FOAM DECONTAMINATION OF LARGE NUCLEAR COMPONENTS BEFORE DISMANTLING

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ABSTRACT

An application of a foam decontamination technique is described that allows declassification of ferritic steel wastes (large internally contaminated valves of complex geometry) for melting and material restricted reusing. Alkaline, acid and neutralizing foam solutions are circulated inside the components for several hours. The qualities of foam required for successful decontamination are indicated in terms of their half-life and coalescence properties.

ADVANTAGES OF FOAM DECONTAMINATION

Until recently, few techniques have been available for decontaminating, to a low level, large internally contaminated components with complex shapes before dismantling. Spraying does not reach all internal surfaces evenly due to preferential flow patterns. Immersion systems are not practicable as the reactants are ineffective on large volumes because of boundary layer phenomena, and because of the excessive reactant volume required (at least three times the component volume). Mechanical methods cannot reach all the internal surfaces, and recovery of the abrasion residue is difficult.

The advantages of foam decontamination by refoaming (1) and by recirculation (2) have been recognized in recent years. They include foam expansion to reach all the interstitial gaps of the component; foam mobility and continuous renewal on component walls, which eliminates boundary layer effects; small liquid volume requirements (ten times less than to fill the component) with the option of using stronger reactants; and continuous filtration of the reactants during the decontamination procedure.

RECIRCULATING FOAM DECONTAMINATION PRINCIPLES

The foam used consists of a gas, generally air, dispersed in a liquid in proportions such that the mixture density more closely approximates that of a gas than a liquid. Suitable surface-active agents are added to lower the surface tension of the liquid containing chemical decontaminants, and thus facilitate the formation of a foam.

Recirculation consists in supplying the component with foam decontaminants; foam flowing out from the top of the component is broken, reformed and recycled back into the unit for periods of up to several hours.

The foam is naturally destroyed by drainage through Plateau borders and by coalescence among bubbles. However, the working life of the foam must be long enough to fill the entire component volume and flow out with sufficient moisture content to get a good decontamination.

Foam Working Life

A test vessel with a volume V is initially filled with foam that is as representative as possible, and the rise in the liquid level L due to coalescence is observed. Experimental observations have shown that:

$$L = L_o(1 - e^{-kt}) \qquad \text{(Eq. 1)}$$

where t is the time (refer to Fig. 1) and L_o the liquid quantity recovered after an infinite time (in fact, the quantity of liquid present in the foam when sampled).

Generally the foam does not break immediately, and fills the entire volume V throughout the observation period. This was the case in these experiments as long as the bulk factor ($f = V/L_o$) did not exceed 40.

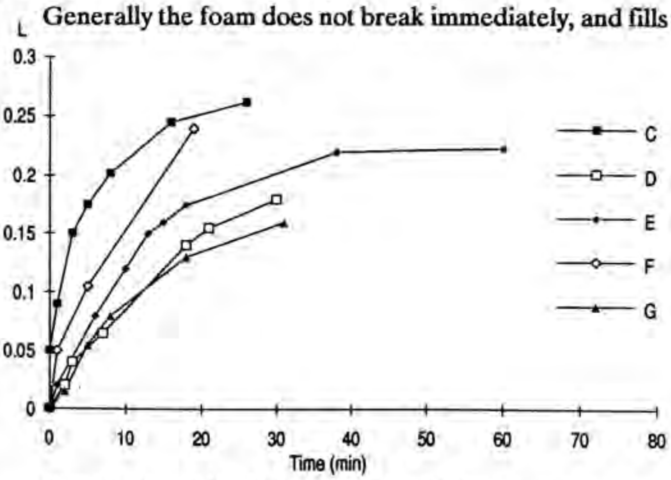

Fig. 1. Rise in the liquid level due to coalescence.

This may also be written: $L' = L_o - L = Lo \bullet e^{-kt}$ (where L' is the residual liquid trapped in the foam). This formula was proposed by Bikerman (3); other similar formulas are presented by Doufare (4).

Figure 2 shows the function $1 - L/L_o$ versus time plotted in semilogarithmic coordinates. If Eq. (1) is verified, the curves should be straight lines; in fact, the actual plot deviates slightly from this simulation.

