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Effect of Surface Modifications on the Performance of Nuclear Grade Activated Carbon for Removal of Methyl Iodide

By

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- ASTM D3803 shows that methyl iodide penetration increases as the moisture content of the carbon increases with %RH:

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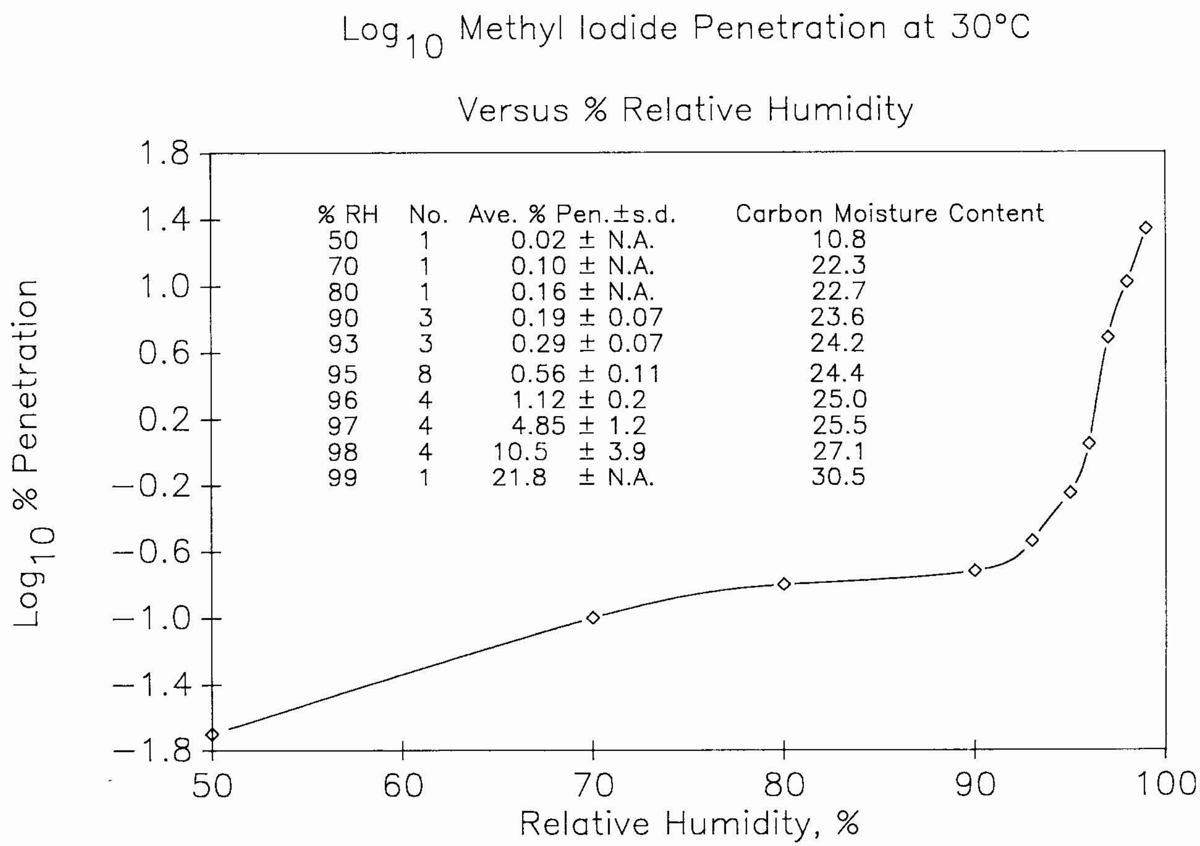

FIG. A1.1 Results of Sensitivity Testing of New, Co-impregnated, 2*N*Carbon Using the IC Protocol and Varying Only Relative Humidity

- It is interesting to note that if we could decrease the amount of moisture adsorbed at 95% RH to that of 90% RH we would:
- Decrease the % penetration from 0.56 to 0.19 (nearly 3 times improvement), while only having to:
- Decrease the moisture content from 24.4% to 23.6% (a 0.8% decrease)

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- If the surface of the activated carbon could be modified to make it a bit more hydrophobic then:
- The methyl iodide performance could be increased as determined by an ASTM D3803 test and perhaps
- Costs could be reduced by requiring less impregnant for the same performance?

