Laboratory Testing of Nuclear Grade- Activated Charcoal Since Implementation of Generic Letter 99-02

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Abstract

NRC Generic Letter 99-02, "Laboratory Testing of Nuclear–Grade Activated Charcoal" was issued June 3, 1999. This generic letter required utilities to change the testing of charcoal samples from each ventilation system to ASTM D3803-1989 using 95% RH for systems with no humidity control and 70% RH for systems with humidity control. This paper discusses the laboratory tests results performed at NUCON since the issuance of the generic letter and their impact.

Introduction

Beginning in June of 1973, the NRC has issued a regulatory guide to provide guidance for the design, testing and maintenance of adsorption units of light water cooled nuclear power plants. This guide (and its subsequent revisions) has relied upon accepted industry test standards for testing new and used (surveillance) carbon samples. As the standards changed over time to reflect more current understanding, the regulatory guide was also revised. The present accepted test method, ASTM D3803-1989, came about as a result of this process of change and revision culminating in Generic Letter 99-02 in June of 1999 and the issuance of Regulatory Guide 1.52 Revision 3 along with its companion, Regulatory Guide 1.140 Revision 2, both in June of 2001. The testing methodologies used in earlier versions of the regulatory guide along with laboratory test results obtained since the publication of the generic letter are discussed.

Test Requirements and Methods for Different Versions of Regulatory Guide 1.52

The test requirements and methods used in the different version of the Regulatory Guide 1.52 are discussed below and summarized in Table 1.

Regulatory Guide 1.52, June 1973

New carbon tests from Table 1 of the guide.

Test 5a:

An elemental iodine carbon qualification test at design basis accident (DBA) temperature and pressure with the DBA temperature and pressure rounded to the next higher decade. The test method used was RDT Standard M-16-1T, June 1972 paragraph 4.5.2 which required pre-equilibration with a 130°C, 95% RH air until the temperature differential across the carbon bed was less than 1°C then injection of 130°C saturated steam with the air flow for 2 hours while maintaining approximately 95% RH and 50 psia steam pressure. Then the challenge gas was

injected for 1.5 hours followed by another 1.5 hours of steam-air flow. Test bed depth specified was 1"and flow velocity specified was 11-13 m/min. Acceptable result: 99.9%. The tests were to be repeated three times and averaged. No tolerances were given for the temperature and RH.

Test 5b:

A methyl iodide test performed according to paragraph 4.5.4 of the same RDT standard except DBA temperature and pressure were used. This test was the same as 5a. except methyl iodide was used as the challenge agent. This was a batch test with an acceptable result of 95% when performed at 95% RH and 99.5% when performed at 70% RH.

Test 5c:

A methyl iodine test performed according to paragraph 4.5.5 of the same RDT standard with the test bed pre-equilibrated with 25°C, 70% RH air (no time duration given), challenged for 2 hours then heated to 180°C with the flow stopped and then continued with airflow for 4 hours. This was a qualification test with an acceptable result of 99%.

Used carbon test from Table 2 of the guide:

For 2 inch deep beds operating inside containment, test at 95% RH at maximum design temperature and design velocity for an elemental iodine penetration of less than1% and a methyl iodide penetration of less than 10% (RDT assumed as test method).

For 2 inch deep beds outside containment with RH control test at 70% RH and maximum design temperature and design velocity with methyl iodide for a penetration less than 1%. For deeper beds, the allowable penetration was less than 0.175% (again, RDT assumed as test method).

No guidance was offered for testing carbon beds outside containment without humidity control.

Regulatory Guide 1.52 Revision 1, July 1976

New carbon tests from Table 2 of the guide:

Test 5a:

This was a methyl iodide removal test performed according to paragraph 4.5.3 of RDT M16-1T, October 1973 at 25°C except at 95% RH. Paragraph 4.5.3 was the same as 4.5.1 except methyl iodide was used as the challenge agent. The test bed was equilibrated with the conditioned air until the differential across the bed

was less than 1°C. The challenge period was 2hours and then the airflow was continued for another two hours. Tolerances were ± 1 m/min for the velocity, ± 2°C for the temperature and ± 5 % for the RH. This was a qualification test to establish the suitability of the product and was to be repeated three times. An acceptable result was 99%.

