

Chapter 6

Design of a Micellar-Based Technique for Recycling Liquid Scintillation Glass Vials

N. G. L. Harding and J. Dixon

University of Glasgow, Department of Pathological Biochemistry, Royal Infirmary, Glasgow, Scotland

INTRODUCTION

Liquid scintillation vials have become, with progressive financial stringencies, an item to be erased from the budget of a laboratory provided an acceptable technique could be developed for recycling used glass vials.

The current cost of glass vials is around ten vials per dollar; so an average research programme where vial consumption is of the order of several thousand per year, discards about one thousand dollars-worth of vials per year. In order to excise this budgetary loss, a reliable recycling technique was required for cleaning a used vial and placing it in circulation again. A prerequisite was that an investigator should not need to check the background count of the used vial in the required isotope channel. Once a certain count-independent method for achieving this had been established it should prove possible to introduce a standard recycling procedure into the laboratory with ready and unanimous acceptance among users of a wide variety of isotopes.

At the outset we anticipated that a major difficulty would be to convince users that the following criteria were acceptable without actually counting each recycled vial:

- (i) the background count is not increased by the washing procedure;
- (ii) chemiluminescence characteristics of the vial remain unchanged;
- (iii) the efficiency of the isotope assay procedure is constant;
- (iv) external standardisation and internal quench estimates remain unaffected;
- (v) improperly washed vials are readily detected and rejected without the need for counting.

PRELIMINARY SURVEY

When we began this programme some years ago there was little detailed information available about the structural relationships between the glass in a scintillation vial and the 'average' residue left after a used vial has been emptied of the majority of its radioactive contents. To investigate this we used a standard glass vial 'contaminated' with a residue of naphthalene-based scintillator containing a variety of inert (non-radioactive)

'contaminant' molecules. At the same time, by using this standard system we were able to investigate available commercial washing reagents in a preliminary study. Results can be summarised as follows:

- (i) Commercial washing powders and liquids were variable in their ability to remove standard contaminants from the glass wall. Some were unable to remove precipitated protein; others could not remove the hydrophobic components of the scintillator; others did not remove all traces of radioactive labels introduced into the 'contamination'.
- (ii) Some of these washing preparations were at a grave disadvantage in that they left residues upon the vial wall. These were later found to interfere with internal quench estimation procedures. Some preparations yielded such ingrained deposits that even subsequent machine washing and acid-soaking failed to regenerate the original optical surface.
- (iii) With some materials there was not a consistent spectrum of cleaning ability: those found not to be satisfactory for hand washing of the vials were also unsatisfactory for machine washing; other materials able to remove certain types of contaminant by hand washing also proved unsatisfactory in the washing machine.

This preliminary study of commercially available preparations confirmed the need for a fundamental investigation of the problem rather than an attempt to modify an existing preparation to give a simple one-step vial-recycling technique.

This basic approach had several advantages, in particular enabling complete control over the composition of washing reagents and the structure-contaminant relationship of ingredients of a scintillator and samples. Accordingly, the programme started at a fundamental level and aimed to produce a simple one-step washing procedure for use in the laboratory.

As a first step we investigated simple procedures for removing standard oleaginous contaminants (Table 1), and confirmed that neither simple anionic nor cationic systems were individually capable of performing the required tasks.

Table 1. Effectiveness of different treatments at removing a standard greasy soil from scintillator vials.

Isopropanol, 2 min	Oily residue and fingerprint remains	Slight wetting
Isopropanol, 2 min + ultrasound	Cleaning complete	Slight wetting
Alkylsulphonate ion	Partial cleaning	No wetting
Quaternary ammonium ion	Partial cleaning	No wetting

Vials were treated with a standard scintillator residue and dried. Soiled vials were treated as shown, at room temperature, and examined for residues, fingerprints and wetting by distilled water.

An impressive finding was that although ultrasonication in isopropanol proved effective for these contaminants, it was ineffective for rapid removal of all scintillant complexes. In addition, these preliminary experiments confirmed that there was an empirical relationship between 'wettability' of the washed surface and the extent of cleaning in the test system.

An important question arising from this first project was whether there was a relationship between the structure of anionic and cationic surfactants and their ability to remove contaminants from the glass wall. To avoid an essentially boundless survey of compounds the following principles directed our attention to several classes of reagent for further study.

STRUCTURAL BASIS OF CONTAMINATION

Two factors set the rate and determine the extent of contamination, namely the glass wall and the impinging molecules.

