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FRACTIONAL HEAT GENERATION RATES IN HANFORD
REACTORS AFTER SHUTDOWN

R. Nilson and R. H. Meichle

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INTRODUCTION

The knowledge of the fraction of decay-heat which is absorbed in fuel elements after a reactor is shutdown is important for many reasons. For example, allowable, reduced flow-rates after shutdown are very sensitive to the manner in which the decay-heat is distributed. Also, the temperature-rise in a discharged, uncooled fuel element is dependent on the total heat generated in the slug. Apart from any heat consideration, the escape of gamma rays from a discharged fuel element is also of importance in certain applications.

A significant refinement in the knowledge pertaining to decay-heat in irradiated uranium in recent years warrants a complete review of the fractional heat generation in the Hanford reactors. Earlier work¹ was based on very qualitative aspects of fission product decay rates and energy spectra following reactor shutdown. The results of these early calculations on the original, solid, Hanford slug indicated that about 20 per cent of the energy generated in a fuel element from both fission product decay and delayed fission escaped from slug. The results reported here show a much smaller escape-fraction. Also, the time-dependence of the escape-fraction is considered for times greater than 100 seconds after reactor shutdown.

SUMMARY

Fractional heat-generation rates are calculated for various Hanford fuel types. The results, condensed to the maximum fraction of energy which escapes from the fuel elements over the period 10^2 - 10^7 seconds after shutdown, are given in table below. Time-dependent curves from 100 seconds to 10^7 seconds after shutdown are given in the body of the report.

<u>Fuel Type</u>	<u>Maximum Fraction Shut Down Heat Escaping Fuel</u>
Original solid Hanford element (1.33" OD)	14%
Present I&E (1.356 - 1.370" OD)	14%
Overbore I&E (1.883" OD)	11%

Absorption of radiation escaping the fuel is almost completely confined to the graphite. The combined coolant water and process tube fractions is less than three percent of the graphite absorption.

¹ HW-33870, "Heat Generation and Total Heat Output From the Pile After Shutdown," S. S. Jones, 11-23-59 (Secret).

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DISCUSSION

The problem of theoretically determining the heat generation rates in irradiated fuel elements after reactor shutdown involves knowledge of the sources of heat, the energy-dependences of the various radiations, how the energy spectrum varies with time, and the probability that particular radiations will escape from the fuel element. Each of these topics is considered in turn.

Heat Sources

The principle sources of energy in irradiated fuel for times greater than 100 seconds after a reactor shutdown are beta and gamma decay of fission products, and beta and gamma decay of U^{239} and Np^{239} . Perkins and King² have calculated fission product, gamma and beta, decay-rates from 123 isotopes due to thermal fission of U^{235} as a function of reactor operation times from one to 1000 hours and as a function of time after shutdown from 100 to 10^8 seconds.

The results of Perkins and King, although falling below Way and Wigner's³ earlier predictions of disintegration rates and total energy release do agree with the Way-Wigner rule-of-thumb expressions⁴ and are in good agreement with experimental measurements.⁵

The data of Perkins and King are split into the seven, decay, gamma-energy groups shown in Table I.

Table I

Decay Gamma-Energy Groups

<u>Group</u>	<u>Gamma-energy range (Mev)</u>
I	0.1 - 0.4
II	0.4 - 0.9
III	0.9 - 1.35
IV	1.35 - 1.8
V	1.8 - 2.2
VI	2.2 - 2.6
VII	2.6

² Perkins and King, NSE, 3, 726 (1958).

³ K. Way and E. P. Wigner, Phys. Rev. 73, 1318 (1958)

⁴ K. Way and E. P. Wigner, Phys. Rev. 70, 115 (1946)

⁵ D. L. Hill and L. Lange, CP-1827 (1944); R. A. Day and C. V. Cannon, CP-2176 (1944); L. Borst, CL-697; E. L. Brady and A. Turkevich, CL-697 (1945); and S. K. et al, CC-1128 (1943).

