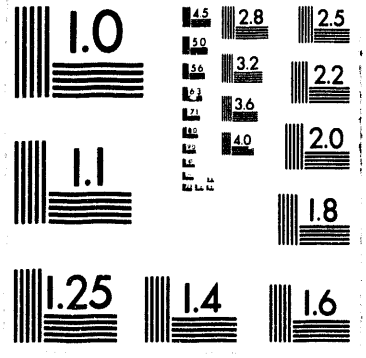
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A Letter Report for Westinghouse Hanford Company

Review of Grout Particulate-Emissions Methodology

M. Y. Ballinger

September 1993

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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Letter Report

REVIEW OF GROUT PARTICULATE-EMISSIONS METHODOLOGY

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Pacific Northwest Laboratory
Richland, Washington 99352

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REVIEW OF GROUT PARTICULATE-EMISSIONS METHODOLOGY

A model has been developed by Westinghouse Hanford Company (Program Release) to estimate the quantity of particulate aerosols that would become airborne during the pouring of grout into a storage vault. Information and equations derived from spill experiments were used in the model to determine release fractions. This letter report discusses the similarities and differences between the spill experiments and the grout vault operations, the applicability of the spill equations, and the use of particle depletion models to account for the residence time of particles in the grout vault. The following equation is recommended for Program Release to estimate the quantity of aerosol generated during pouring:

$$F = 1.26E\text{-}5\ Fr^{0.35}\ Arch^{0.45}\ (Rho_a/Rho_l)^{2.2}$$

A method is also recommended to estimate aerosol deposition in the grout vault.

1.0 Similarities/Differences Between Spill Experiments and Grout Operations

The spill experiments involved spills of various solutions and powders from a height of 1 to 3 m in a large tank (20 m^3). The following parameters were varied: spill height (1-3 m), material quantities (125-1000 cc for liquids), material form (powders, solutions, slurries), and solution characteristics (density, viscosity, surface tension). In each experiment, the duration of the spill was on the order of a few seconds and ventilation flow was provided by pulling air through particulate samplers. A flow rate of about 5 m^3/min was used. At this flow rate, the residence time of air in the tank was 4 minutes. Experiments were performed at room temperature and under low relative humidities (RH).

Grout operations consist of continuous pouring of a slurry into a large vault (6000 m^3). The temperature and relative humidity in the vault are elevated (40°C and 100% RH). The volumetric flow rate through the vault is supplied by a ventilation system that draws 1.7 m^3/s through the vault. As the vault fills, the residence time of air in the vault decreases from 59 minutes to 7 minutes. Key parameters associated with both the grout pouring operations and the experimental slurry spills are presented in Table 1.

Table 1. Comparison of Spill Experiments and Grout Operations

	Grout Operations	Spill Experiments (Slurry Spills)
Solution Density, g/cc	1.6	1.12 - 1.41
Solution Viscosity, poise	0.022	0.01 - 0.05
Surface Tension, dyne/cm	70	58 - 68
Ventilation Flow, m^3/s	1.7	0.09
Residence Time, min	7 - 59	4
Spill Height, m	1 - 10	3
Quantity Spilled, cc	3785/s	1000

Solution characteristics roughly correspond to experimental values. The height of much of the grout spill is greater than that of the experiments, but is not considered extreme for model application.

One major difference between grout pouring operations and the spill experiments is that the grout is released through a semi-continuous process and the experiments were single-event spills. The proposed model (Program Release) accounts for this difference by representing the grout pour as a series of small spills (less than 1 second in duration). The representation of the grout pour as a series of small spills appears acceptable for the following reasons:

1) One of the parameters in the equations derived from the experimental spills is the radius (R) of an equivalent sphere of liquid spilled. For pour times of less than 1 second, the equivalent sphere radius is on the order of the actual radius of the pour. Grout is poured through a 5-cm (2-inch) diameter pipe so the radius as calculated using a 0.02-second factor is the actual radius of the pour (vol = 3785 cc/s x 0.02 s =75.7 cc, $R = (3/4\ vol/\pi)^{1/3} = 2.6$ cm). For pours of less than 1 second, the equivalent sphere radius is less than 9.6 cm (3.8 inch).

