Determinations of Po^{210} therefore should be made according to this standard method rather than by a method which does not approach the manner of human cigarette puffing.

As shown in Table 1, there are considerable differences between the mainstream smoke from various brands of cigarettes. These differences are statistically significant wherever two means differ by more than three standard errors. The values are not related to the size of the cigarette tested, nor are they directly related to the presence or absence of a filter. There is, however, a marked relation between the Po^{210} content and the amount of particulate matter in the mainstream smoke, as shown in Table 2. The mean ratio of radioactivity to particulate matter is 2.64. With the exception of cigarette D, all of the values for this ratio are within 13 percent of the mean ratio. It is reasonable to assume that a relation exists between the Po^{210} content and the amount of particulate matter, because the polonium is apparently adsorbed on smoke particles during the combustion process. Brand characteristics in terms of Po^{210} content appear to depend on the amount of particulate matter in the mainstream smoke, and neither directly on the presence or absence of a filter nor on the nature or construction of the filter.

THOMAS F. KELLEY

Bio-Research Consultants, Inc., Cambridge, Massachusetts 02141

References and Notes

1. E. P. Radford, Jr., and V. R. Hunt, *Science* **143**, 247 (1964).
2. I. Michelson, *ibid.*, p. 917.
3. E. P. Radford, Jr., and V. R. Hunt, *ibid.* **144**, 366 (1964); K. W. Skrable, F. J. Haughey, E. L. Alexander, *ibid.* **146**, 86 (1964); E. P. Radford, Jr., V. R. Hunt, J. B. Little, *ibid.*, p. 87.
4. H. Eisler, *Science* **144**, 952 (1964).
5. C. L. Ogg, *J. Assoc. Offic. Agr. Chemists* **47**, 356 (1964).
6. E. P. Radford, Jr., V. R. Hunt, D. Sherry, *Radiation Res.* **19**, 298 (1963).
7. E. L. Wynder and D. Hoffman, *New England J. Med.* **262**, 540 (1960); E. L. Wynder and D. Hoffman, *Adv. Cancer Res.* **8**, 249 (1964); J. R. Newsome and C. H. Keith, *Tobacco Sci.* **1**, 58 (1957).
8. B. Pfyl, *Z. Unters. Lebensm.* **66**, 501 (1933).
9. J. A. Bradford, W. R. Harlan, H. R. Hammer, *Ind. Eng. Chem.* **28**, 836 (1936).
10. R. B. Derr, A. H. Riesmeyer, R. B. Unangst, *ibid.* **27**, 771 (1937).
11. W. Wolman, *J. Amer. Med. Assoc.* **152**, 917 (1953).
12. M. O. Schurr and J. C. Richards, *Tobacco Sci.* **1**, 13 (1957).
13. A. E. O'Keeffe and R. C. Lieser, *ibid.* **2**, 73 (1958).
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