The anatomy of the glass wall is a subject of some doubt. The precise composition of the wall will vary with the type of glass used, and in turn this will determine the solution properties of the glass. For example, silica, borosilicate, aluminosilicate and high-lead glasses have excellent water resistance, in contrast to the poor water resistance of high-alkali, phosphate and borate glasses. For the purposes of the present paper, it is useful to assume that the wall consists of an outer, hydrated layer and an inner, structural layer based upon the tetrahedral silicon atom. Normally the hydrated layer of the wall will have hydroxyl or alkoxy moieties in contact with the internal fluid layer. These are illustrated in Fig. 1.

The nature of the impinging molecules is complex as diverse types and amounts find their way as samples and scintillators into a scintillation vial. Several factors thereby complicate the chemistry of contamination: labelled molecules introduced into glass vials will react at varying rates with the vial wall; sample preparation and counting may be performed at a variety of temperatures, both above and below ambient resulting in varying rates of interaction with the vial wall; finally, as the composition of the vial wall may vary according to the batch of the glass, contaminant properties may not always be consistent.

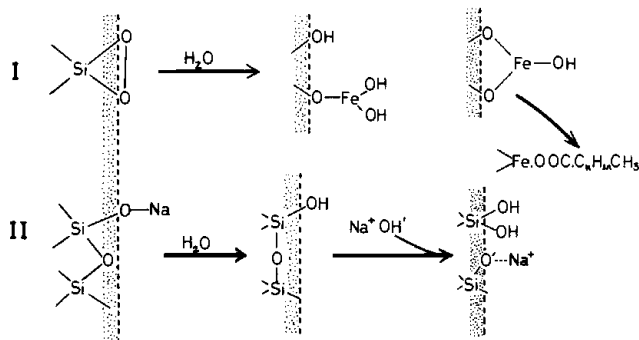


Fig. 1. Glass vials: hydration and derivatisation of wall silicon atoms. The shaded area refers to the glass wall.

Despite this spectrum of difficulties, contaminants are able to bond to the vial wall by rather a restricted number of mechanisms. As a result it will be predicted that any failure to remove a contaminant by a properly designed reagent could be ascribed to a rate-dependent failure to attack a large lump of contaminant adhering to the glass wall rather than to an aberrant physicochemical interaction. Clearly, any washing reagent must therefore be able to reverse the common modes of contaminant bonding. For convenience we have classified the three modes of contaminant bonding, though other and more complex classifications come to mind. As an operational tool the following classifications have proved fundamental to the design procedure.

Covalent bonding

This is likely to be an unusual mechanism in the formation of contamination complexes with the glass wall. Nevertheless, it has some important aspects, of which one is illustrated in Fig. 1. A reaction between iron or manganese and the wall hydrate layer leads to covalent bonding of an iron atom in alternative modes. Coupling to both oxygen atoms of the silicon tetrahedron forms a ferric hydrosilicate in which a reactive hydroxyl group is attached to the iron atom. This hydroxyl can in turn be displaced by other residues, a carboxylate derivative is shown in Fig. 1, to yield large metallo-organic complexes. The stains which can be left on glass surfaces by iron-rich solutions are familiar, and those who have tried to remove them will know that they are removed with appreciable difficulty. This type of contamination is important in two ways: first, the iron atom, if radioactive, presents a radioassay problem for the cleaning process; second, if non-radioactive, the iron is troublesome in acting as an intermediate for formation of metallo-organic contaminants.

Ionic processes

For convenience, both ionic bonds and ion-exchange processes can be considered together. An underlying mechanism is illustrated in Fig. 1. In the presence of hydroxyl ion, the wall hydrate layer is susceptible to attack at the oxygen atom which joins two silicon atoms. This structure is cleaved, leaving an anionic oxygen atom able to partake in ionic processes. At the same time, hydroxyl density in the wall hydrate layer appears to increase as the remaining silicon atom sequesters an hydroxyl residue. Thus, as the wall gains negative charge, counter-ions would be expected to be absorbed from aqueous solutions. Anions such as borate, carbonate and phosphate are known for their ability to adhere to glass, though fortunately dissociation is relatively simple under non-etching conditions.

Etching is an important part of the contamination procedure, and as already described seems to be a common feature of many washing solutions. In such cases etching is often accompanied by a deposition of virtually insoluble white material. The dependency of vial erosion upon hydroxyl ion is shown in Fig. 2, which also shows the erosion of a commercial laboratory washing reagent and compares these with a current vial recycling reagent specifically designed. The etched surface of the vial is greatly increased in surface area and contains pits for entrapment of contaminants by the various binding mechanisms. Reproducible low-background counts during vial recycling has been a guiding principle requiring a washing procedure free from the 'etching pitfall'.

Hydroxyl ion is very important in the physical chemistry of the glass surface owing to the ability to break bonds in the hydrate layer and create etched zones.