2) The equation is not very sensitive to the time chosen. The release fraction is proportional to $R^{-0.35}$. Increasing the time factor from 1 second to 100 seconds (and increasing the volume spilled) would cause the release fraction to be reduced by 42%. Decreasing the time factor from 1 second to 0.01 second causes the release fraction to increase by 71%.

The particles of primary concern in the grout (radioactive cesium) are soluble. In the slurry spill experiments, the soluble aerosol (uranine) was the portion of the aerosol that was measured. Consequently, results from the experiment are considered appropriate for determining the release fraction of soluble materials. Insoluble particles may not be as accurately predicted.

The high humidity in the grout vault indicates that any aerosol from a grout pour would be less likely to decrease in size because of evaporation than would aerosols in the spill experiments. Ballinger et al. (1988) show how to correct for the evaporation and settling that occurs in the experimental chamber (called the Radioactive Aerosol Release Tank or the RART) but that would not occur in the actual grout pour.

The "initial aerosol" is the aerosol that was originally formed from the spill. In the spill experiments, this aerosol changed by evaporation and settling before it reached the collection devices (impactors). It is appropriate to use the equations derived from the initial aerosol data for the grout operations, particularly if other credit is taken for aerosol depletion. Thus, data from the impactors was corrected to account for the change. Both the impactor data and the corrected data were reported in Ballinger et al. (1988). The corrected data are referred to as the initial aerosol data.

As noted in Table 1, ventilation conditions in the grout vault differ markedly from those in the experimental chamber. The smaller residence time in the RART would allow less particle depletion than in the vault. In

addition, filters collecting the aerosol in the RART were only 1-2 m from the spill site. In contrast, the vault exhaust is located in an upper corner, 20 m or more from where the grout is poured. Therefore, filter loadings in the experimental chamber are expected to be higher than would occur in the vault exhaust.

The primary particle depletion mechanism expected under vault conditions is gravitational settling. Thermophoresis (as a result of a temperature gradient within the vault) could contribute to depletion; however, this effect is expected to be minor compared to settling. An analysis (including equations) of the effects of gravitational settling in the vault is presented in Section 4.0.

2.0 Models to Estimate Release Fractions from Slurry Spills

Several equations are given in Ballinger et al. (1988) to predict the fraction (F) of material that will aerosolize during spills of solutions (including slurries). These equations are:

Equations for Measured Data:

$$F = 8.12E\text{-}10\ Arch^{0.55} \qquad \text{eqn 1}$$

$$F = 2.3E\text{-}5\ Arch^{0.44}\ (Rho_a/Rho_l)^{2.4}\ Fr^{0.38} \qquad \text{eqn 2}$$

Equations for Corrected Data:

$$F = 8.9E\text{-}10\ Arch^{0.55} \qquad \text{eqn 3}$$

$$F = 6.31E\text{-}6\ Arch^{0.45}\ (Rho_a/Rho_l)^{2.2}\ Fr^{0.35} \qquad \text{eqn 4}$$

Definitions are:

$$Fr = V^2/(g * R) = 2H/R$$

V = spill velocity, cm/s
g = gravity constant, cm/s^2
H = spill height, cm
R = radius of equivalent sphere, cm

$$Arch = \text{Archimedes Number} = Rho_l^2\ H^3\ g/u^2$$

Rho_l = liquid density, g/cc
μ = liquid viscosity, poise (g/cm s)
Rho_a = density of air, g/cc

The following analysis is provided to determine the general applicability of equations 1 through 4 to slurry spills (the equations were developed from spills of all types of solutions) and to determine the most appropriate equation for the grout process.

