HISTORY OF RADIOIODINE CONTROL

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Abstract

The release and immobilization methodologies of radioiodine in nuclear power facilities is reviewed. The evaluation is restricted to the release mechanisms and control technology of gaseous releases under normal operating conditions. The major released radioisotope forms are typically removed by special processes, depending on the chemical properties of the released iodine fission product form. The progression to the current understanding of these processes is described.

Radioiodine Control

Under normal operating conditions, radioiodine and other fission products are released from the fuel rods due to cladding failure and subsequent liquid leaks from the primary or in case of PWRs by primary to secondary leaks , steam generator tube ruptures and subsequent out leakage from the steam generator systems [1], [2]. There are numerous good descriptions of the cladding failure caused releases into the primary coolant in the literature, [3], [4], [5]. In the earlier days of nuclear power operations, the fuel rod maintained integrity was assumed to be 99 %. However, in the subsequent years improvements in fuel rod manufacturing and their quality assurance, this value has significantly increased. Typically, plant technical specifications in the US, limit the specific activity in the reactor coolant systems to < 1.0 microCi/g dose equivalent ^{131}I . The maximum short time, less than 48 hours duration, ^{131}I dose equivalent is 60 times this value. Recent studies indicate that these values are not exceeded in the reactor coolant even with iodine spiking due to power transients [6].

Radioiodine Forms

The halide releases from the fuel are typically in elemental form, but in some cases volatile halides, such as CsI may be also released into the primary coolant, where additional reactions may take place and acid halides (e.g., HI) may then be formed via reaction with the radiolytically generated hydrogen and hypoacids (e.g., HOI) via reaction with hydroxyl radicals. There are indications, that some of the inorganic iodine forms once released to the reactor coolant can react even with the very low level of organic impurities in the coolant as indicated by the presence of a large fraction of organic radioiodine in the steam jet air ejector of BWR condensers, [7]. Some of these inorganic iodine forms may be released from the coolant system via the above discussed leaks, or in some from the degassing systems of the primary coolant.

While elemental iodine and hydrogen iodide, at the concentrations present will be gaseous products, the hypoiodic acid can be airborne only as part of the water droplet, because it is not stable in a gaseous form. Many early evaluations of radioiodine control discuss the presence of HOI [8], [9], [10], [11], however, subsequent evaluations have determined that the life of the water free, truly gaseous HOI is in the millisecond range [12],[13],[13], [14]. In most cases, in the past, when air sampling methods identified iodine species as HOI, the actual form was either an organic iodine or possibly hydrogen iodide. In general, it is conservative to "convert" reported HOI form to organic iodine form rather than into HI form, when dealing with airborne iodine measurement results from earlier literature. When the inorganic radioiodine forms are released into the containment, reactor building and other environments, additional chemical reactions can and do take place between the inorganic iodine forms and the organic compounds present in those areas, creating a large fraction of organic iodides.

Under normal operating conditions, when the concentration of the radioiodine forms is low, the available organic reactant ratio is very high, and in many cases, the bulk of the normally released originally inorganic iodine can convert to organic forms. While the organic iodide is commonly assumed to be methyl iodide, other organic forms are also produced. The type of organic compounds formed are dependent on the airborne organic reactants being present or on the type or organic solids on which inorganic iodine forms may initially deposit (paints, coatings, electrical insulation, etc.) and subsequently react. In case of off-normal operation releases the potential exists for a much larger amount of radioiodine to be released from the primary system, in that case the organic reactant to iodine ratio is lower and a larger fraction of the radioiodine forms stay in the inorganic form.

Organic iodine forms other than methyl iodide have been identified e.g., longer alkyl chain (ethyl, propyl) iodides, methyl di- and tri- iodides, allene iodide, etc. Both the method of deposition and the subsequent removal mechanism for the various iodine forms are different [17].

Radioiodine can also be present as a neat aerosol (such as CsI) or in adsorbed or absorbed form on existing solid or liquid aerosols. The iodine forms adsorbed or absorbed on aerosols may be in a transient stage, i.e. the adsorbed molecule can at a later time desorb from the solid surface and the liquid substrate in which the iodine absorbed can evaporate at which time the initially particulate radioiodine can reconvert to a gaseous form. This type of transition behavior affects conclusions drawn from short term filtration/ adsorption tests, where the initially mechanically filtered out radioactivity can become airborne after some residence time on the particulate filter.

The main grouping of radioiodine forms are:

a) elemental iodine
b) hydrogen iodide
c) organic iodides
d) iodine salt aerosols
e) the above four forms adsorbed or absorbed on aerosols.

The distribution of these iodine forms is dependent on the environment where the original release from the primary or secondary systems took place. The distribution between the various forms can be significantly different during normal and refueling operations, due to the typical presence of a much larger amounts of maintenance related organic compounds being present in the containment during refueling operations. The airborne iodine form distribution is also different for large accidental releases which are not discussed in detail in this paper.

Airborne Iodine Concentrations

The total iodine concentration in all cases will be a sum of the normal background stable iodine concentrations and the both stable and radioiodine forms being released from the fuel. Atmospheric iodine concentrations are reported in the various literature to be as follows:

Particulate or attached to particulate forms: 0.5 to 5.0 x 10E-9 g/m^3

Gaseous phase : 0.1 to 1.0 X 10E -8 g/m^3

Therefore when non - isotope - specific removal mechanisms are used for iodine control, the presence of the background stable iodine and the simultaneous to the radioactive, stable iodine released concentration also needs to be considered . Even in some presumed chemical removal reactions, such as the methyl iodide quarternary salt formation by reaction with tertiary amines, the compared to the radioactive species much larger quantity, the initial deposition of the larger quantity of stable iodine forms will have an effect by converting the initial chemical reaction removal to the isotope exchange type removal processes or exposure ambient air which contains stable iodine forms would perform in a an identical manner.