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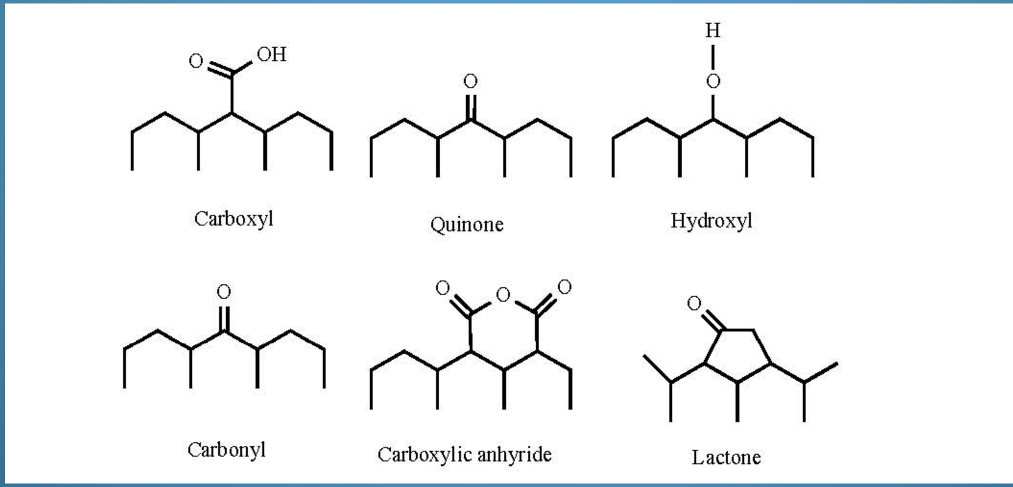
- The functional groups on the surface of the carbon can be modified to change it’s nature:



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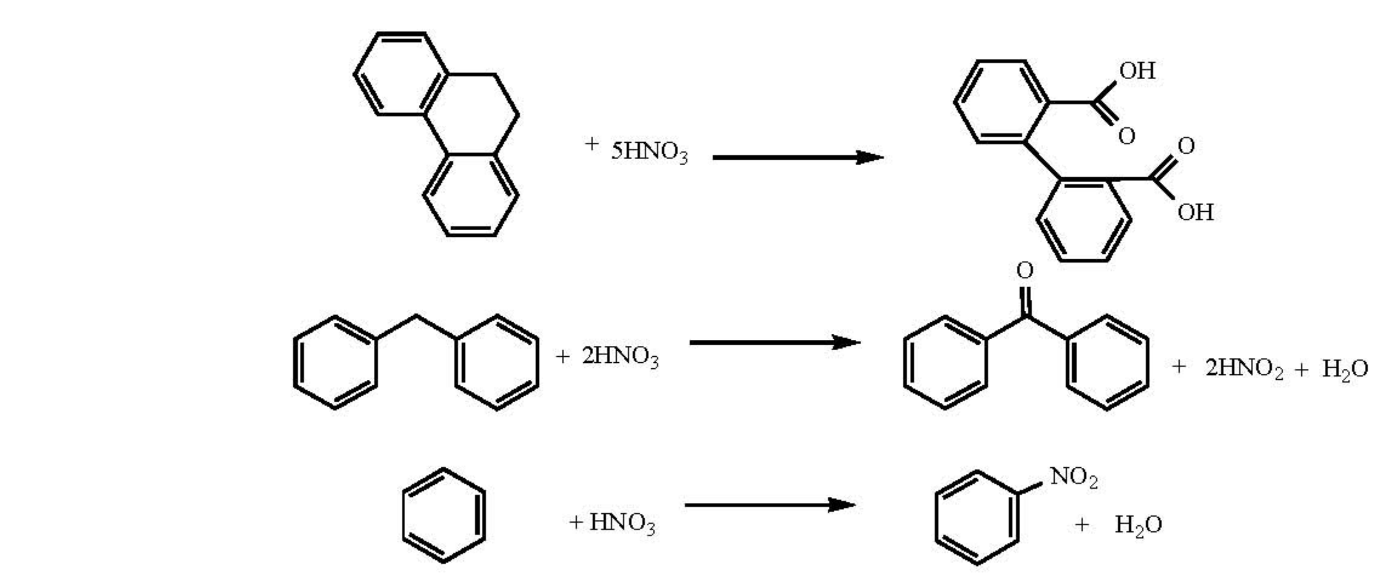
- The surface of the carbon can be modified chemically (HNO3, H2O2, O3 for example)