The term "half-life" ($t_{1/2}$) will be used here to represent the time after which $L = L_o/2$ (a more accurate term might be the "half-liquefaction time"). Equation (1) may then be written as follows:

$$\frac{L}{Lo} = 1 - e^{-0.693\, t/t_{1/2}}$$

Moreover, if f_o is the initial bulk factor, then:

$$f_o = \frac{V}{L_o} \qquad f' = \frac{V}{L'} \qquad f' = \frac{V}{L_o - L} = \frac{V}{L_o \cdot e^{-kt}}$$

$$\frac{f'}{f_o} = e^{+0.693\, t/t_{1/2}} \qquad \text{(Eq. 2)}$$

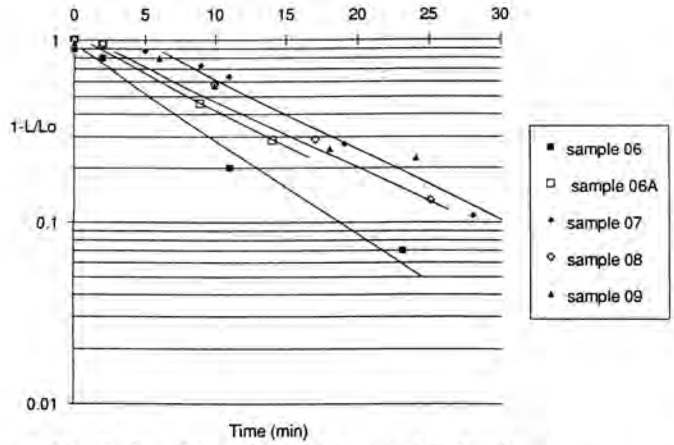

Fig. 2. Fraction of liquid trapped in the foam.

It is easy to identify the measured half-life of each numbered foam composition on Fig. 2, representing one experiment. The test foams included a standard acid composition (06a) with a half-life of 8 minutes, and the same foam with 0.5% polyethylene glycol additive, which extended the initial half-life to 14 minutes (09); after 90 minutes in operation, the half-life was only 12 minutes (07). These foams were sampled during the actual application described in the next chapter.

During foam recirculation in the contaminated vessel, foam is injected continuously at a rate R into a vessel with a volume V. For what value of R will it overflow from the top of the vessel with a bulk factor f'? The filling time t_f must be less than the time required for the foam to reach f':

$$t_f < t'$$

where $t_f = V/R$, f' becomes R/L' and t' is determined from Eq.(2) as follows:

$$t' = \frac{t_{1/2}}{0.693} Ln \frac{f'}{f_0}$$

Hence:

$$\frac{R}{V} > \frac{0.693}{t_{1/2}} \cdot \frac{1}{\text{Ln}\frac{f'}{f_o}} \qquad \text{(Eq.3)}$$

Bradley (1) reports that the maximum filled volume $V_{max} < 2\, t_{1/2} \cdot L \cdot f = 2\, t_{1/2} \cdot R$, or:

$$\frac{R}{V} > \frac{1}{2t_{1/2}}$$

The correspondence with Eq.(3) is obvious if:

$$Ln \frac{f'}{f_0} = \frac{1}{2 \times 0.693} \; i.e. \; \frac{f'}{f_0} = 2.06$$

(for example: $f' = 30$ and $f_o = 14.5$).

This is the minimum flow rate R corresponding to the volume V to be filled, for which the foam will overflow with a bulk factor f' that will allow it to be recycled, assuming the foam solidity properties are maintained as long as $f' < 40$.

If f_0 is limited to 7 to ensure satisfactory decontamination of the low points in the vessel, then:

$$\frac{R}{V} > \frac{0.693}{\text{Ln}\frac{40}{7}} \cdot \frac{1}{t_{1/2}} = \frac{1}{2.5\, t_{1/2}} \qquad \text{(Eq. 4)}$$

This is very similar to the formula in Ref. (1). Note that the maximum filling time may also be determined from Eq.(4):

$$\frac{R}{V} = \frac{1}{t_f}$$

Hence an easily applicable formula:

$$t_f < 2.5\, t_{1/2} \qquad \text{(Eq. 5)}$$

As foam fills the component it stratifies from the initial bulk factor f_0 to the overflow bulk factor f' (Fig. 3). The liquid fraction L is assumed to be drained continuously from the component. For simplicity, it is assumed that the foam is unaffected by pressure variations due to the weight of the overlying foam; this hypothesis is acceptable for a preliminary approximation.

If $\overline{L}$ is the liquid holdup present as foam in the component, then:

$$\overline{L} = \frac{V}{\overline{f}} \qquad \text{where } \overline{f} \text{ is the mean bulk factor}$$

$$\overline{f} = \frac{f_o}{t'} \int_0^{t'} e^{kt}.dt = \frac{f_o}{k \cdot t'} (e^{kt'} - 1)$$

$$\overline{L} = V \frac{\text{Ln}\frac{f'}{f_o}}{f' - f_o} \qquad \text{(Eq. 6)}$$

Examples of $\overline{f}$ are shown in Table I.