Table I, shows an iodine form distribution from an early analytical effort at a US BWR, Oyster Creek.

The previously published results have been modified here by eliminating the HOI form and adding the quantities previously identified as HOI to the organic iodide column. This may be only partially appropriate because some of the iodine may have been in HI form also.

Table 1. Iodine form distribution.

Location	Particulate	Elemental	Organic
Reactor Building	9	28	62
Radwaste building	*	16	84
Turbine building	17	43	40
Steam jet air ejector	*	<1	99
Gland seal and mechanical vacuum pump exhaust	*	8	92
Feedwater and condensate pump area	8	57	35
Reheater protection system and lubrication oil storage area	5	15	80

- The sampling took place downstream of a HEPA filter, thus only volatile species distribution is made. The particulate or attached to particulate forms have not been measured.
- The original tabulation is from ref [7] and [16].

The high proportion of organic radioiodine forms under normal operating conditions, as indicated by the above tabulation, should be expected, due to high concentration of background organic contaminant to iodine form ratio. As the total released iodine concentration increases, as is the case in off normal events, the ratio of inorganic to organic iodine forms will increase.

Radioiodine Removal Mechanisms

The various airborne iodine fission products can be removed and immobilized by several different process mechanisms. In most cases, the removal mechanisms have to accommodate all of the potentially existing forms simultaneously and often a compromise needs to be made when optimizing the selected removal process conditions for the required treatment of all of the airborne radioiodine forms.

Elemental iodine is generally removed by a physical adsorption process, which is surface area dependent and follows the classical surface adsorption based rate controlling steps, which are:

a) Diffusion to the adsorbent surface
b) Pore diffusion
c) Adsorption on the surface

In some cases, depending on the type of adsorbent used. there may be an additional step, i.e.,

d) Chemical reaction on the surface.

It is important to understand, that in the absence of the latter, the physical adsorption is a reversible process and the adsorbed iodine will be at an equilibrium with its gaseous concentration. In a dynamic system the elemental iodine adsorbed on the inlet of the adsorbent bed will , progress through the adsorbent bed by a series of adsorption/desorption (as in gas chromatography) steps. Depending on the rate of transport, some of the ^{131}I may decay to gaseous xenon and because the xenon is more weakly adsorbed, and proceed faster through the adsorbent bed , the decayed iodine isotope location can be "regenerated" for additional ^{131}I adsorption. This type of ,decay based, regeneration results in the decrease in the rate of the transport of the radioiodine in an adsorbent bed, but the rate of transport of any stable iodine which is also present does not change.

In the matrix of some iodine adsorbents, chemicals capable of reacting with the adsorbed iodine, are naturally present (e. g., alkali salts in vegetable based carbons) or are added on purpose (impregnated adsorbents) to convert the elemental iodine to a salt which is a form of immobilization of the iodine subsequent to adsorption on the surface. The iodine which reacts to form a salt on the adsorbent surface is no longer retained by physical adsorption on the surface and the adsorption principals of transport and retention apply only in attracting the iodine to the surface of the adsorbent

However, when both physical adsorption and chemical reaction on the adsorbent surface are used to immobilize the elemental iodine, the rate controlling parameters can and do conflict. As an example, increased temperature results in decreased adsorption capacity (shorter adsorbed stage residence time on the surface), although not in decreased adsorption rate, while the chemical reaction rate significantly increases with increased temperature. As an example the natural alkalinity in nutshell based carbons have been found successful in removing iodine even at 1000 C from a helium stream. While the large surface area of the carbon does adsorb the elemental iodine even at this high temperature, if physical adsorption was the only process, there would be only infinitesimal adsorption capacity, but the alkalinity present on the surface of the coconut shell carbon is capable of converting the elemental iodine to the iodide form and retaining it on the surface not in a per se adsorbed but in a reacted salt form.

Very small quantities of potential reactants for the elemental iodine, have a significant effect on the retention of the adsorbed elemental iodine even under ambient temperature conditions. Therefore, it is very rare that physical adsorption alone is the process involved in elemental iodine form immobilization.

In most environments, where iodine immobilization is needed, there are other compounds which are also adsorbed on the adsorbent surfaces. Depending on the nature of these compounds the adsorption rate and or the adsorption capacity may be lowered for radioiodine. The most common "other" chemical present in these radioactive iodine vapor processing environments is water, although various trace, volatile organic compounds are also typically present. The co-adsorption of water can strongly affect the adsorption of the elemental iodine, based on the adsorbent type, because the shape of the water adsorption isotherm (adsorption capacity versus water concentration, i. e. relative humidity) is different for different matrix adsorbents.

In carbon based adsorbents, there are two sharp breakpoints in water adsorption capacity, one at about 50 % RH and another at about 95 % RH. In other, mineral based, adsorbents the water capacity breakpoints are at different RH conditions. The presence of adsorbed water will lower both the adsorption rate and the adsorption capacity of the adsorbent. The adsorption rate is lowered because there are additional rate controlling steps (e. g. dissolution of the iodine in the adsorbed water layer) and the capacity is lowered because some of the surface sites are occupied by water molecules. However, in cases where, in addition to physical adsorption, a chemical reaction on the surface is needed to immobilize the elemental iodine,, the presence of some adsorbed water can enhance the on - the - surface chemical reaction steps.

