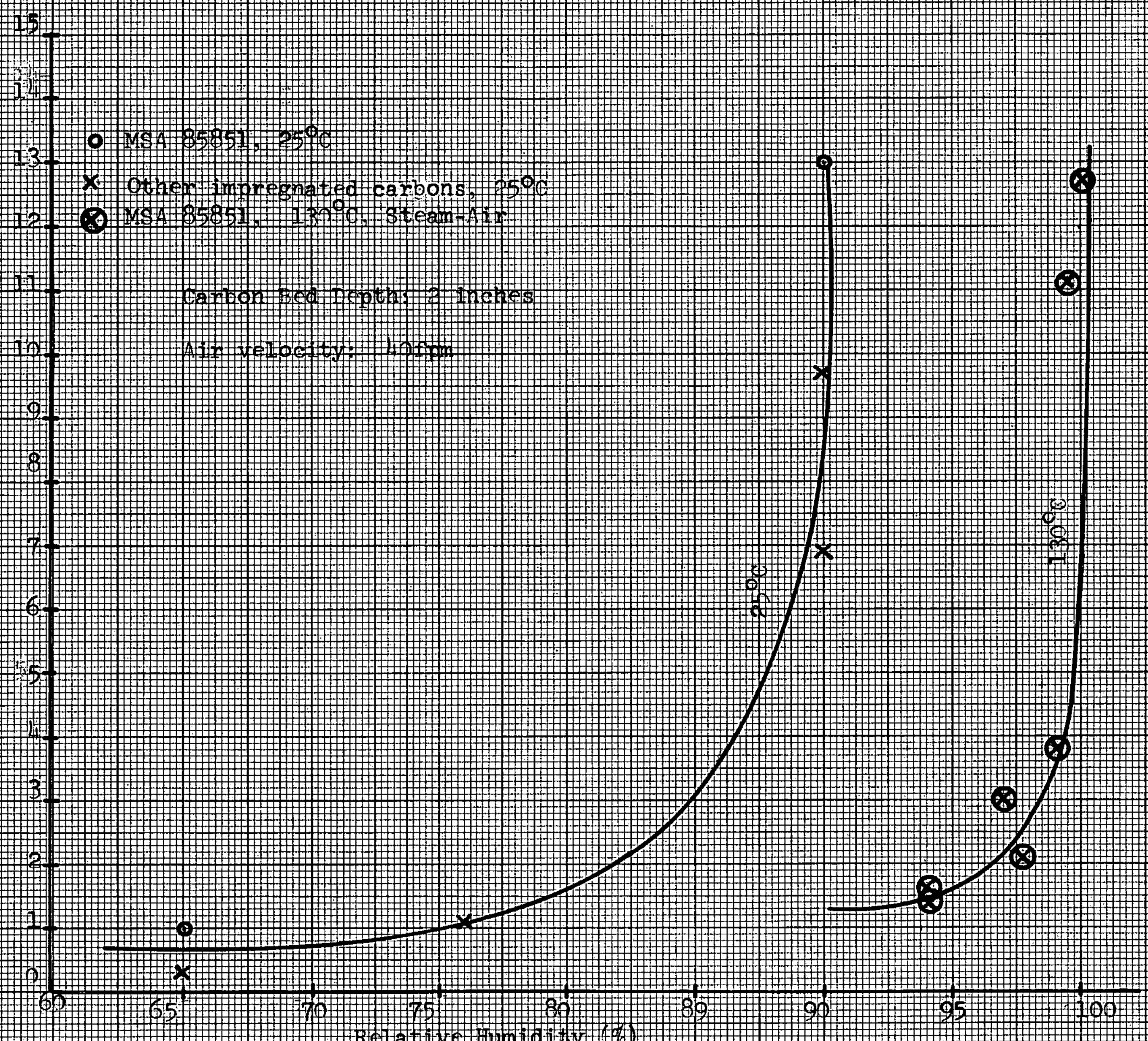
Attachment 1

Consolidated Edison requests a change on Page 4.5-4 of the Technical Specifications in the test conditions specified for measuring iodine removal efficiency of the charcoal filters. This revision would require that a test of the charcoal's ability to remove methyl iodine from the air be performed under test conditions which would yield the most accurate results achievable with the current state of the art. Presently, other Westinghouse PWR facilities are analyzing the iodine removal efficiency of their charcoal filters by performing tests run under conditions similar to those being proposed in this letter.