In this analysis, the equations are used to see how well they predict the release from the slurry experiments. The parameters in the experiments are

H =	300 cm	
vol =	1000 cc	$R=(3/4\ vol/\pi)^{1/3}$
R =	6.2 cm	

$g =$ 980 cm/s^2
$Rho_a =$ 0.00121 g/cc (Welty, Wicks, and Wilson 1976)
$Fr =$ 96.9

Results from the spill experiments (with varying solution densities and viscosities) are presented in Table 2. These values were obtained from Table A.2 of Ballinger and Hodgson (1986) and Appendix A of Ballinger et al. (1988). Table 2 presents the actual release fractions obtained in the experiments as well as the release fractions predicted using equations 1 and 2.

As can be seen in Table 2, equation 1 overestimates the release fraction by several orders of magnitude, and equation 2 tends to underestimate the release fraction by about half. If the density of air is used instead of the density of the liquid in the Archimedes number in equation 1, there is a much closer agreement with experimental data (as shown in last column in Table 2). The values presented in Table 2 neglect both the evaporation and the settling of particles.

Table 2. Computed Equation Values for Slurry Experiments (Measured Equations)

					------ Release Fractions -------			
						Predicted		
Run No.	Rho_l (g/cc)	Viscosity (poise)	Arch	Density Ratio	Actual	Eqn 2	Eqn 1	Eqn 1[a]
2	1.123	0.032	3.3E+13	0.00108	8.7E-06	8.7E-06	0.022	1.2E-05
4	1.155	0.030[b]	3.9E+13	0.00105	1.1E-05	8.8E-06	0.024	1.3E-05
1	1.189	0.049	1.6E+13	0.00102	9.2E-06	5.5E-06	0.015	7.5E-06
5	1.201	0.031	4.0E+13	0.00101	1.8E-05	8.1E-06	0.024	1.2E-05
3	1.334	0.013	2.8E+14	0.00091	4.6E-05	1.5E-05	0.072	3.2E-05
8	1.345	0.013	2.8E+14	0.00090	2.7E-05	1.5E-05	0.072	3.2E-05
6	1.286	0.013	2.6E+14	0.00094	3.0E-05	1.6E-05	0.069	3.2E-05
7	1.407	0.029	6.2E+13	0.00086	1.6E-05	6.7E-06	0.031	1.3E-05
				Average	2.1E-05	1.0E-05	0.041	1.9E-05

[a] The density of air was used instead of solution density in the Archimedes number.
[b] Viscosity was not measured for run 4. This value is assumed based on the similarity to other slurry properties.

Data on the initial aerosol are also given in Ballinger et al. (1988) and are used to derive equations 3 and 4. Table 3 compares predictions of the release fraction using equations 3 and 4 to the initial aerosol release fraction. Initial aerosol data are back calculated from the experimental results by estimating the evaporation and settling of airborne particles before they reached the collection devices.

Again, equation 3 overestimates the release fractions by several orders of magnitude, but if the density of air is used, equation 3 works fairly well (agrees with experimental to within 40%). Equation 4 underestimates release fractions by a factor of about two.

3.0 Application of Models to Grout Vault Operations

Equation 4 is the most appropriate model to apply to the grout operations because it reflects the initial aerosol that would be generated from a liquid spill. The use of this equation allows evaporation and settling conditions within the grout vault to be considered separately from those in the spill experiments. If equation 4 is multiplied by a factor of 2, the results seem to reasonably and conservatively reflect the quantity of initial aerosol (before evaporation and settling) that was produced by the slurry experiments. Because of the discrepancy about which density to apply in equations 1 and 3, equation 3 is not recommended, and the use of equation 4