The adsorption and accumulation of organic materials on adsorbent surfaces can be of a transient nature. Organic molecules, which are only weakly adsorbed (typically higher volatility), may pass through an adsorbent bed reasonably fast, while larger molecular weight (typically lower volatility) compounds will be more strongly adsorbed and retained on the adsorbent surface for a longer period of time. The time of retention and its effect is dependent on the operating conditions and the depth of the adsorbent bed as shown on Figure 1 from Ref. [18].

Long term retention of organic compounds can have another detrimental effect on the overall radioiodine control, because the adsorbed elemental iodine can convert to less strongly adsorbed organic forms on the adsorbent surface and pass through the adsorbent bed at a faster rate than unreacted elemental iodine.

Early iodine control technology considered the only hazard to be from the elemental iodine form and therefore "unimpregnated" carbon adsorbents were used for radioiodine control [19], [20, [21], [22]. The control and immobilization of elemental iodine was successfully accomplished by a 25 mm deep carbon bed operated at 40 cm/second velocity through the adsorbent bed, i. e. an ~ 0.06 second residence time.

In the early 1960s, it was found that, particularly at low total iodine concentrations, a large fraction of the total iodine may be present in less strongly adsorbed non-elemental forms. The most common and largest organic fraction was identified as methyl iodide. Methyl iodide is adsorbed much more weakly than elemental iodine and its adsorption capacity on all adsorbents is significantly reduced under high humidity conditions.

A large variety of adsorbents were evaluated unsuccessfully, to find a higher adsorption capacity adsorbent for methyl iodide which would have reasonable adsorption capacity at high relative humidity.

None of the adsorbents, tested to date, have the required physical adsorption capacity for methyl iodide to be acceptable as a control step. Evaluation of the various modified adsorbents resulted in two type of treated adsorbents which were capable of controlling radioactive methyl iodide, those which decontaminate the radioactive methyl iodide by isotope exchange and those which are capable of initially chemically reacting with adsorbed methyl iodide generally leading to a combined reaction and isotope exchange process, [22], [23], [24], [25].

The isotope exchange process consists of depositing stable iodine or iodine compounds on the carbon surface, which as long as present in a significant excess is capable of decontaminating the radioactive methyl iodide by isotopic exchange:

$$CH_3{}^{131}I \quad + \quad K\,{}^{127}I \quad \leftrightarrow \quad K\,{}^{131}I \; + \; CH_3\,{}^{127}I$$

gas phase surface surface gas phase

It is important to note that the isotope exchange process is a reversible process and a significant excess of the stable iodine form is required for the process to be highly efficient. This type of organic iodide removal process consists of significantly larger number of steps than the physical adsorption process alone:

a) Diffusion of active form to the grain surface
b) Diffusion of active form into the pores
c) Physical adsorption of active form
d) Isotopic exchange
e) Desorption of inactive form from the surface
f) Diffusion of stable form from pores
g) Diffusion of stable form into the gas phase

Generally, at velocities below about 40 cm/sec, the rate controlling steps are the bulk diffusion steps i. e. steps a and g, while at higher than 40 cm/sec velocity through the adsorbent,bed pore diffusion becomes tha rate controlling step (steps b and f). However, the change in rate controlling step is dependent on other parameters also such as adsorbent particle size, temperature, adsorbent type, etc., [27], [28], [29], [30].

The gas velocity at which the change from laminar to turbulent flow change occurs makes the often practiced "normalization" of the methyl iodide removal efficiency to constant residence time (retention index) questionable covering rate controlling step transition ranges.

The isotope exchange process, while capable of removing methyl iodide at a much higher efficiency than physical adsorption alone is still strongly affected both by the coadsorbed water and coadsorbed other organic compounds. The dependence on relative humidity is indicated by Figure 2 and Table 2 from Ref. [31].

Table 2. The effect of relative humidity on methyl iodide removal for a 50 mm bed operated at 20 cm/sec.

Relative humidity %	Water adsorbed wt %	Penetration of $CH_3{}^{131}I$ % (100 - Efficiency)
50	10.8	0.02 +/- NA
70	22.3	0.10 +/- NA
80	22.7	0.16 +/- NA
90	23.6	0.19 +/- 0.07
93	24.2	0.29 +/- 0.07
95	24.4	0.56 +/- 0.11
96	25.0	1.12 +/- 0.2
97	25.5	4.85 +/- 1.2
98	27.1	10.5 +/- 3.9
99	30.5	21.8 +/- NA

The data indicates that at about 95 % relative humidity, the adsorbed water starts to fill the pores of the adsorbent, and therefore significantly reducing the available surface area of the adsorbent for the isotope exchange step to take place. Above 95 % relative humidity, it is also becoming difficult to control the relative humidity within the system and the reproducibility of the measurement begins to suffer, [31]

Another common process is to use both chemical reactivity and isotope exchange by impregnating adsorbents with the tertiary amine triethylene diamine. This compound is capable of complexing with methyl iodide to form a quaternary salt, [32], [33], [34]. However, it has to be noted that in most cases, particularly in low concentration non-accident conditions, there is a significant chemical form abundance of the non radioactive iodine form and, on exposure to air, a large fraction of the tertiary amine will convert to the stable iodine form quaternary salt and the later stages of the methyl iodide decontamination process will take place by isotope exchange between a non-radioactive iodine quaternary salt and the radioactive methyl iodide, [28], [35], [36].

The rate of isotope exchange processes, analogous to chemical reaction on the surface will be directly proportional to temperature. Thus, while the physical adsorption capacity is inversely proportional to temperature, the isotope exchange (and other chemical reaction steps) are enhanced at higher temperatures, as long as the reactant or stable exchange material is retained on the adsorbent at the operating temperatures. The variation in short term removal efficiency of methyl iodide with temperature for different type of adsorbents and operating conditions is shown on Figure 3 and Figure 4 from Ref. [27] and [29].