In support of this proposal to change the test conditions, a report by R. E. Adams consultant to Consolidated Edison dealing with the validity of the results obtained by the testing method prescribed by Technical Specifications and that prescribed here in is included. In the report, Mr. Adams, states that the performance of new charcoal filters have been established under a wide range of conditions by the tests performed at the Oak Ridge National Labratories. The periodic testing of the filters required by the Technical Specifications should therefore, be used only to established that the charcoal has not degenerated to a point where it can no longer meet the iodine removal efficiencies as given in the Specifications. Tests performed at a temperature of 25°C, ambient pressure and a relative humidity of 70% will provide this information and in fact, as a result of the lower temperature, will yield more conservative results than those provided by a test done at the present required conditions.

The revised Page 4.5-4 of the Technical Specification is attached.

Figure I Comparison of Data From 25°C and 130°C Tests



## References


1. Adams, R.E., Ackley, R.D., and Browning, W.E., Jr., "Removal of Radioactive Methyl Iodide from Steam-Air Systems", USAEC Report ORNL-4040 (January 1967)

2. Ackley, R.D., and Admas, R.E., "Removal of Radioactive Methyl Iodide form Steam-Air Systems (Test Series II)", USAEC Report ORNL-4180 (October 1967)

3. "Connecticut Yankee Charcoal Filter Tests", Connecticut Yankee Atomic Power Company Report CYAP-101 (December 1966)

4. Ackley, R.D. and Adams, R.E., "Trapping of Radioactive Methyl Iodide from Flowing Steam-Air: Westinghouse Test Series", USAEC Report ORNL-TM-2728 (December 1969).

5. Swanks, J. H., "In-place Iodine Filter Tests at the High Flux Isotope Reactor", USAEC Report ORNL-TM-1677 (December 1966).

6. Personal Communication by R. E. Adams with Members of Reactor Operations Division and Inspection Engineering Division of the Oak Ridge National Laboratory.

7. Ackley, R.D., Combs, Zell and Adams, R.E., "Aging, Weathering, and Poisoning of Impregnated Charcoals Used for Trapping Radioiodine", USAEC Report ORNL-TM-2860 (March 1970).

8. Davis, R.J. and Ackley, R.D., "Long-Term Effects of Radio-iodine Trapping by Charcoal", 12th AEC Air Cleaning Conference, CONF-720823 (January 1973).

9. Wilhelm, J.G., Dillman, H.G., and Gerlach, K., "Testing of Iodine Filter Systems Under Normal and Post-Accident Conditions", 12th AEC Air Cleaning Conference, CONF-720823 (January 1973).

10. Bennett, E.C. and Strege, D.E., "Evaluation of Weathered Impregnated Charcoals for Retention of Iodine and Methyl Iodide", Douglas United Nuclear Report DUN-7985 (June 1972).

11. "Gas-Phase Adsorbents for Trapping Radioactive Iodine and "Iodine Compounds", USAEC RDT Standard M16-1T (June 1972)

12. Adams, R.E., Ackley. R.D., and Shields, R.P., "Application of Impregnated Charcoals for Removing Radioiodine from Flowing Air at High Relative Humidity", pp. 387-402, IAEA Symposium on Treatment of Airborne Radioactive Wastes, New York, August 26-30, 1968.

7. From the standpoint of nuclear safety philosophy, test results from room temperature methyl iodide tests on exposed carbon would be more conservative than high temperature steam-air tests.

iodide efficiency test under conditions of high temperature, high relative humidity and high pressure.

The 25°C, 70% relative humidity, methyl iodide test is considered more applicable for the following reasons:

1. High temperature, steam-air tests are very difficult to perform and no such capability to perform this test is available in industry at the present time.

2. High temperature, steam-air tests are very succeptible to steam condensation in the carbon bed and test results are not valid if this occurs. Invalid test results could cause replacement of carbon in a reactor filtration system where such replacement is not needed.

3. The very performance of the high temperature, steam-air test could very easily mask the presence of carbon degradation that is being sought by the performance of the test.

4. Room temperature tests are much less difficult to perform and laboratories capable of providing this service now exist.