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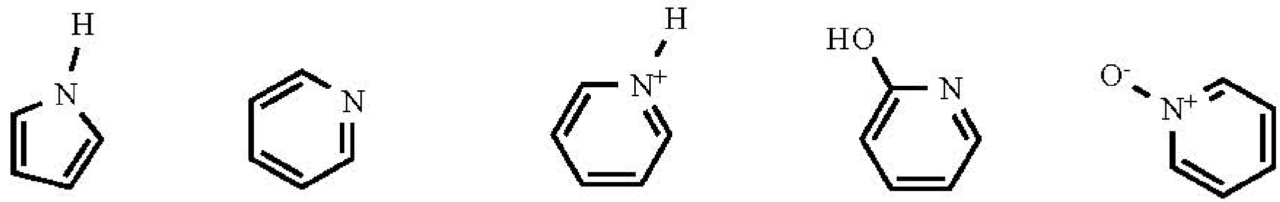
- The surface of the carbon can be modified by incorporation of nitrogen:



Pyrrole Pyridine Pyridinium Pyridone Pyridine-N-oxide

- The surface of the carbon can be modified by heat treatment in nitrogen or hydrogen
- High-temperature treatments remove oxygen from the active sites at the edges of graphite-like crystallites (graphene layers)
- The character of the residual carbon active sites formed depends on the gaseous atmosphere in which the treatment is carried out:

- Upon treatment with H_2 at 950 °C, activated carbon will adsorb very little oxygen at room temperature
- Heat treatment with N_2 and the activated carbon will adsorb as much as 20 times more O_2 under the same conditions

- To explain this difference in surface chemistry it has been postulated that treatment in H_2 not only removes oxygen (as CO and CO_2)
- But also removes (hydrogasifies) some of the very reactive residual carbon atoms (e.g., as CH_4)
- In contrast, treatment in an inert gas removes the oxygen but leaves behind many highly (re)-active carbon atoms.