The operating velocity versus efficiency at identical residence times can be different as shown on Figure 5 from Ref. [29]. This variation occurs because of changes in the carrier gas velocity affect the rate controlling steps of methyl iodide removal. A similar relationship shown on Figure 6 from Ref. [27], where the assumed velocity independent Performance Index is used. The plot clearly indicates the break in efficiency, in the ~ 40 cm/sec velocity range, which proves that the relationship between methyl iodide removal efficiency and residence time is not linear. As discussed above, the changes in rate controlling steps and the magnitude of the change is also strongly dependent on the particle size of the adsorbent. Adsorbent particle size effects are shown on Figure 7 from Ref. [29]. Reduction in particle size affects both the bulk and the pore diffusion steps and therefore can be a very significant cause of an increase in the methyl iodide removal efficiency under otherwise similar test conditions, [29].

The control of larger molecular weight iodides is a more complex process. While these compounds are less prevalent than methyl iodide, their presence has been identified in minute quantities and their typically lower long term removal efficiency may be one explanation for, the mentioned in the literature, of "more penetrating" iodine forms. (In some cases adsorbent test geometry and possible settling in the adsorbent bed in horizontal test beds may also be a cause of a constant rate, low level, penetration.) The larger molecular weight organic iodides are more strongly physically adsorbed than methyl iodide, but their isotope exchange rate is significantly slower and the combined effect can be good short term but diminished long term removal efficiency,[17], [37], [38].

The control and immobilization of the volatile HI form is the least studied among the compounds needing control. Generally, the low concentration of airborne HI occurs by chemical reaction between the HI and the alkalinity of the adsorbent surface. However, if the adsorbent alkalinity is not controlled, the physical adsorption capacity being a reversible process, faster penetration than other iodine forms may occur, but in this case, the adsorbed water can enhance retention.

Formulas Used in Expressing Radioiodine Removal

To enable any comparative evaluation of test data, it is important to have the following information with tolerances if applicable regarding any adsorbent evaluation:

a) Challenge agent type
b) Challenge agent concentration
c) Bed depth
d) Carrier gas velocity
e) Temperature
f) Pressure
g) Adsorbent particle size
h) Carrier gas relative humidity under test conditions
i) Duration of pre-equilibration with carrier gas at indicated relative humidity
j) Duration of loading
k) Duration of any post loading gas sweep
l) Adsorbent type
m) History of the adsorbent, if used

Typically, the following are used to express the performance of the adsorbent:

Removal efficiency, (%):

The fraction of the challenge agent radioactivity retained, expressed as the percent of the introduced total activity.

Penetration, (%):

100 - removal efficiency.

Decontamination Factor, (DF):

100/Penetration %

Performance Index, (K), sE-1 :

log DF/residence time in seconds

Caution has to be used in all cases in the extrapolation of any of the above test removal quantities to conditions other than the parameters under which the test has been performed.