Test 5.b:

This was a methyl iodide removal test as 5.a. except 80°C and 95% RH was required for the test and 25°C for pre-and post-loading sweep. This was a batch test with 99% as an acceptable result.

Test 5.c:

This was a methyl iodide removal test to be performed for qualification purposes on carbon beds used in containment according to paragraph 4.5.4 of RDT. This paragraph is identical to paragraph 4.5.2 except methyl iodide is used. The test parameters to be used were 130 ± 2°C, 95 ± 5% RH (air pressure 20 psia +2, steam 95 ± 1% of the saturated vapor pressure at the test temperature). An acceptable result was 98%.

Test 5.d:

This was an elemental iodine retention batch test performed according to a test by the Savannah River Laboratory. The loading with iodine was performed at 25°C at a low but unspecified humidity for ten minutes followed by elution at 180°C for 4 hours. Acceptable results were 99.9% for loading and 99% for loading pus elution.

Used Carbon Tests from Table 3 of the Guide:

Test 5.c:

This test was for systems inside containment for a methyl iodide penetration of less than 10%.

Test 5.b:

This test was for systems operated outside the primary containment with humidity control. Tested at 70% RH for a methyl iodide penetration of less than 1% for 2 inch beds and less than 0.175% for beds greater than or equal to 4 inches.

No guidance for systems without humidity control outside containment.

Regulatory Guide 1.52 Revision 2, March 1978

New carbon tests:

Table 5.1 of ANSI N509-1976 replaced Table 2 of revision 1 of the guide and was identical to it.

Used carbon tests:

Table 2 of revision 2 of the guide was identical to Table 3 of revision 1 of the guide except the required tests refer to Table 5.1 of ANSI N509-1976 rather than to Table 2 of the guide.

Discussion of problems and shortcomings of the test methods:

The problems and shortcomings associated with the testing methodology used in earlier versions of the regulatory guide have been discussed elsewhere (see for example EGG-CS-7653, "Final Technical Evaluation Report for the NRC/INEL Activated Carbon Testing Program", April 1987). The recommendation from this report resulted in a testing protocol that formed the basis for ASTM D3803-1989, "Standard Test method for Nuclear-Grade Activated Carbon". This became the test method recommended in NRC Generic Letter 99-02: Laboratory Testing of Nuclear-Grade Activated Charcoal, June 3, 1999 and incorporated in Regulatory Guide 1.52 Revision 3, June 2001.

Regulatory Guide 1.52 Revision 3, June 2001

New Carbon Tests:

Original and replacement batches of impregnated activated carbon must meet Section FF-5000 of ASME AG-1-1997.

Qualification Tests:

FF-5211:

This is a methyl iodide removal efficiency test performed at 80°C and 95% RH according to ASTM D3803-1989. An acceptable result is not less than 99.0%

FF-5212:

This is an elemental iodine removal efficiency test performed at 30°C and 95% RH according to ASTM D-3803-1989. An acceptable result is not less than 99.9%.

FF-5213:

This is a methyl iodide removal efficiency test performed at 130°C and 95% RH according to ASTM D3803-1989. An acceptable result is not less than 98%.

There are a couple things wrong with this testing methodology. First, ASTM D3803-1989 ***is only a 30°C and 95% RH methyl iodide removal test.*** One could make a good argument for use of the 30°C, 95% RH elemental iodine test using this procedure since changing the challenge agent should not make a difference. The same cannot be said for the higher temperatures, although one might assume that if the tolerances for the parameters were maintained for the higher temperatures then the results should be acceptable. But this is just an assumption and has not been verified. Some practical problems also arise when one considers holding the temperature to ± 0.2 °C at 130 °C. Secondly, the highest temperature test is considered the most difficult, (has the lowest efficiency) when it is common knowledge that just the opposite is true.

Batch Tests:

FF-5221:

This is a methyl iodide removal efficiency test performed at 30°C and 95% RH in accordance to ASTM D3803-1989. A minimum acceptable result is 97%.

FF-5222:

This an elemental iodine retention efficiency test performed at 180°C in accordance with ASTM D3803-1989. A minimum acceptable result is 99.5%.