5. Moisture condensation at 70% relative humidity is not a problem and test results would be more consistent and acceptable.

6. Room temperature tests would be more sensitive in detecting carbon degradation by adsorbed organic material; little, if any, desorption of organic material would occur during startup of the tests or during conduct of the test.

might well be destroyed by the very act of performing the methyl iodide test under the assumed accident conditions.

It might be argued that the same process could occur during introduction of steam-air mixtures into the carbon filters following the assumed accident. This could happen, but to be consistent with the safety philosophy of the nuclear industry, no credit should be taken until the process has been demonstrated and proven.

Based upon the foregoing discussion, an examination of the applicability of the room temperature methyl iodide test is in order. A test that will measure the degradation of carbon by adsorbed organic material under test conditions unlike those of application might be more acceptable than a test under the conditions of application that will introduce an unknown factor by changing the condition of the carbon during the performance of the test. As discussed earlier, the room temperature test has been used at several laboratories to detect carbon degradation by absorbed organics. The room temperature test will produce conservative results in view of the possible "re-activation" of the carbon during the 130°C tests. This conservatism is in keeping with the AEC nuclear safety philosophy.

Conclusions

Serious consideration should be given to the use of the 25°C, 70% relative humidity, methyl iodide efficiency test to monitor carbon degradation in field applications, rather than the methyl industrial community in the near future. Also, from a technical viewpoint, there appears to be a more subtle factor that must be considered. To prevent unwanted steam condensation in the testing apparatus, it is necessary to heat the test apparatus and influent air to a constant temperature somewhat higher than the desired temperature of testing prior to the addition of steam; this step requires several hours to perform. Upon initial addition of steam, the temperature of the carbon under test rises significantly because of heat evolved during initial adsorption of water vapor by the carbon; several hours are required to dissipate this heat and trim the temperature of the system to the value desired. In all, the carbon is subjected to high temperature air and steam-air flow for as much as 5 or 6 hours prior to the introduction of radioactive methyl iodide. These preheating steps would not affect the physical condition of <u>new</u> impregnated carbon being tested for qualification as to use in steam-air atmospheres; however, these preheating steps could very well affect the physical condition of <u>exposed</u> carbon being tested for degradation. It has been noted earlier that adsorbed organic materials are the predominant cause of carbon degradation for methyl iodide trapping (ignoring for the moment the influence of condensed steam on methyl iodide efficiency). The preheating steps could, very possibly, desorb and elute organic material from the carbon prior to the introduction of the radioactive methyl iodide; so, in effect, the test would be performed on carbon that has been "reactivated" and unlike the bulk of exposed carbon still contained in the air filtration systems of the reactor containment. The evidence of degradation

These test procedures are based upon the experience derived during the extensive R&D program on carbon carried out at ORNL during the period 1965-1970. To illustrate the type of data produced under these two sets of testing conditions, Figure I was developed; data were taken from References 4 and 12. The two curves are very similar in shape with the 130°C curve displaced to higher values of relative humidity for a given value of methyl iodide penetration. This behavior is consistent with the observation that, within limits, elevated temperature (and possibly increased pressure) enhances the isotopic exchange process and thereby increases the methyl iodide trapping efficiency. From the graph, it may be observed that tests at 25°C, 70% relative humidity and 130°C, 95% relative humidity would each produce an efficiency of about 99%. At 90% relative humidity, the 25°C test would produce an efficiency of about 90% while the 130°C test would produce an efficiency of about 99%. These observations, of course, assume that no condensation of steam takes place in the 130°C tests.

For PWR application, either set of test conditions might be considered for use in monitoring carbon degradation. One usually assumes that test data obtained under actual conditions of application are more acceptable; however, this assumption may not be true in this case. Steam-air tests are difficult to perform and require rather expensive and sophisticated equipment as well as an experienced operator; for this reason, the capability for adequately performing such tests may not be available within the efficiency, the efficiency for elemental iodine was practically unchanged.

## Field Monitoring to Detect Carbon Degradation

As a consequence of the reported degradation of impregnated carbon in field applications, it is essential that the condition of the carbon be monitored periodically because of the safety-related function of these air filtration systems. Ideally the condition of the carbon would be measured with an inplace test, but no such test procedure is available for commercial nuclear power reactors. The alternative is to remove a sample of the exposed carbon periodically for testing in the laboratory. If the methyl iodide efficiency has dropped below the value dictated by the Technical Specifications, then the exposed carbon should be replaced with new carbon of known properties and efficiencies.