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An example of decay rates from the Perkins-King compilation is shown in Figure 1 for the case of a 1000 - hour reactor operation. D is the disintegration rate in disintegrations/sec-watt, Γ_T is the total, gamma-energy, decay rate in Mev/sec-watt, Γ_i are the group, gamma-energy, decay rates in Mev/sec-watt and B is the beta-energy, decay rate in Mev/sec-watt. For the case shown the average gamma energy per disintegration is ~ 0.8 Mev at 10^2 seconds and gradually decreases, being about 0.50 Mev at 10^5 seconds and 0.30 Mev at 10^7 seconds. The average beta energy per disintegration varies from .75 Mev at 10^2 seconds to about 0.30 Mev in the region of 10^6 seconds.

The decay of U^{239} is relatively simple. The half-life of the isotope is 23.5 minutes and it decays to Np^{239} by the emission of a beta-particle of maximum energy, 1.21 Mev. Assuming an allowed beta spectrum, the average beta particle energy is $\sim .45$ Mev. The beta decay is followed by a .074 Mev gamma ray.

Time After Shutdown (Sec.)

Fission Product

Figure 1. Fission Product Decay Rates Due to 1000-Hour Reactor Operation

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The decay of Np^{239} is more complex. The gamma spectrum accompanying the 2.33 day, beta decay to Pu^{239} according to Roos⁶ is shown in Table II. The Np^{239} beta decay consists of at least four transitions, 90% of which involve E_{max} of 0.34 - .44 Mev⁷. The average beta decay energy, assuming again an allowed spectrum, is $\sim .15$ Mev.

Table II

Approximate Gamma Spectrum Accompanying Np^{239} Decay

<u>E_γ (Mev)</u>	<u>Mev per Decay</u>
0.05 - .10	0.035
0.10 - .15	0.046
0.15 - .20	0.000
0.20 - .25	0.091
0.25 - .30	0.125
0.30 - .35	0.027
	0.284

The decay energy from U^{239} and Np^{239} is computed in Appendix A. The U^{239} and Np^{239} energy-decay rates are shown in Figure 2. A comparison with the curves of Figure 1 shows, that for times up to $\sim 3 \times 10^5$ seconds after reactor shutdown, the uranium and neptunium activities are competitive with the fission product activities.

Absorption Probabilities

All the energy of fission-product, U^{239} , and Np^{239} beta-decays remains in the fuel element. Thus, to determine the fraction of energy which appears in the various parts of the lattice, it is only necessary to determine the escape-probability of the gamma-energy from the fuel element.

Define $f(E)$ as the fraction of gamma rays of energy E which escapes from the fuel element. The total energy escaping the fuel is

$$F = \int f(E) \Gamma(E) dE, \quad (1)$$

where $\Gamma(E)$ is the total gamma energy appearing per interval dE at E from fission product decays (including U^{239} and Np^{239} decays). Thus, the fraction of energy remaining in the fuel is

$$H = \int B(E) dE + \int [1 - f(E)] \Gamma(E) dE, \quad (2)$$

where $B(E)$ is the total beta decay-energy per interval dE at energy E .

⁶ M. Roos, J. N. E. Part B: Reactor Technology, 1, 98 (1959)

⁷ UCRL-8289, "Decay Schemes," Strominger and Hollander, 1948 (Unclassified).

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Figure 2. U_{239} and Np_{239} Decay Rates - 1000 Hr Operation

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If the slug is in the reactor, the energy F passing out will be absorbed in either water, process tube or graphite. (An insignificant amount will be absorbed in the fuel cladding and the internal water of externally-and-internally-cooled fuel elements). Defining as W , T , and G the fractions, respectively, for the external cooling-water, process tube and graphite, we have

$$W = \int f(E) \Gamma(E) w(E) dE, \quad (3)$$

$$T = \int f(E) \Gamma(E) [1 - w(E)] t(E) dE, \quad (4)$$

$$G = \int f(E) \Gamma(E) [1 - w(E)] [1 - t(E)] dE, \quad (5)$$

where $w(E)$ and $t(E)$ are, respectively, the absorption-probabilities of gamma rays of energy E for water and process tube. All gammas not absorbed in the fuel, water or tube are absorbed in the graphite.