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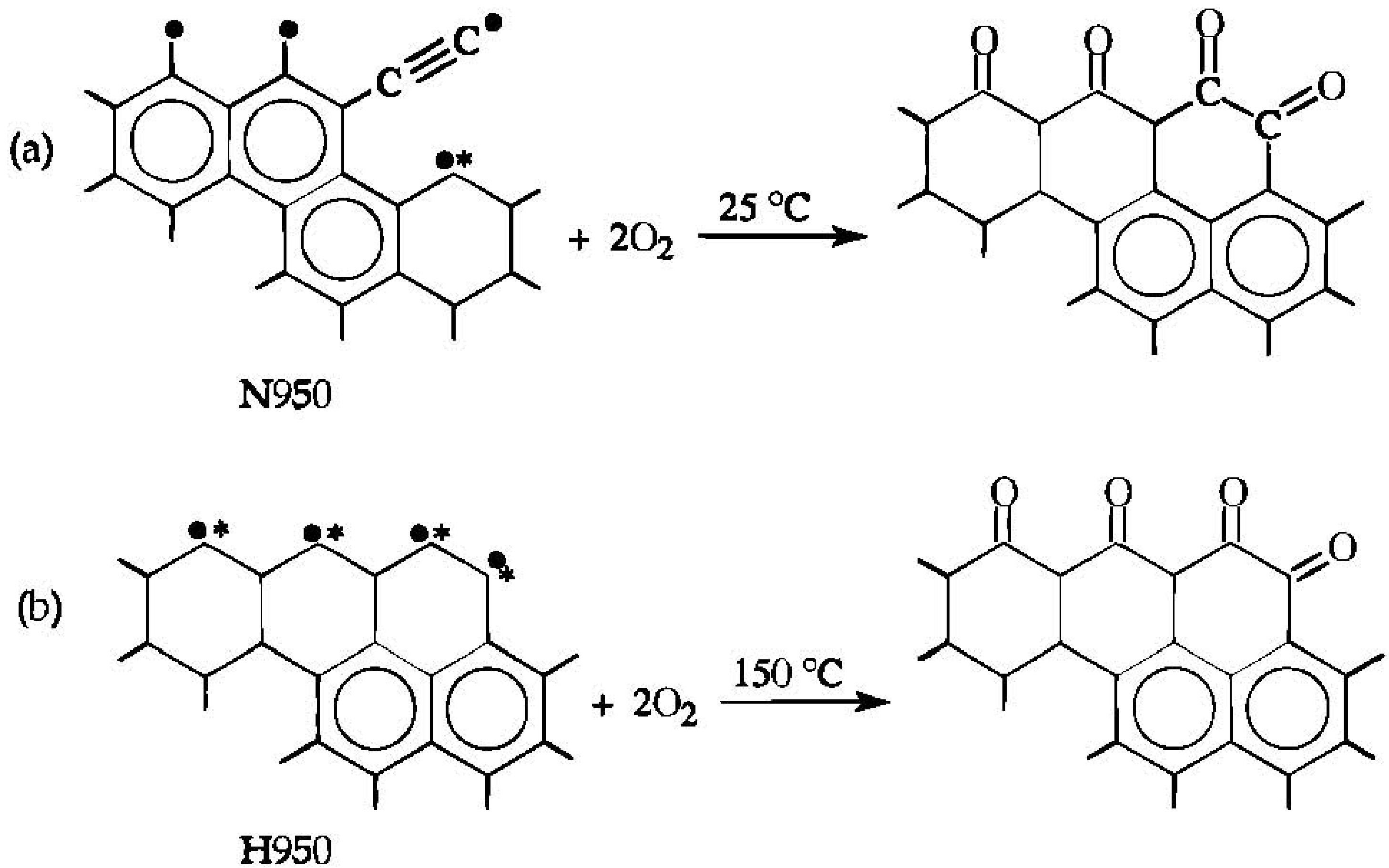

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Why is this important to us?

For a couple of reasons:

1. It is generally felt that oxygen surface complexes are responsible for the ageing effect of nuclear grade carbons
2. And these same oxygen complexes are hydrophilic and thus increase methyl iodide penetration when tested according to ASTM D3803