## Laboratory Testing of Exposed Carbon Samples

The methyl iodide efficiency value for the exposed carbon sample, as determined by laboratory test, provides the basis for determining if degradation of the carbon has proceeded to the extent that new carbon is required in the carbon adsorber system. Laboratory test procedures suitable for this purpose are contained in RDT Standard M16-1T (June 1972) (11). The two test procedures are identical except for experimental conditions. One procedure is carried out at 25°C, atmospheric pressure, and 70% relative humidity (room temperature test); the other procedure is carried out at 130°C, 50 psia, and 95% relative humidity (steam-air test).

The results of a similar study at ORNL on carbon by another manufacturer, but similar in type to MSA-85851, indicated some greater losses in methyl iodide efficiencies when samples of the carbon were exposed to flowing air from the LITR-BSR off-gas and the HFIR off-gas over a six-month period (8).

Comparable experience with carbon degradation has also been reported with continuously operating off-gas systems associated with West German nuclear power reactors (9).

A program is underway by Douglas United Nuclear to study carbon degradation in the off-gas systems of the N Reactor (Hanford). An example of gross degradation of carbon by adsorption of an unidentified organic material may be observed in the first progress report (10). The methyl iodide efficiency for a one-inch depth of an impregnated carbon similar to MSA-85851 dropped from 92% to about 10% over a six-month period of exposure; infrared spectroscopy indicated that the exposed carbon contained 29.5 mg of an unidentified complex hydrocarbon per gram of carbon.

To summarize, reported experimental data support the following conclusions: (1) simple aging of impregnated carbon in a protected atmosphere does not seem to affect the methyl iodide efficiency; (2) continuous exposure to flowing air containing traces of organic vapors does reduce the methyl iodide efficiency and the extent of degradation will depend upon the actual conditions at the site of application; (3) in all studies reported, while the continuous exposure to flowing air produces a decrease in methyl iodide subjected to inplace tests with radioactive iodine and radioactive methyl iodide at approximately six-month intervals. During the period December 1966-October 1969, the carbon filters (2-inch combined depth of carbon) contained MSA-85851 impregnated carbon. Over a 820-day period of continuous exposure, the methyl iodide efficiency dropped from 99.7 to 54%; during the same period, the elemental iodine efficiency dropped from 99.95 to 99.85%. Details of the test procedures have been reported (5) and the test data were obtained from records at ORNL (6).

Another study of carbon degradation was also performed at ORNL (7). This study involved the measurement of methyl iodide efficiency (under ambient conditions) for carbon samples exposed in three ways: (1) carbon stored in closed containers for simple aging effects; (2) carbon continuously exposed to flowing clean, humidified air (influent air passed through HEPA filters and carbon beds to remove particulates and organic vapors; and (3) carbon continuously exposed to flowing air (not pretreated) removed from the atmosphere within the ORR reactor building. The results for MSA-85851 carbon indicated that simple aging of the carbon had no noticeable effect on methyl iodide efficiency over the 17 months of the project; continuous exposure to pre-cleaned air in the laboratory over a 12-month period reduced the methyl iodide efficiency (ambient conditions) from about 99% to about 97%; continuous exposure to flowing air from the interior of the ORR building, over a 15-month period, reduced the methyl iodide efficiency from about 99% to about 91%.

a second program of tests was performed at ORNL during 1969; this program, sponsored by the AEC and Westinghouse, consisted of 18 tests and was performed using MSA-85851 impregnated carbon (4). A new and enlarged testing apparatus was constructed at ORNL to allow very sensitive control of the operating parameters of temperature and pressure and thus avoid unwanted (and sometimes unobserved) steam condensation in the carbon under test at very high relative humidities. Test results indicated methyl iodide efficiencies ranging from 98.6 down to 88.9% at calculated relative humidities ranging from 94 up to 99.6%.