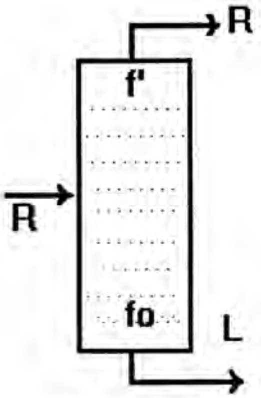

Fig. 3. Foam stratification.

Coalescence on Vessel Walls

Wall coalescence intuitively appears to have a decisive effect on decontamination. By renewing the liquid film in contact with the wall, coalescence eliminates the boundary layer phenomenon and allows fresh molecules to react with the contaminated wall.

In order for a liquid to wet a solid surface, the adhesion energy must be greater than the liquid cohesion energy. This may be expressed as follows (5):

$$\gamma_{sv} > \gamma_{lv} + \gamma_{sl}$$

TABLE I

Coalescence Data

Half-life (min)	f_o	f'	Max filling time (min)	Minimum R/V	Actual flow rate (m^3h^{-1})	Actual filling time (min)	$\bar{f}$
Valve (5 m³)							
6.00	5.00	35.00	16.85	3.56	20	15	15.4
8.00	5.00	35.00	22.46	2.67	20	15	15.4
14.00	8.00	35.00	29.82	2.01	17	18	18.2
Heat Exchanger (27 m³)							
14.00	6.00	35.00	35.63	1.68	60	27	16.4

where γ_{sv} is the surface tension at the solid-vacuum interface, γ_{lv} the surface tension at the liquid-vapor interface, and γ_{sl} the surface tension at the solid-liquid interface.

This equation is easily verified for common metals requiring decontamination (e.g. steel, copper, aluminum). However, the metal surface is usually covered by a greasy film from one micrometer to several millimeters thick, causing γ_{sl} to drop significantly. The addition of a surface active agent can reduce the solid-liquid surface tension by 50%: the hydrocarbon chain is absorbed by the greasy film, while the polar group remains in the aqueous phase.

However, it is often noted that the foam persists longer in contact with the wall than in the foam mass: the foam tends to "adhere" to the wall, while it is destroyed in the center. The Plateau theory indicates the value of Plateau suction in Plateau capillaries within the foam:

$$\Delta P = \frac{2\gamma_{lv}}{r_B}$$

where r_B is the mean bubble radius and γ_{lv} is the surface tension. By comparison, in contact with the vessel walls, the presence of a flat surface results in less effective drainage by reducing the Plateau suction to:

$$\Delta P = \frac{\gamma_{lv}}{r_B}$$

The controlled and repetitive addition of small quantities of C5 volatile alcohol has been observed to increase the relative wall drainage. This may be attributable to the fact that the volatility of the alcohol increases the internal pressure in the bubbles, causing them to burst more readily on contact with the wall. This method was adopted for the applications discussed below.

APPLICATION TO LARGE VALVES WITH COMPLEX INTERNAL GEOMETRY

A special dolly (Fig. 4) was designed on the basis of the preceding theoretical considerations to test the

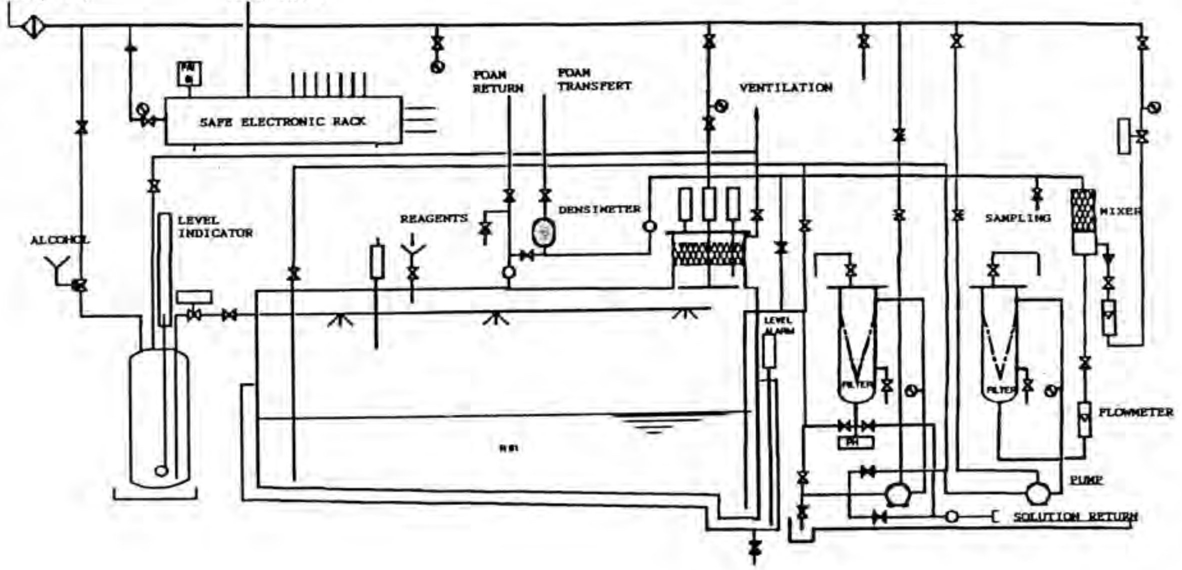

Fig. 4. Schema of decontamination dolly.

decontamination of a series of large valves with complex internal configurations. The valves were made of ordinary low-carbon steel and were taken from the secondary system of a gas-*cooled reactor* undergoing decommissioning.