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Figure 1

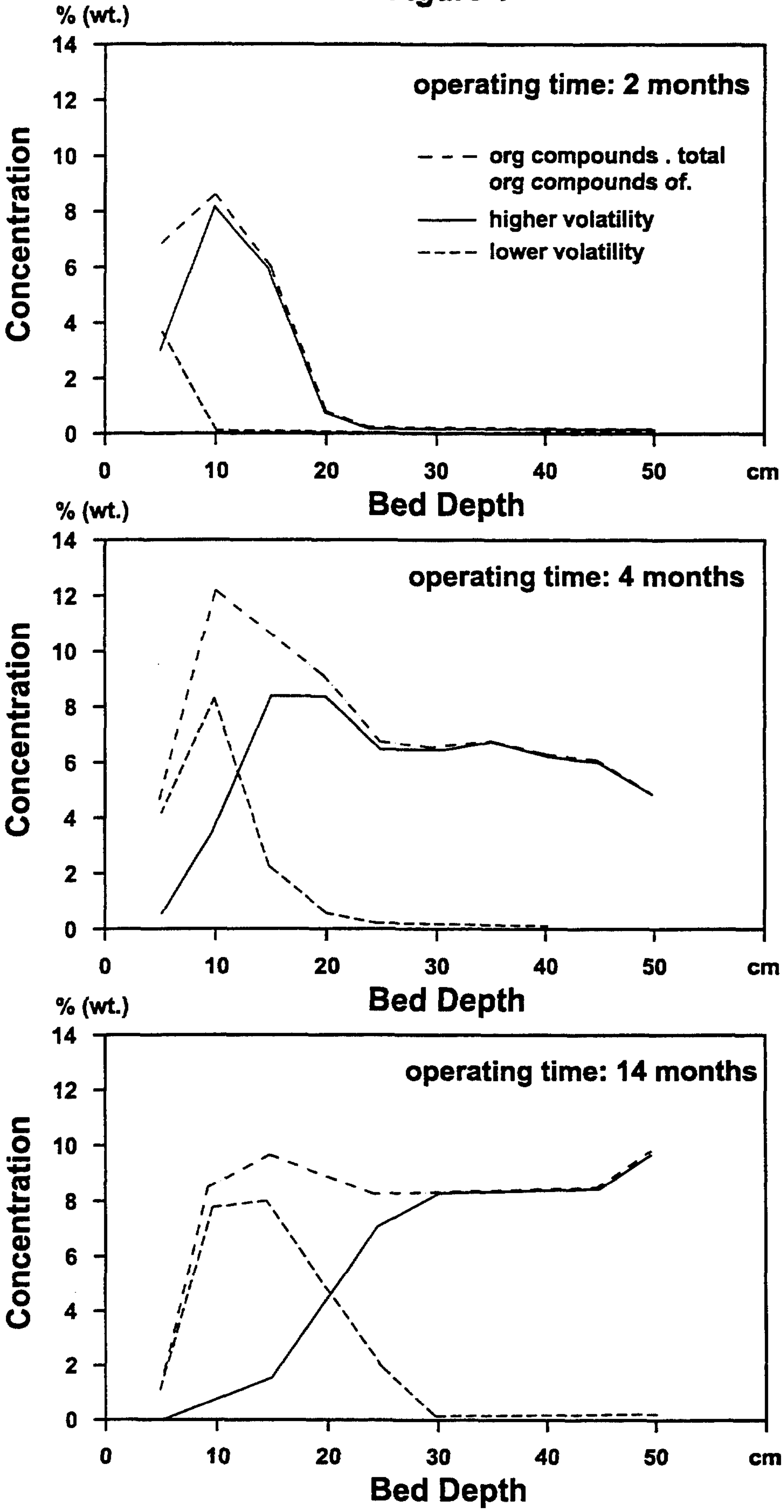
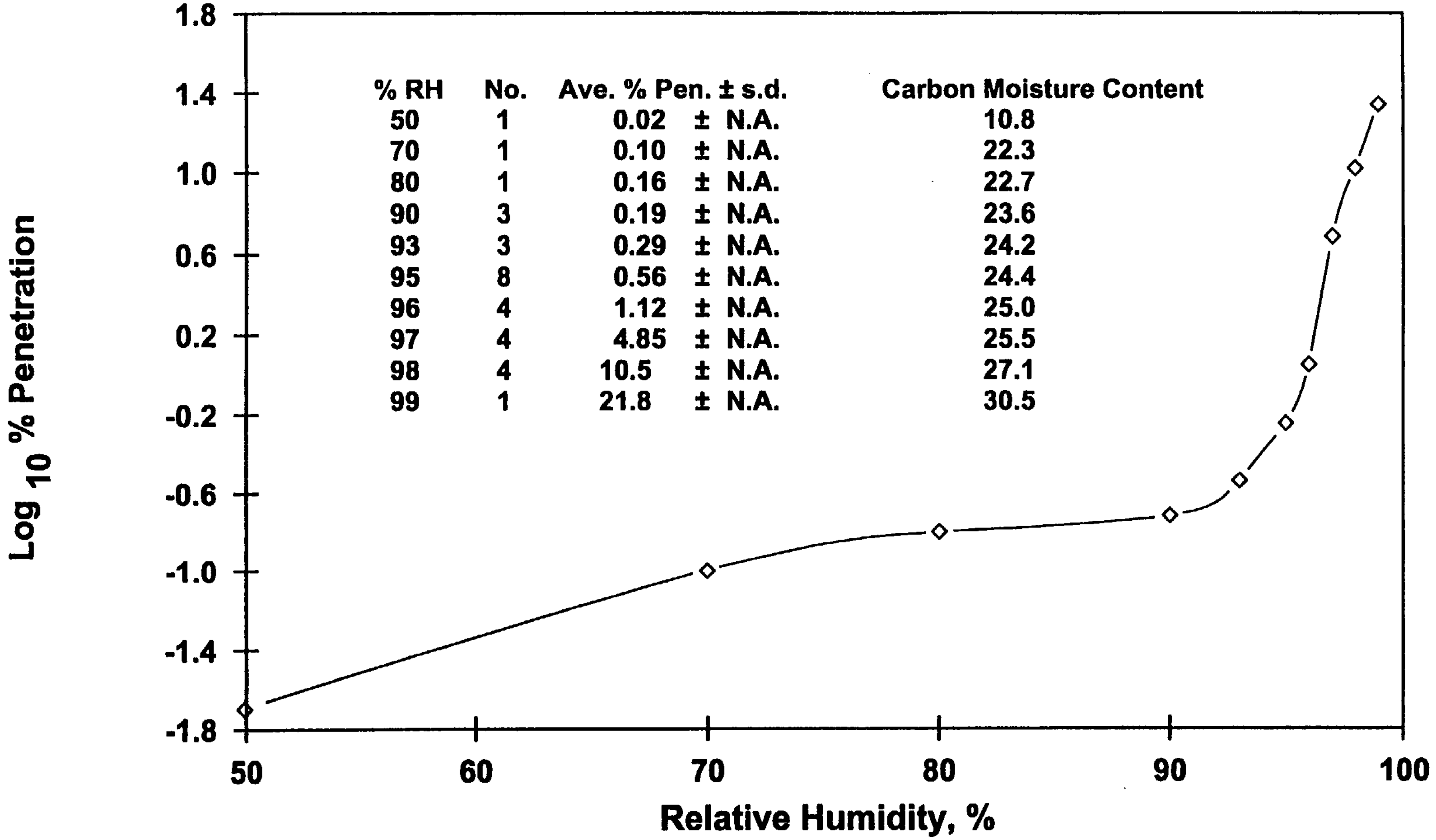

% (wt.)
Concentration
14
12
10
8
6
4
2
0
operating time: 2 months
org compounds . total
org compounds of.
higher volatility
lower volatility
0
10
20
30
40
50
cm
Bed Depth
operating time: 4 months
operating time: 14 months