If the gamma rays are assumed to travel straight paths of lengths l through absorbing media of gamma ray energy-absorption coefficients $\mu(E)$, then $w(E)$ and $t(E)$ can be expressed as

$$w(E) = 1 - \exp \left[-\mu_w(E) l_w \right] \quad (6)$$

$$t(E) = 1 - \exp \left[-\mu_t(E) l_t \right]. \quad (7)$$

The average chord lengths for isotropic radiation assumed to originate at the inside wall of a cylindrical shell has been derived by Memmert⁸.

$$\bar{l} = r_o \left[E(\alpha, \pi/2) - \frac{r_i}{r_o} \right], \quad (8)$$

where $E(\alpha, \pi/2)$ is the complete elliptic integral of the second kind

$$E(\alpha, \pi/2) = \int_0^{\pi/2} d\phi \sqrt{1 - \sin^2 \alpha \sin^2 \phi}, \quad (9)$$

r_i and r_o are the inside and outside radii of the cylindrical shell, and $\sin \alpha = \sin r_i/r_o$.

⁸ AERE-Trans-853, "Resonance Absorption in Heterogeneous Systems," G. Memmert, 1960. (Unclassified).

The value of f for a solid cylindrical fuel element assuming a uniform generation of gamma rays throughout the fuel element can be obtained from the curves in Case et al.⁹. For hollow fuel rods filled with water, it is sufficiently accurate to assume (if the inner radius is less than half the outer radius), that f is equal to the f computed for a solid rod of reduced $\mu' = \mu(1 - e^2/a^2)$ where e and a are the inner and outer radii of the fuel element. The gamma energy absorption coefficient for water is assumed to be negligible compared to uranium in this approximation.

A more exact determination of the gamma escape fraction f can be obtained using the method of Story¹⁰ as follows.

First, consider the fission product gamma rays which should have a source distribution in the fuel element corresponding to the thermal neutron distribution within the fuel rod. The escape fraction $f(E)$ is then

$$f(E) = \frac{1}{2} [f'(E) + f''(E)] \quad (10)$$

The $f'(E)$ and $f''(E)$ in the above expression correspond to the Story's escape probabilities $1 - K(\gamma)$ and $1 - K^*(\gamma)$ for infinite cylindrical rods for a uniform source density and a surface-plus-uniform source density, respectively.) Thus, the $f(E)$ desired is just the average of the minimum $f'(E)$ (uniform source-density) and the maximum $f''(E)$ (surface-plus-uniform source-density).

The U^{239} and Np^{239} fission product gamma rays will have a source distribution which in part follows the thermal neutron distribution within the fuel rod. However, a certain fraction of the resonance captures in U^{238} will be surface captures and for such a gamma-source distribution a larger fraction will escape than for the thermal distribution.

Story¹⁰ quotes a rule-of-thumb which seems to be fairly well born out in measured Pu^{239} buildup distributions in cylindrical fuel rods. The rule is that for every 100 resonance neutrons absorbed with a thermal-like distribution, a further $28.2/a$ are captured in the surface of the rod. (The parameter a is the rod radius in cm.)

We therefore assume that the U^{238} activation distribution is made up of a true thermal distribution fraction τ (due to the thermal neutron captures in U^{238}) and a resonance-distribution fraction ρ which in turn is composed of a fraction $(1 + \frac{.282}{a})^{-1}$ having a thermal-like distribution and a fraction $\frac{.282}{a} (1 + \frac{.282}{a})^{-1}$

having solely a surface distribution. Then, in calculating $f(E)$, we take a fraction $\tau + \rho (1 + \frac{.282}{a})^{-1}$ times the $f(E)$ of Equation (10) and a fraction $\frac{.282}{a} (1 + \frac{.282}{a})^{-1} \rho$ times an escape probability $f''(E)$. The escape-fraction $f''(E)$ is for a surface source and in Story's notation is $1 - K^*(\gamma)$.

⁹ Case, de Hoffman and Placzek, "Introduction to the Theory of Neutron Diffusion," Volume 1, (1953).

¹⁰ AERE-TR-2218, "Escape of Gamma Radiation From Uranium Rods in a Pile Heat Evolved in the Moderator," J. S. Story, 1957 (Unclassified).