Unfortunately, ***there is no guidance whatsoever*** in ASTM D3803-1989 for performing **FF-5222** (the elemental iodine retention test).

Used carbon tests:

ASTM D3803-1989:

This is a methyl iodide removal efficiency test performed at 30°C and 95% RH or at 70% RH if the system has humidity control. The acceptance criterion is a methyl iodide penetration less than or equal to 2.5%

It's worth noting that the used carbon's allowable penetration is more stringent than the new carbon's.

Summary of Test Results Since June 1999

The average efficiency for 500 ASTM D3803-1989 methyl iodide removal efficiency tests performed on 2 inch beds at 30°C and 95 % RH since June of 1999 by NUCON's laboratory was 96.7 %. The minimum efficiency was 36.18% for a sample that had always been tested at 130°C and 95% RH. During the same time period, the average removal efficiency for 352 tests performed on 2 inch beds tested at 30°C and 70% RH was 98.75% with a minimum efficiency of 54.73%. These results indicate that carbon beds are not being replaced unnecessarily as a result of adopting D3803-1989 as the test method.

Discussion and Recommendation

The process of using consensus standards to form the basis of new regulatory guidance has worked successfully in the past and should continue to do so in the future. Although the argument can always be made that this process puts current practice behind the "state of the art", interested parties are assured of the opportunity to be heard at any time during the evolution of test methods and requirements. For the first time in my experience, the regulatory guide is ahead of the AG-1-code for the testing of nuclear grade activated carbon. Hopefully, the code will soon be revised to reflect current practice.

Table 1 Regulatory Guide1.52 Carbon Test Requirements

USNRC Regulatory Guide 1.52 Rev. 0-1973 (RDT M16-1T, June 1972)

Table 1 New Carbon

Test	Temp °C	RH %	Bed Depth inches	Pressure kPa	Velocity m/min	Pre-Equil hours	Equil hours	Challenge minutes	Elution minutes	Agent	Conc. mg/m3	Efficiency %	Notes
5a	130	95	2	394.5	12.2	0	2	90	90	I_2	17.5	99.9	Qual
5b@95%	130	95	2	394.5	12.2	0	2	90	90	CH_3I	1.75	95.0	Batch
5b@70%	130	70	2	394.5	12.2	0	2	90	90	CH_3I	1.75	99.5	Batch
5c	25-180	70	1	101.3	12.2	0	0	120	240	I_2	75	99.0	Qual

Table 2 Used Carbon

5a	130	95	2	394.5	12.2	0	2	90	90	I_2	17.5	99.9	inside
5b	130	95	2	394.5	12.2	0	2	90	90	CH_3I	1.75	90.0	inside
5b	130	70	2	394.5	12.2	0	2	90	90	CH_3I	1.75	99.0	outside
5b	130	70	4, 6, 8, 15	394.5	12.2	0	2	90	90	CH_3I	1.75	99.8	outside

Table 1, continued:

USNRC Regulatory Guide 1.52 Rev. 1-1976 (RDT M16-1T, Oct 1973)

Table 2 New Carbon

Test	Temp °C	RH %	Bed Depth inches	Pressure kPa	Velocity m/min	Pre-Equil hours	Equil hours	Challenge minutes	Elution minutes	Agent	Conc. mg/m3	Efficiency %	Notes
5a	25	95	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	Qual
5b	25-80-25	95	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	Batch
5c	130	95	2	374.8	12.2	0	2 (≤ 1°C)	120	120	CH_3I	1.75	98.0	Qual
5d	25-180	n/a	1	101.3	12.2	0	0	10	240	I_2	75	99.9-99.0	Batch

Table 3 Used Carbon

5c	130	95	2	374.8	12.2	0	2 (≤ 1°C)	120	120	CH_3I	1.75	90.0	inside
5b	25-80-25	70	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	outside
5b	25-80-25	70	4, +	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.8	outside

Table 1, continued:

USNRC Regulatory Guide 1.52 Rev. 2-1978
(ANSI N509-1976)