Figure 2

Log_{10} Methyl Iodine Penetration at 30° C
Versus % Relative Humidity

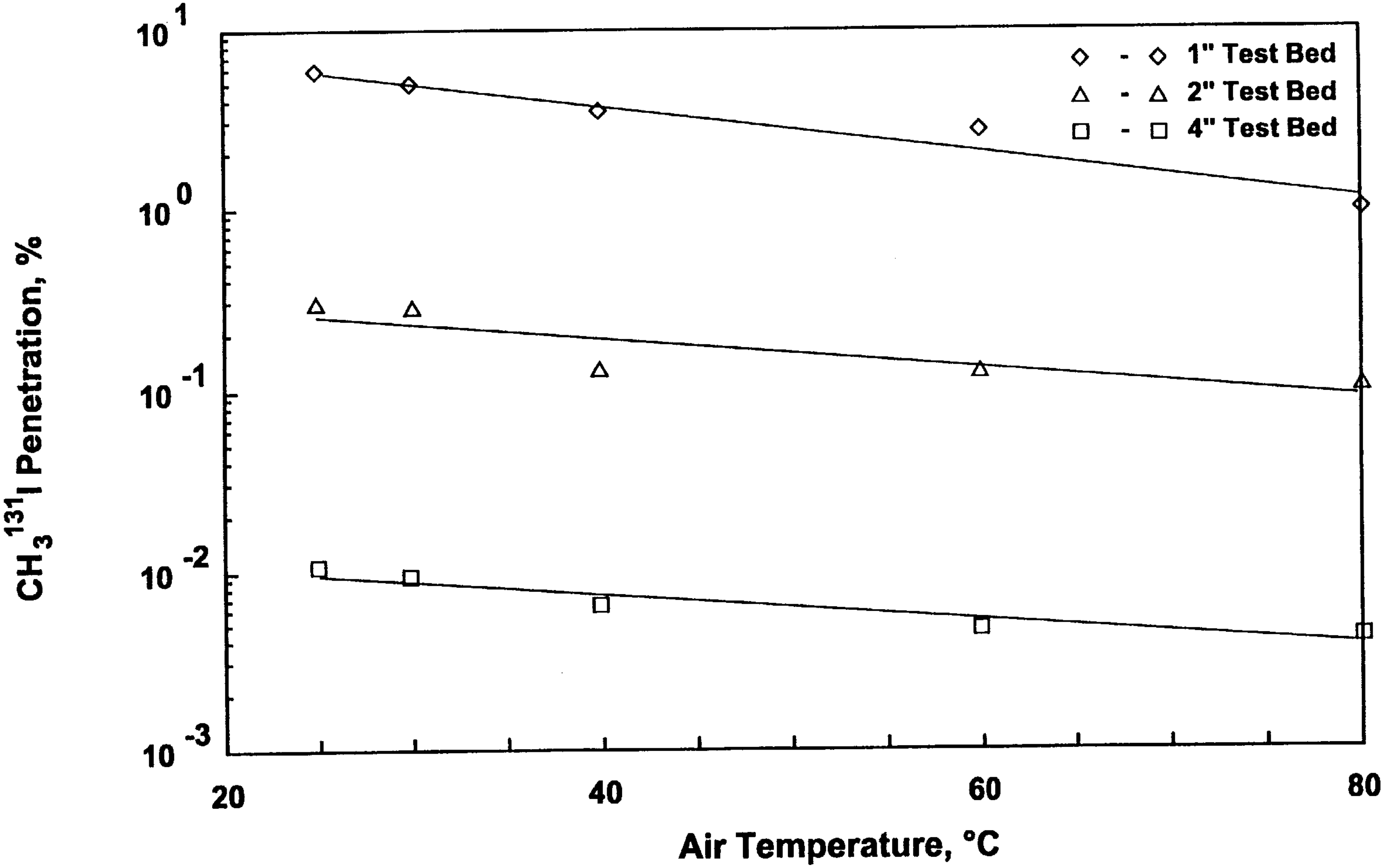


% RH	No.	Ave. % Pen. ± s.d.	Carbon Moisture Content
50	1	0.02 ± N.A.	10.8
70	1	0.10 ± N.A.	22.3
80	1	0.16 ± N.A.	22.7
90	3	0.19 ± N.A.	23.6
93	3	0.29 ± N.A.	24.2
95	8	0.56 ± N.A.	24.4
96	4	1.12 ± N.A.	25.0
97	4	4.85 ± N.A.	25.5
98	4	10.5 ± N.A.	27.1
99	1	21.8 ± N.A.	30.5

Results of Sensitivity Testing of New, Co-Impregnated, 2N Carbon Using the IC Protocol and Varying Only Relative Humidity