Table 3. Release Fractions from Slurry Experiments - Initial Aerosol

	Release Fractions				
		Predicted Values			
Run No.	Actual	Eqn 3	Eqn 4	(Eqn 4)x 2	Eqn 3[a]
2	9.6E-06	0.024	1.1E-05	2.2E-05	1.3E-05
4	1.3E-05	0.027	1.1E-05	2.3E-05	1.4E-05
1	1.0E-05	0.016	7.1E-06	1.4E-05	8.2E-06
5	1.9E-05	0.027	1.1E-05	2.1E-05	1.4E-05
3	5.2E-05	0.078	2.0E-05	4.0E-05	3.5E-05
8	3.0E-05	0.079	2.0E-05	4.0E-05	3.5E-05
6	3.4E-05	0.075	2.1E-05	4.2E-05	3.5E-05
7	1.9E-05	0.034	9.1E-06	1.8E-05	1.5E-05
Average	2.3E-05	0.045	1.4E-05	2.8E-05	2.1E-05

[a] The density of air was used instead of solution density in the Archimedes Number.

with an additional multiplication factor of 2 is recommended. Thus, the equation is

$$F = 1.26E\text{-}5\ Fr^{0.35}\ Arch^{0.45}\ (Rho_a/Rho_l)^{2.2} \qquad \text{eqn 5}$$

Example values for the grout vault are:

Rho_a = 0.00113 g/cc for air @ 40°C
Rho_l = 1.6 g/cc
Rho_a/Rho_l = 0.00071
g = 980 cm/s^2
μ = 0.022 poise
Vol = 3785 cc (assuming a 1-second spill)
R = 9.6 cm $\quad R = (3/4\ vol/\pi)^{1/3}$

In these example calculations, the initial velocity of the spilled material is neglected. Table 4 shows the range of release fractions at the extreme grout vault conditions.

Table 4. Release Fractions for Grout Spills

Height (cm)	Arch	Fr	Release Fraction
1036	5.8E+15	214	1.2E-04
102	5.5E+12	21.1	2.3E-06

4.0 Depletion of Spill Particles in the Vault

Particles to be generated in the grout pouring operation are assumed to be similar in size to those from slurry spill experiments. Ballinger et al. (1988) shows an average Aerodynamic Mass Median Diameter (AMMD) of 3.1 μm and average geometric standard deviation (sigma-g) of 6.7 for slurry spills. The initial similarity in particle size distributions may not persist after a period of time; the higher humidity and a lower ventilation rate in the grout vaults should produce a lower rate of evaporation and a higher rate of gravitational settling and alter the particle size distribution from what existed in the spill experiments.

An initial aerosol size distribution was computed for the slurry spill experiments. An evaporation/settling code was developed and used to simulate conditions in the experimental chamber and back calculate what the original spill aerosol would have been given the amount collected on the experimental apparatus (cascade impactors). As shown in the document (Ballinger et al. 1988), the initial aerosol from slurry spills has an average AMMD of 15.8 microns and a sigma-g of 10.1. This size distribution, when plotted on probability paper, gives the fraction of particles in each size range. Figure 1 shows the plot. Table 5 shows the mass fraction of the total aerosol in each size range. Sizes of 1, 3, 5, 7, 10, 20, 50, and >50 (assumed to be 100) microns were chosen for the calculations of particle settling.

Settling velocities were determined using the following equation (Chan, Ballinger, and Owczarski 1989):

$$Vs = Rho_p \, D^2 \, g \, Cm/(18 \, \mu)$$

Vs = settling velocity, cm/s
Rho_p = particle density, g/cc
D = particle diameter, cm
g = gravity constant, cm/s^2
Cm = Cunningham correction factor, dimensionless
μ = gas viscosity, poise

This equation predicts the settling velocity of particles in still air. The pouring operation may produce turbulent eddies that would decrease settling velocity. This effect is neglected in the following analyses because of the lack of quantitative data on turbulence created from spills and its effect on settling velocity.

The size measured in the experiments was in Aerodynamic Equivalent Diameter which means that the particles captured exhibited the behavior of a sphere of unit density (1 g/cc) with the measured diameter. Thus, Rho_p is 1 g/cc.