Section 6a.2 New Carbon

Test	Temp °C	RH %	Bed Depth inches	Pressure kPa	Velocity m/min	Pre-Equil hours	Equil hours	Challenge minutes	Elution minutes	Agent	Conc. mg/m3	Efficiency %	Notes
5a	25	95	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	Qual
5b	25-80-25	95	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	Batch
5c	130	95	2	374.8	12.2	0	2 (≤ 1°C)	120	120	CH_3I	1.75	98.0	Qual
5d	25-180	n/a	1	101.3	12.2	0	0	10	240	I_2	75	99.9-99.0	Batch

Table 2 Used Carbon

5c	130	95	2	374.8	12.2	0	2 (≤ 1°C)	120	120	CH_3I	1.75	90.0	inside
5b	25-80-25	70	2	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.0	outside
5b	25-80-25	70	4, +	101.3	12.2	0	16 (≤ 1°C)	120	120	CH_3I	1.75	99.8	outside

Table 1, continued:

USNRC Regulatory Guide 1.52
Rev. 3-2001

New Carbon Section 4.11 (ASME AG-1-1997 *AG-1 does not provide guidance for parameters as tested IAW D3803-89*)

Test	Temp °C	RH %	Bed Depth inches	Pressure kPa	Velocity m/min	Pre-Equil hours	Equil hours	Challenge minutes	Elution minutes	Agent	Conc. mg/m3	Efficiency %	Notes
FF-5211	80	95	2	101.3	12.2	16	2	60	60	CH_3I	1.75	99.0	Qual
FF-5212	30	95	2	101.3	12.2	16	2	60	60	I_2	17.5	99.9	Qual
FF-5213	130	95	2	101.3	12.2	16	2	60	60	CH_3I	1.75	98.0	Qual
FF-5221	30	95	2	101.3	12.2	16	2	60	60	CH_3I	1.75	97.0	Batch
FF-5222	30-180	95	2	101.3	12.2	16	2	60	60	I_2	75	99.5	Batch

Used Carbon Table 1 (D3803-1989) *RG 1.52 Rev. 3 does not provide guidance for Elemental Iodine test parameters*

	30	95	2	101.3	12.2	16	2	60	60	I_2	1.75	97.5	
	30	95	2	101.3	12.2	16	2	60	60	CH_3I	1.75	97.5	
	30	95	4, +	101.3	12.2	16	2	60	60	I_2	1.75	99.5	
	30	95	4, +	101.3	12.2	16	2	60	60	CH_3I	1.75	99.5	

Reference List

1. American Society for Testing and Materials, "Standard Test Method for Nuclear-Grade Activated Carbon", ASTM Designation: D3803-1989. Reapproved 1995

2. American Society of Mechanical Engineers, "Nuclear Power Plant Air Cleaning Units and Components", ANSI/ASME N509-1976.

3. American Society of Mechanical Engineers, "Code on Nuclear Air and Gas Treatment," ASME AG-1-1997.

4. NRC Generic Letter 99-02, "Laboratory Testing of Nuclear-Grade Activated Charcoal," June3, 1999.
5. Scarpellino, C.D. and Sill, C.W., "Final Evaluation Report for the NRC/INEL Activated Carbon Testing Program", EGG-CS-7563, April 1987.

6. United States Atomic Energy Commission, "Gas-Phase Adsorbents for Trapping Radioactive iodine And Iodine Compounds", Standard RDT-M16-1T, June 1972 and October 1973.

7. United States Atomic Energy Commission, "Design Testing, and Maintenance Criteria for Atmospheric Cleanup System Air Filtration and Adsorption units of Light-Water-Cooled Nuclear Power Plants", Regulatory Guide 1.52, June 1973.

8. United States Nuclear Regulatory Commission, "Design, Testing, and Maintenance Criteria for Engineered-Safety-Feature Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants", Regulatory Guide 1.52, Revision 1, July 1976.

9. United States Nuclear Regulatory Commission, "Design, Testing, and Maintenance Criteria for Post Accident Engineered-Safety-Feature Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants", Regulatory Guide 1.52, Revision 2, July 1978.

10. United States Nuclear Regulatory Commission, "Design, Inspection, and Testing Criteria for Air Filtration and Adsorption Units of Post Accident Engineered-Safety-Feature Atmospheric Cleanup Systems in Light-Water-Cooled Nuclear Power Plants", Regulatory Guide 1.52, Revision 3, June 2001.