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Curves of $f(E)$ are given in Figure 3 for the following slug types.

<u>Case</u>	<u>Description</u>
A	Solid slug in BDF Pile
B	Solid slug in K Pile
C	OIIN slug in BDF Pile
D	KIIN slug in K Pile
E	CIIN slug in C Pile
F*	CVIN slug in C Pile

The pertinent dimensions for these cases are given in Appendix B.

Calculations

The gamma ray escape and absorption probabilities for each region of a lattice cell were calculated for each of the seven groups rather than using the integral expressions. Thus,

$$H = B + \sum_{i=1} [1 - f(\bar{E}_i)] \Gamma(\bar{E}_i), \text{ etc.},$$

where $\bar{E}_i$ is the average energy of the gamma rays of each group.

The gamma ray absorption coefficients for uranium, water, aluminum and zirconium were taken from References 11 and 12.

RESULTS

The principal results of these calculations are shown in Figure 4. Here are shown for the 1000-hour operation case the fractional energy escaping the various fuel geometries. For all practical purposes the results for the solid and present-size I&E slugs are identical. Only in the overbore geometry are the results significantly different.

The shape of the curves can be qualitatively explained by referring to the manner in which the fission product gamma spectrum varies with time. The rise in the escape-fraction beyond 10^5 seconds is due to the increased fraction of harder gamma rays (notably groups IV, V and VII) at this period in the decay (See Figure 1). The escape-fraction decreases beyond 10^6 seconds as again the average gamma energy decreases.

* Case F differs from the others in that the process tube is Zr-2 instead of aluminum.

11 ANL-5800, "Reactor Physics Constants," 1958.

12 C. M. Davisson, "Interaction of -Radiation with Matter," Chapter II of Beta- and Gamma-Ray Spectra copy," K. Siegbahn, Interscience Publishers (1955).

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Figure 3B

Gamma Escape Probability $f(E)$
Present I&E Slug
(Cases C,D,E)

Figure 3C
Gamma Escape Probability $f(E)$
C-Overbore IRE
(Case F)

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Figure 4
Escape Fractions of Decay Heats
(1000 Hour Operation)
A Solid and Present I&E Fuel
B CVI Fuel Element (Overbore)

Fraction Energy Escaping Fuel

.14
.13
.12
.11
.10
.09
.08

10^2

10^3

10^4

10^5

10^6

10^7

Time After Shutdown (Sec.)

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The fractions of radiation absorbed in the cooling water external to the fuel element and in the process tube are negligible (< three percent) of the fraction absorbed in the graphite. For all practical purposes, therefore

$$G + H = 1.$$

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APPENDIX A

U²³⁹ and Np²³⁹ Contributions to Decay Heat

Equilibrium U²³⁹ and Np²³⁹

The net formation rates of U²³⁹ and Np²³⁹ in the reactor during equilibrium operation are

$$\dot{N}_{29} = R_{28}(\eta, r) - (\phi \sigma_{29} + \lambda_{29}) N_{29}, \quad (A1)$$

$$\dot{N}_{39} = N_{29} \lambda_{29} - N_{39} (\phi \sigma_{39} + \lambda_{39}), \quad (A2)$$

where $R_{28}(\eta, r)$ = the U²³⁹ formation rate from radiative captures in U²³⁸ (caps/cm³-sec),

ϕ = thermal flux,

σ_{29} = absorption cross section of U²³⁹,

σ_{39} = absorption cross section of Np²³⁹,

λ_{29} = decay constant of U²³⁹,

λ_{39} = decay constant of Np²³⁹,

N_{29} = U²³⁹ concentration (nuclei/cm³), and

N_{39} = Np²³⁹ concentration (nuclei/cm³).