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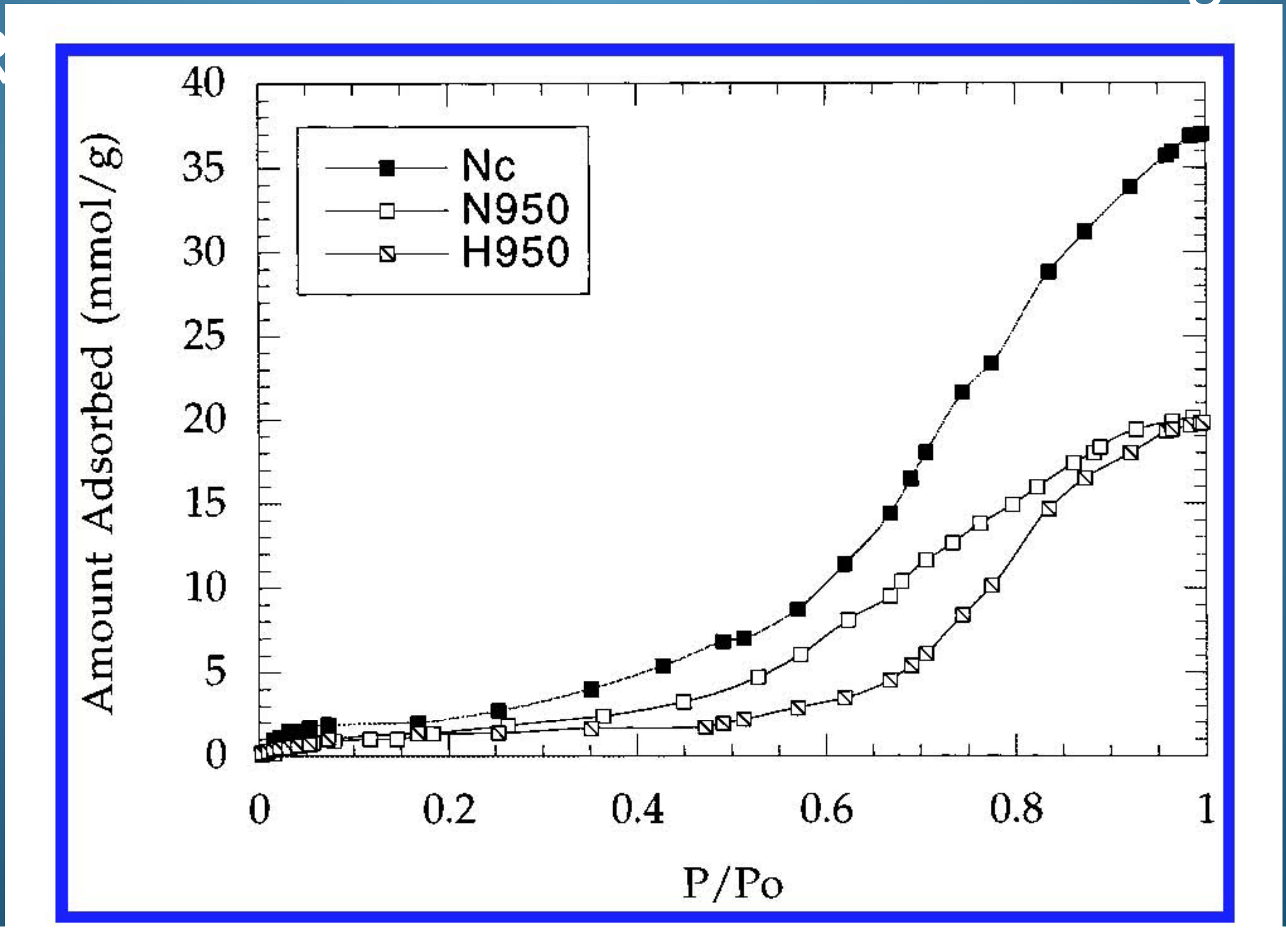
We can see that both H2 treated and N2 treated carbons adsorb less water at a given %R



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- We are especially interested in the reduction in moisture adsorbed at high %RH
- Others have found that the oxygen complexes remove by nitrogen heat treatment are quickly replaced by exposure to ambient air
- The hydrogen treated carbon appears stable in air even at 150 ^{o}C

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- To test this thinking the methyl iodide penetration was determined according to ASTM D3803 on a sample of AddSorb RC3-8x16, KI-TEDA co-impregnated carbon,
- A sample prepared from the same base carbon but oxidized with HNO_3 and then KI-TEDA impregnated
- And a sample prepared from the same base material heat treated with H_2

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- Results
- Untreated AddSorb RC3 - 8x16:
  - Methyl iodide penetration: 0.37%
  - Moisture content: 23.8%
- Oxidized AddSorb RC3 – 8x16:
  - Methyl iodide penetration: 10.1%
  - Moisture content: 28.7%
- H_2 treated AddSorb RC3- 8x16
  - Methyl iodide penetration: 0.10%
  - Moisture content: 22.0%

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- Conclusions:
- H_2 heat treatment shows promise as a way to improve the performance of nuclear grade carbons (an existing activation kiln operation can be modified to make this base in situ)
- This also has the potential to improve ageing characteristics
- N- incorporated carbons will be investigated in the future as well