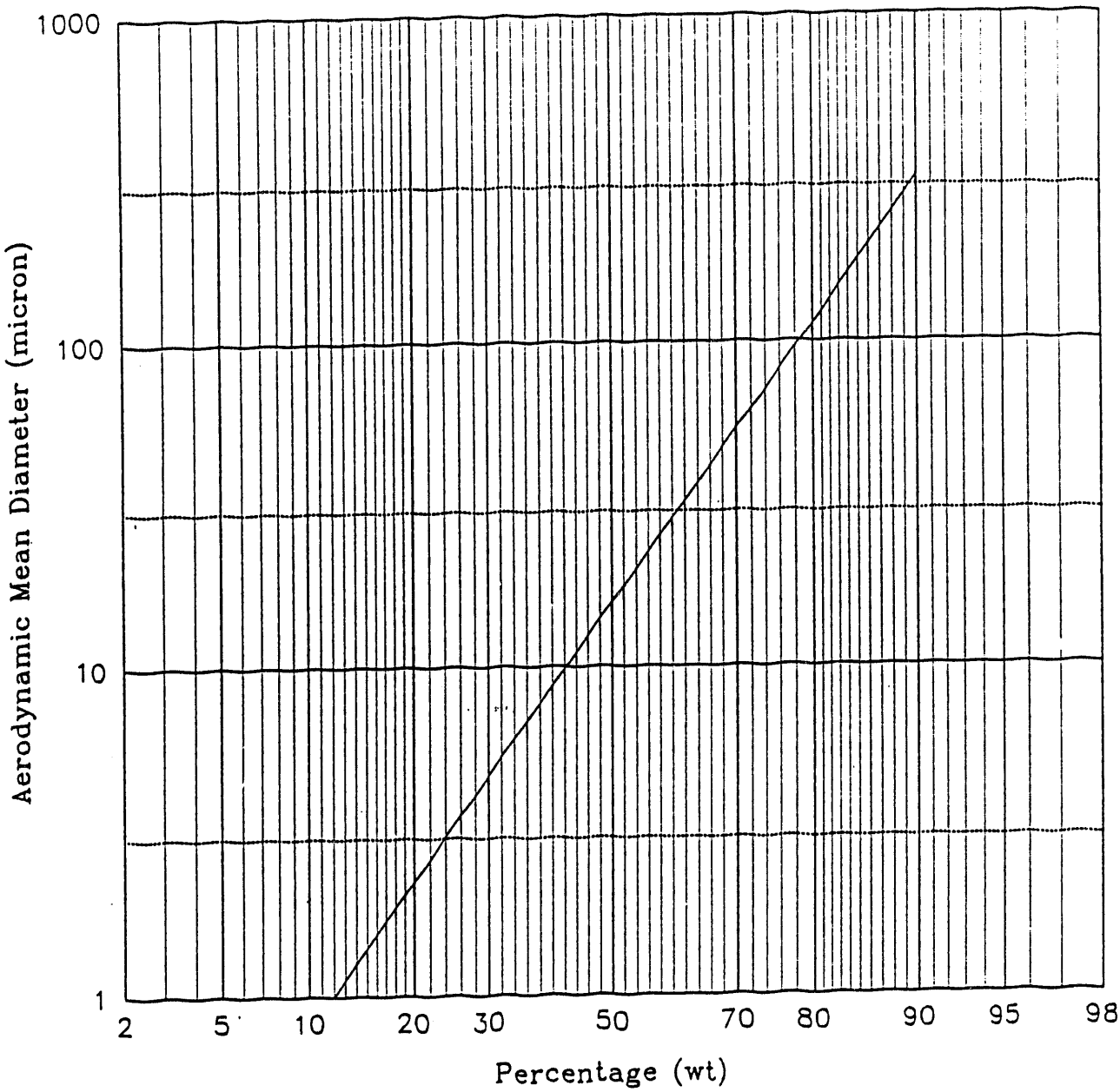

Figure 1. Size Distribution of Particles from Slurry Spills (Average Initial Aerosol)

$g = 980\ cm/s^2$
Cm = 1 for particles <1000 μm
μ = 0.00019 poise @T = 40°C (Welty, Wicks, and Wilson 1976)

Vs is calculated using these values and is shown in Table 5.