The solutions to (A1) and (A2) are readily given by

$$N_{29}(t) = \frac{R_{28}}{\alpha} [1 - e^{-\alpha t}], \text{ and} \quad (A3)$$

$$N_{39}(t) = \frac{R_{28} \lambda_{29}}{\alpha} \left[\frac{1}{\beta} + \frac{1}{\beta - \alpha} \left(\frac{\alpha}{\beta} e^{-\beta t} - e^{-\alpha t} \right) \right], \quad (A4)$$

where

$$\alpha = \phi \sigma_{29} + \lambda_{29}, \quad (A5)$$

$$\beta = \phi \sigma_{39} + \lambda_{39}. \quad (A6)$$

For $\phi = 3 \times 10^{13}$, the $\phi \sigma$ -terms in (A5) and (A6) are negligible compared with the decay terms. Hence,

$$\alpha = \lambda_{29},$$

$$\beta = \lambda_{39},$$

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and B(5) and B(4) simplify to

$$N_{29}(t) = \frac{R_{28}}{\lambda_{29}} [1 - e^{-\lambda_{29}t}], \quad (A7)$$

$$N_{39}(t) = R_{28} \left[\frac{1}{\lambda_{39}} + \frac{1}{\lambda_{39} - \lambda_{29}} \left(\frac{\lambda_{29}}{\lambda_{39}} e^{-\lambda_{39}t} - e^{-\lambda_{29}t} \right) \right]. \quad (A8)$$

The values of λ_{29} and λ_{39} are respectively, 4.92×10^{-4} ($T_{1/2} = 23.5$ min) and 3.44×10^{-6} ($T_{1/2} = 2.33$ days). Since $\lambda_{29} \gg \lambda_{39}$, the term $\lambda_{39} - \lambda_{29}$ in Equation (A8) can be approximated to $-\lambda_{29}$, which further simplifies the expression (A8).

$$N_{39}(t) = R_{28} \left[\frac{1}{\lambda_{39}} + \left(\frac{1}{\lambda_{29}} e^{-\lambda_{29}t} - \frac{1}{\lambda_{39}} e^{-\lambda_{39}t} \right) \right]. \quad (A8')$$

U²³⁹ and Np²³⁹ - Concentration After Shutdown

If the reactor has been operating for a period T seconds and then shut off the concentrations of U²³⁹ and Np²³⁹ as a function of time after shutdown are as follows:

$$N_{29}(t) = \frac{R_{28}}{\lambda_{29}} [1 - e^{-\lambda_{29}T}] e^{-\lambda_{29}t}; \quad (A9)$$

$$N_{39}(t) = R_{28} \left[\frac{1}{\lambda_{39}} + \left(\frac{1}{\lambda_{29}} e^{-\lambda_{29}T} - \frac{1}{\lambda_{39}} e^{-\lambda_{39}T} \right) \right] e^{-\lambda_{39}t} + \frac{R_{28}}{\lambda_{29}} (1 - e^{-\lambda_{29}T}) (e^{-\lambda_{39}t} - e^{-\lambda_{29}t}). \quad (A10)$$

U²³⁹ and Np²³⁹ Disintegration Rates and Energy-Decays After Shutdown

The disintegration rates for U²³⁹ and Np²³⁹ after reactor shutdown are given by

$$R_{29}(t) = N_{29}(t) \lambda_{29}, \text{ and} \quad (A11)$$

$$R_{39}(t) = N_{39}(t) \lambda_{39}. \quad (A12)$$

The energy decays can then be obtained by multiplying (A11) and (A12) by the energy emission per disintegration. As discussed in the text, the energy emissions per disintegration are as follows,

$$\begin{aligned} E_{29} (\beta) &= .45 \text{ Mev/dis,} \\ E_{29} (\gamma) &= .074 \text{ Mev/dis,} \\ E_{39} (\beta) &= .15 \text{ Mev/dis, and} \\ E_{39} (\gamma) &= .284 \text{ Mev/dis.} \end{aligned} \tag{A13}$$

Derivation of R_{28}

The U^{238} neutron capture rate must be determined as a function of power in the pile so that the U^{239} and Np^{239} decay-energies can be considered on the same scale as the other fission product decay-energies compiled by Perkins and King.