FIGURE 3

PARAMETRIC STUDY OF TEMPERATURE EFFECTS ON $CH_3{}^{131}I$ PENETRATION AT 95%, 40 FPM



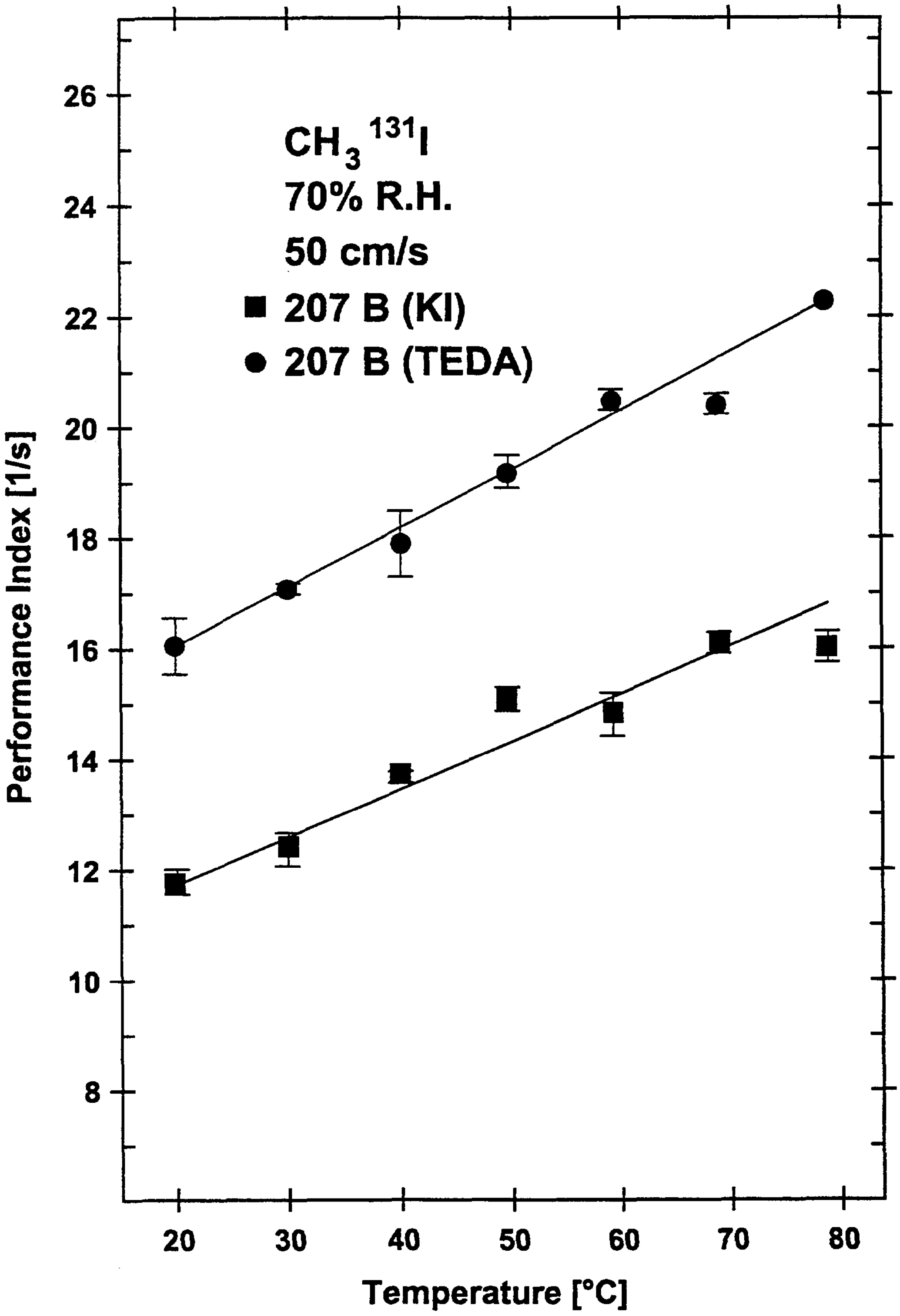


Figure 4. Performance index as a function of temperature with different impregnated activated carbons.