## Degradation of Impregnated Carbons in Field Applications

Degradation of carbon in field applications with respect to methyl iodide trapping efficiency can occur by adsorption of organic vapors onto the carbon surfaces. This degradation is, of course, more rapid and more severe when the carbon is continuously exposed to flowing air containing these vapors. To date, little experience has been gained regarding degradation of carbon contained in standby systems not exposed to air flow. Examination of the experience gained in continuous flow systems would be of value to point out what one might expect, to a lesser degree, for carbon systems on standby.

The HFIR reactor at Oak Ridge utilized a continuous flow air filtration system to exhaust air from the reactor building; the effluent air normally contains no radioactivity and would contain radioactivity only in the event of an accident. The systems are

## Qualification of Impregnated Activated Carbons

The applicability of the type of impregnated carbon contained in the Indian Point Unit No. 2 containment filter systems (MSA-85851) has been well-documented in the literature. This carbon was included in laboratory studies (1,2) at the Oak Ridge National Laboratory (ORNL) during 1966-67 and in engineering scale studies by Westinghouse (3) in 1966. Methyl iodide efficiencies above 90% were reported for relative humidities <93% in the ORNL reports; efficiency values for relative humidities >93% were suspect because of experimental difficulties inherent in performing tests at humidities approaching 100% at temperatures around 280°F and pressures around 60 psia. Results of the Westinghouse tests performed in a circulating loop system indicate that the MSA carbon produced instantaneous methyl iodide efficiencies of 85-90% under relative humidities ranging from 92 to 95%; temperatures were around 260°F and pressures around 40 psig.

The conduct of lab tests under high temperature, high pressure, steam conditions at relative humidities approaching 100% is not a simple matter. Slight changes in the operating parameters of temperature and pressure can cause condensation of water in the carbon under test resulting in a large decrease in methyl iodide efficiency. For instance, in the proximity of saturation at these temperatures, a change of only 1°F results in a change of about 1.6% in relative humidity. Because of some uncertainties regarding the ORNL tests of 1966-67 at relative humidities >90%, vapors in the normal containment atmosphere and the presence of large quantities of water vapor in the post-accident environment can produce a lowered methyl iodine trapping efficiency by adsorption on the carbon surfaces. In the case of PWR applications, the moisture content of the post-accident atmosphere cannot be controlled and the limitations on carbon efficiency must be determined by laboratory study and allowances made to predict the probable carbon efficiency. Degradation of the carbon by adsorbed organic material (or by other unanticipated conditions) can be monitored over the service life of the carbon by periodically removing samples of the exposed carbon for laboratory analysis.

A logical plan, therefore, to ensure that the impregnated carbon filters installed in a PWR containment will function efficiently in a post-accident environment, would be to qualify the particular carbon, in the laboratory, under the anticipated conditions of temperature, humidity, and pressure; handle the other variables such as carbon bed depth, air flow velocity, etc. by careful design; and monitor for possible degradation of the carbon as a result of organic adsorption or simple aging by removing carbon samples periodically for laboratory determination of methyl iodine efficiency under assumed accident conditions. If the laboratory test indicated a lowering in methyl iodide efficiency below a predetermined value, then the exposed carbon filters would be replaced with new ones.

R. E. Adams 11/12/73

Monitoring of Degradation of Impregnated Activated Carbon in Standby Air Filtration Systems of PWR Containment

The application of impregnated activated carbon as a means of removing radioactive iodine and methyl iodide from containment atmospheres following an accident is now accepted and air filtration systems containing impregnated carbon are utilized in many nuclear power reactors. Laboratory research and field experience have demonstrated that this type of activated carbon is very efficient for the trapping of radioactive elemental iodine under most any condition of application in nuclear power reactor systems and further, retains this capability under adverse conditions of exposure. The same is not true for the isotopic-exchange trapping of radioactive methyl iodide. Laboratory research has shown that this mechanism of trapping can be enhanced or degraded in many ways. The effects of variables such as carbon bed depth, air flow velocity, carbon particle size, quantity of iodine impregnation, etc. can be handled by specifications and careful design of the system. However, the influence of the two major degrading factors (moisture and organic vapors) is dictated by the conditions in the containment environment preceeding and following an accident. Both of these materials interfere with the isotopic-exchange mechanism by adsorbing on the carbon surfaces and limiting the approach of the radioactive molecules of methyl iodide to molecules of impregnant iodine (or iodide) whereby isotopic exchange takes place. The presence of organic