We define the following rates:

$$F = \phi N_{25} \sigma_{f25} (1 + \delta), \text{ and} \tag{A14}$$

$$B = \phi N_{25} \sigma_{a25}. \tag{A15}$$

F is the fission rate per cm^3 of fuel and B is the U^{235} burnout rate per cm^3 of fuel. In the above expressions,

$$N_{25} = U^{235} \text{ nuclei/cm}^3,$$

$$\sigma_{f25} = U^{235} \text{ thermal neutron fission cross section,}$$

$$\sigma_{a25} = U^{235} \text{ thermal neutron absorption cross section,}$$

$$\delta = \text{fast } (U^{238}) \text{ to thermal fission ratio.}$$

The U^{238} - n, γ rate is equal to the conversion ratio times the N^{235} burnout rate, by definition.

$$R_{28} = \phi N_{25} \sigma_{a25} CR. \tag{A16}$$

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The power density (watts per cm^3) is proportional to the fission rate.

$$P = \phi N_{25} \sigma_{f25} (1 + \delta) H, \quad (\text{A17})$$

where H is the heat of fission, 3.2×10^{11} watt-sec. Equations (A16) and (A17) can be solved simultaneously to yield R in terms of P.

$$\begin{aligned} R_{28} &= \frac{CR \sigma_{a25} P}{\sigma_{f25} (1 + \delta)} \cdot 3 \times 10^{10} \quad (\text{A18}) \\ &= \text{U}^{239} \text{ production rate/cm}^3 \text{ fuel/watt-cm}^{-3}. \end{aligned}$$

U²³⁹ and Np²³⁹ Contribution to Decay Heat - 1000 Hour Reactor Operation.

For T = 1000 hours, Equation (A9) and (A10) for the nuclear concentrations after shutdown reduce to

$$N_{29}(t) = \frac{R_{28}}{\lambda_{29}} e^{-\lambda_{29} t}, \quad (\text{A19})$$

$$N_{39}(t) = \frac{R_{28}}{\lambda_{39}} e^{-\lambda_{39} t} + \frac{R_{28}}{\lambda_{29}} \left(e^{-\lambda_{39} t} - e^{-\lambda_{29} t} \right). \quad (\text{A20})$$

Then the disintegration rates are for any time t after shutdown,

$$R_{29}(t) = N_{29}(t) \lambda_{29} = R_{28} e^{-\lambda_{29} t},$$

$$R_{39}(t) = N_{39}(t) \lambda_{39} = R_{28} e^{-\lambda_{39} t} + R_{28} \frac{\lambda_{39}}{\lambda_{29}} \left(e^{-\lambda_{39} t} - e^{-\lambda_{29} t} \right).$$

The equations reduce numerically to the following when the values of the constants are inserted.*

$$* \lambda_{29} = 4.92 \times 10^{-4}$$

$$\lambda_{39} = 3.44 \times 10^{-6}$$

$$CR \cong .85$$

$$\frac{\sigma_{a25}}{\sigma_{f25}(1+\delta)} \cong 1.13$$

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$$R_{29}(t) = 2.88 \times 10^{10} P e^{-\lambda_{29} t} \text{ dis/sec-cm}^3$$

$$R_{39}(t) = 2.88 \times 10^{10} P e^{-\lambda_{39} t} \text{ dis/sec-cm}^3$$

Since the units of P are watts/cm³, dividing out P yields the disintegration rates in units of dis/sec-watt which is the same unit that Perkins and King use in their fission product decay compilation. Finally, multiplying the disintegration rules by the appropriate beta- and gamma-energy values per disintegration yields the curves shown in Figure 2 of the text.

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APPENDIX B

Dimensions of Particular Cases Considered

<u>Case</u>	<u>Fuel Element (Inches)</u>		<u>Water Annulus (Inches)</u>	
	<u>Outer Diam.</u>	<u>Inner Diam.</u>	<u>Inner Diam.</u>	<u>Outer Diam.</u>
A	1.336	0	1.438	1.591
B	1.336	0	1.438	1.659
C	1.356	.416	1.445	1.591
D	1.370	.491	1.460	1.659
E	1.370	.481	1.460	1.623
F	1.883	.452	1.991	2.147

<u>Case</u>	<u>Process Tube (Inches)</u>	
	<u>Inner Diam.</u>	<u>Outer Diam.</u>
A	1.591	1.729
B	1.659	1.806
C	1.591	1.729
D	1.659	1.806
E	1.623	1.764
F	2.147	2.237

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