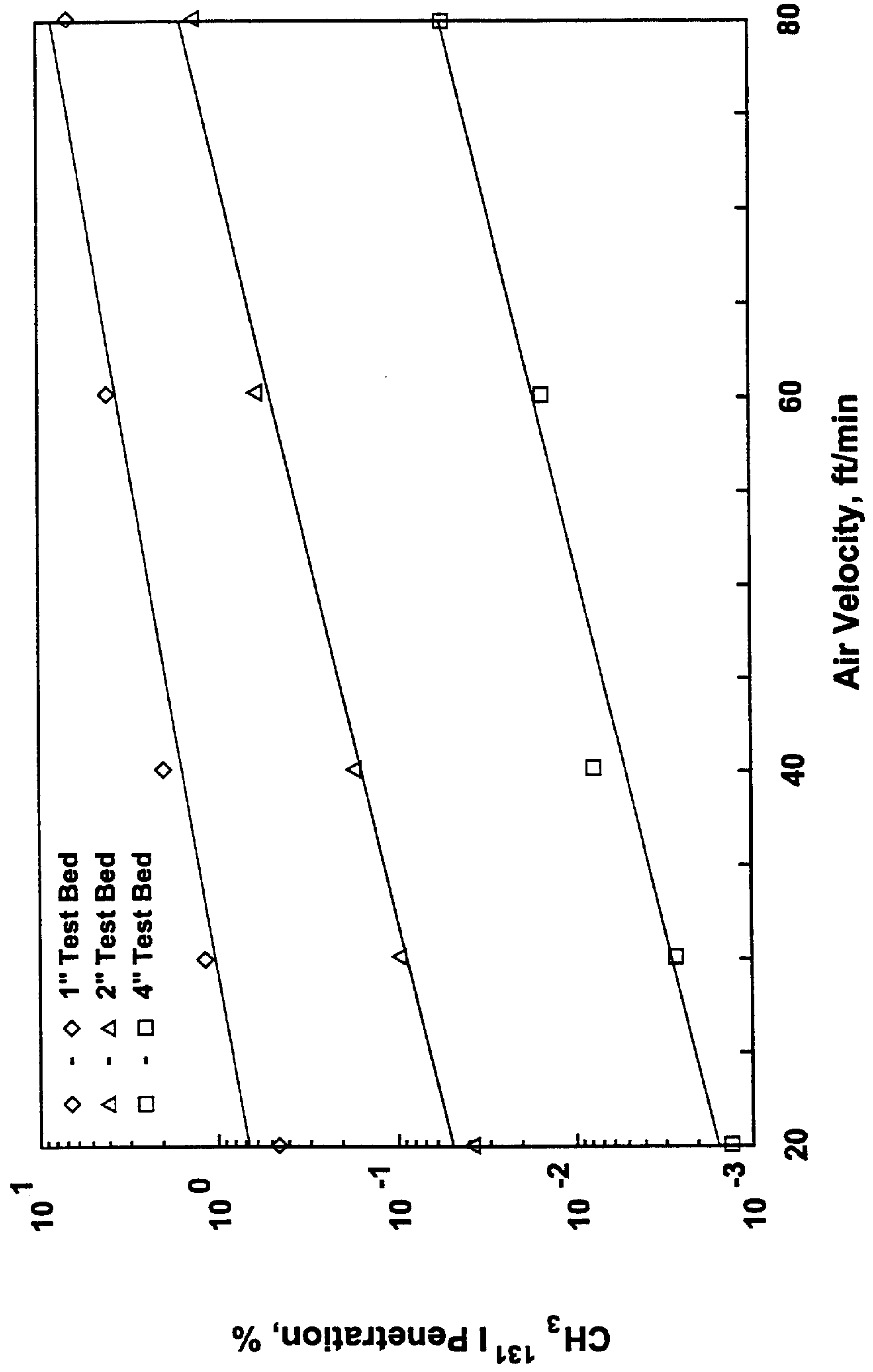


FIGURE 5

PARAMETRIC STUDY OF VELOCITY EFFECTS
ON CH$_3$^{131}I PENETRATION AT 30°C, 95% RH

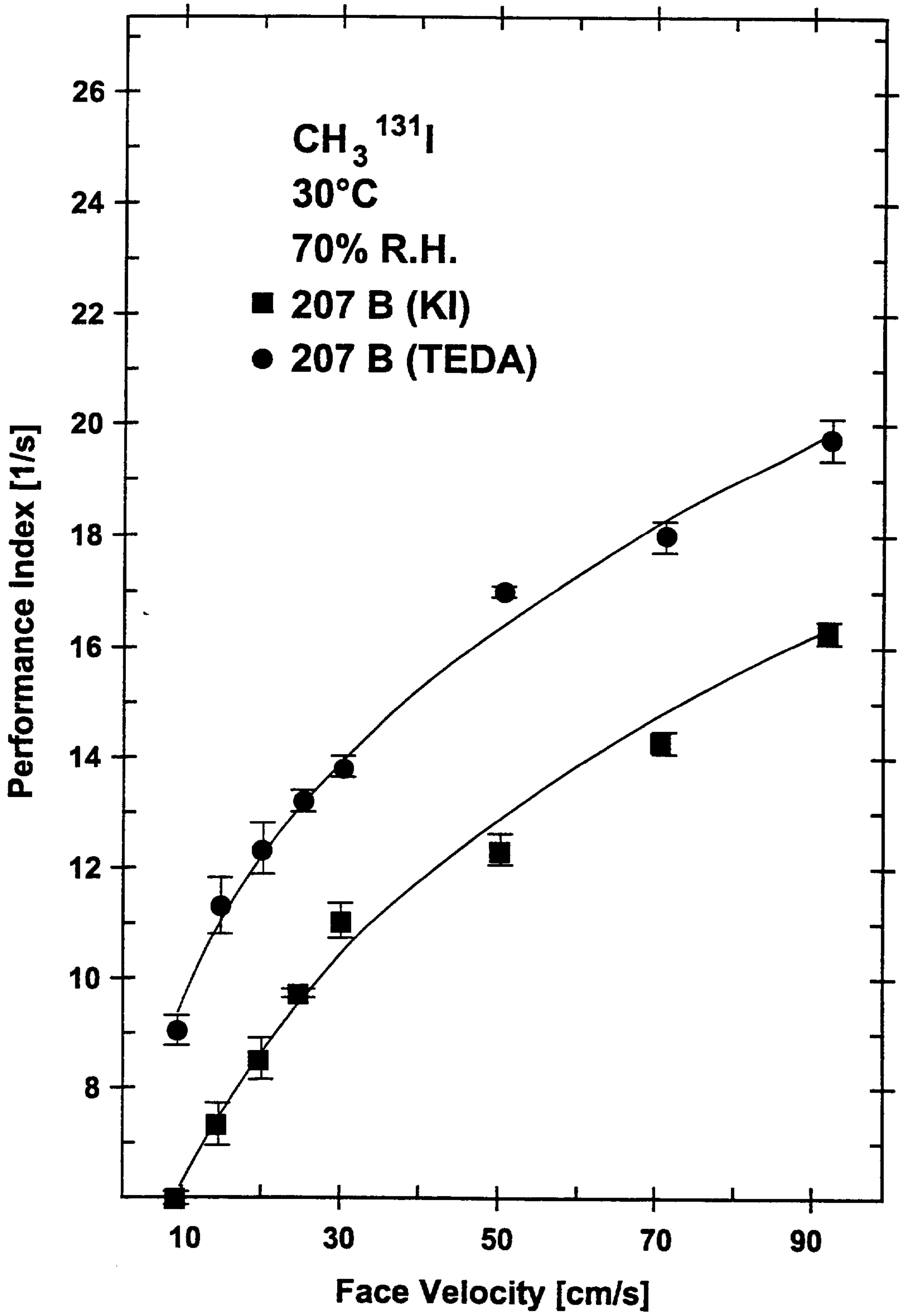


Figure 6. Performance index as a function of face velocity with different impregnated activated carbons.

FIGURE 7

PARAMETRIC STUDY OF PRTICLE SIZE EFFECTS ON $CH_3{}^{131}I$ PENETRATION AT 30°C, 95% RH