The test dolly includes a 4 m^3 rectangular tank inside a *safety vessel*, a liquid injection pump in the foam generator, and a scavenge pump to recover the liquid overflow from the valve. The pumped fluids are filtered to 20 μm and 100 μm. The foam is produced in high efficiency mixer by injecting compressed air. The foam density is monitored in both the feed and recovery pipes, and the final off-gas stream is monitored. Chemical injection systems may be set up as required. Containment and a negative pressure gradient are provided for the entire unit as standard practice in contaminated environments. The complete dolly is light enough to be transported to the decontamination site. Biodegradable surface-active products are used in the process so the liquid waste stream can be recovered without difficulty by the onsite Liquid Waste Treatment Station, where the radionuclides are removed by coprecipitation.

Procedure

The component is first washed with 3N sodium hydroxide using an unctuous foam with a minimum half-life of 8 minutes and a low bulk factor between 5 and 8. This operation may last several hours. On these valves covered with a mixture of lubricating oil and dust particles, the process is more effective at higher temperatures (30-40°C).

The valve is then decontaminated for 3 to 4 hours with a sulfuric and phosphoric acid mixture well suited to surface etching of ferritic steel, applied as a foam with similar properties. It is then rinsed with a slightly alkaline foam to obtain a relatively neutral final pH on the valve walls, and thus prevent further surface damage.

TABLE III

Liquid and Radioactivity Balances

Phase	Liquid Volume (liters)	Dissolved Activity (MBq)
Alkaline	600	2
Acid	628	4.2
Rinse	605	0.3

Operating costs are optimized by using the same reactants twice to decontaminate two identical valves during the same operation. The reactants are mixed to obtain a final pH of 5 to 6 before they are pumped to the liquid waste treatment facility.

Example

Table II summarizes the results obtained with a valve 1.2 m in diameter weighing 5 metric tons, with a unit volume of 3.3 m^3 and a total unit surface area of 24 m^2. The table indicates the total radioactive values (mainly ^{60}Co and ^{137}Cs) measured with a surface probe to assess the total contamination and a cotton smear test to evaluate the free contamination before and after treatment. After the foam process, most of contamination measurements were under the detection tresholds.

TABLE II

Radioactivity Measurements ($Bq \cdot cm^{-2}$) inside Valve 31

Reference Location	Before Treatment		After Treatment	
	Smear	Probe	Smear	Probe
1	15	12	<0.2	<1
2	15	15	<0.2	<1
3	<0.2	<1	<0.2	<1
4	<0.2	<1	<0.2	<1
5	25	35	<0.2	<1
6	6	12	<0.2	<1
7	4	8	<0.2	<1
8	45	20	<0.2	<1
9	65	30	<0.2	<1
10	6	<1	<0.2	<1
11	40	25	<0.2	<1
12	<0.2	<1	<0.2	<1
13	50	30	<0.2	<1
14	<0.2	<1	<0.2	<1
15	20	10	<0.2	<1
16	<0.2	<1	<0.2	<1

The liquid and radioactivity balances are indicated in Table III.

An additional 0.6 MBq was retained by the filters, as estimated from dose rate measurements. The average dissolved metal thickness was 10 μm; an average of 15 $\mathrm{Bq \bullet cm^{-2}}$ was therefore removed from the two valves. The residual contamination did not exceed 0.1 $\mathrm{Bq \bullet g^{-1}}$, allowing the valves to be melted down for recycling in an approved steel mill.

Similar results have been obtained on valves 1.6 m in diameter with a unit volume of 7 $\mathrm{m^3}$.

The same dolly is expected to be used to decontaminate a heat exchanger with a surface area of 1000 $\mathrm{m^2}$ and an internal volume of 27 $\mathrm{m^3}$.

CONCLUSION

On the basis of a theoretical analysis of recirculating foam, a dolly was designed and built for local decontamination of large components with complex shapes. Suitable decontamination procedures involving four hours of foam recirculation have yielded very low residual contamination levels, allowing the metal to be melted down in an approved steel mill and reused under control.

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