Table 5. Calculation of Total Particle Depletion in Grout Vault

Size (μm)	Fraction < Size[a]	Settling Velocity (cm/s)	Fraction in Size Depositing H=1036 cm	Fraction in Size Depositing H=102 cm	Total Fraction Depositing H=1036 cm	Total Fraction Depositing H=102 cm
1	0.12	0.003	0.02	0.02	0.002	0.002
3	0.12	0.026	0.17	0.15	0.020	0.018
5	0.07	0.072	0.40	0.37	0.028	0.026
7	0.04	0.140	0.63	0.59	0.025	0.024
10	0.07	0.287	0.87	0.84	0.061	0.059
20	0.12	1.146	1.00	1.00	0.120	0.120
50	0.15	7.164	1.00	1.00	0.150	0.150
100	0.31	28.655	1.00	1.00	0.310	0.310
				Total	0.717	0.708

[a] Fraction of particles less than stated size and greater than size above.

The fraction of particles depositing at each time step can be computed (Beddow 1980) by

$$Dep = 1-\exp(-Vs\ l/w\ h) \qquad \text{eqn 6}$$

Dep = fraction deposited
l = distance to travel to filters, cm
w = flow velocity, cm/s
h = distance particle must fall, cm

Vault dimensions are 123.5 ft x 50.5 ft x 34 ft. The grout is poured into the vault center, and the exhaust filters are located in an upper corner. (Air inlet is in an opposite corner). Thus, l is $(62^2 + 25^2 + 34^2)^{0.5}$ = 74.9 ft (2280 cm) when the vault is empty. The lowest spill height is 3.3 ft, so l is = 66.8 ft (2036 cm) when the vault is full. The vault flow rate is 1,700,000 cc/s. The average flow velocity when the vault is empty is calculated by dividing the volumetric flow by the height and the average of the other two dimensions.

H (empty) = 34 ft = 1036 cm
L = 123.5 ft = 3764.28 cm
W = 50.5 ft = 1539.24 cm
w (empty) = 0.62 cm/s
H (full) = 102 cm
w (full) = 6.3 cm/s

The distance the particle must fall is assumed to be half of the spill height or 518 cm when empty, 51 cm when full.

Using these values, the fraction of particles in each size range is computed for both full and empty vault conditions. These values are shown in Table 5.

The total fraction of material deposited is calculated by multiplying the incremental fraction of each size by the fraction deposited. Table 5 also shows these values.

The results show that about 71% of the aerosol would deposit in the grout vault. Also, the depletion rate is fairly constant throughout the pouring process. When the vault is empty, the aerosol has farther to fall, but a slower flow velocity and slightly longer distance to travel to get to the filters. When the vault is full, the distance to fall is much less, but the flow velocity is greater and distance to the filters slightly shorter. The trade-off during the pour between flow velocity and fall height appears to be about even.

Using the release fractions computed in Table 4 and applying a factor of 0.3 for the quantity of aerosol that would remain airborne suggests that from 7.0E-7 to 3.6E-5 of the grout would remain in the air long enough to challenge the ventilation system.

5.0 Recommendations

The spill data and models presented in Ballinger et al. (1988) can be used to estimate the release of airborne particles from grout pouring operations. By dividing the pour into intervals of 1 second or less, the grout model can obtain a reasonable estimate of aerosol generated in the vault. It is recommended that the following equation be used in Program RELEASE:

$$F = 1.26E\text{-}5\ Fr^{0.35}\ Arch^{0.45}\ (Rho_a/Rho_1)^{2.2} \qquad \text{eqn 7}$$

This equation does not underestimate the quantity of initial aerosol from the experimental slurry spills and can be used in combination with a credit for particle depletion.

An assumption of about 70% settling may be used because total particle settling does not vary significantly with vault fill (the trade-off between distance particles must fall, and flow velocity is about equal). If a more detailed estimate is desired, the settling equations may be incorporated into Program RELEASE.

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