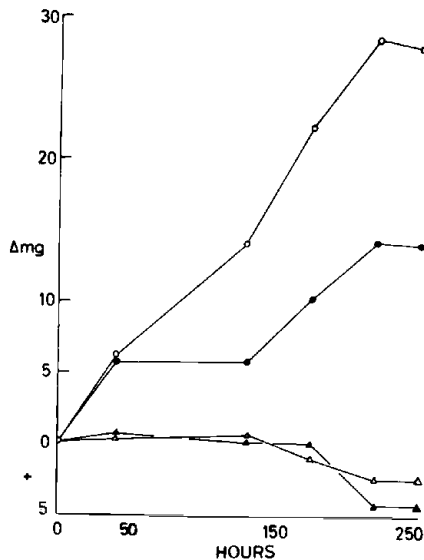


Fig. 2. Erosion of scintillator vials by hydroxyl ion at room temperature. Note that loss of weight (mg/vial) on the ordinate is expressed above zero and weight gain below. O = pH 10.5; ● = pH 8.5; △ = aqueous solution of a micellar reagent at pH 4.0 and at tenfold concentration pH 4.0 (▲).

In the presence of high concentrations of hydroxyl ion, a progressive erosion of the hydrate layer would be expected as the process shown in Fig. 1 extends into and beyond the normal hydrate layer of a glass wall. This is conveniently termed etching. During these studies we also observed that those vials etched with certain preparations during washing became unsuitable with some quenching reagents. This problem is described in further detail below.

Yellowing of quenched standards

A surprising finding during the preliminary screen of available products was that of interference between the washing procedure and subsequent performance of the vial when used with quenched standards. This is illustrated in Table 2, which shows that the phenomenon was restricted to a particular product (W3, W3A) in the presence of carbon tetrachloride. A visual yellowing of the scintillator eventually occurred in these vials and was associated with a progressive fall in light output of the scintillator. The phenomenon was not dependent upon the presence of tritium and did not occur with acetophenone-quenched scintillator.

Electrostatic bonding mechanisms

For convenience a variety of different mechanisms is classified within this general heading, as in practice similar approaches would be used to dissociate material bound to the glass wall by this type of mechanism. An example is the vial which has been allowed to dry when containing a naphthalene-rich scintillator. A familiar incrustation of naphthalene and scintillator is obtained, and is not easy to remove from a glass surface by aqueous solvents. However, addition of toluene rapidly results in dissolution of the complex. There are a series of complicated interactions which describe such a deceptively simple washing procedure and it is beyond the scope of the present paper

Table 2. Yellowing of quenched standards by washed vials.

Wash reagent	NE 233	+CCl ₄	+ C ₈ H ₈ O
W1] ³ H omitted
W2			
W3		y	
W3A		y	
W1] Plus 10 ⁵ dpm ³ H
W2			
W3		y	
W3A		y	

Standard glass vials were washed with a variety of commercially available washing reagents (W2, W3, W3A) and with a micellar wash reagent (W1) based upon the polyethersilane core mentioned in the text and legends to Figs. 9 and 10. The dried vials were dosed with a standard scintillator (NE 233) and left unquenched or dosed with the quenching reagents shown. Identical groups of vials were left without added radioactivity or contained the amount of tritiated toluene shown. Washing reagents W3 and W3A produced yellowing of the scintillator fluid upon standing in daylight for a few days (y).

to consider these in detail. It is sufficient here to mention that non-polar solvent-solute interactions are a fundamental design requirement for a washing procedure of the type specified. The problem is to obtain such an effect in an aqueous environment.

In terms of the bulk of interacting material within the vial this general class of mechanism probably predominates, and may of course be superimposed upon a covalent mechanism. For example, the contaminant iron atom shown in Fig. 1 may eventually terminate in a long hydrophobic alkyl chain which would then prove a powerful attractant by means of hydrophobic interactions with other molecules. This illustrates the problem of developing a suitable strategy for dissecting such mixed-bond complexes. A reagent capable of dissociating the hydrophobic complex would not necessarily cleave the covalent bond, though once the covalent bond had been cleaved, the complex could be removed intact provided the solubility characteristics were appropriate.

DESIGN PRINCIPLES

Both the preliminary survey of existing materials and a subsequent theoretical consideration of the problem of mechanisms of contamination yield the following points summarised for the design procedure:

- (i) many commercial cleaners give rise to time-dependent etching and leave white deposits in the glass wall. These later interfere with quench techniques (Table 2).
- (ii) hydroxyl ion is an important determinant of certain mechanisms which bond molecules to the glass wall (Fig. 1).
- (iii) etching of the glass wall depends upon the concentration of hydroxyl ion and other anions and is to be avoided (Fig. 2).
- (iv) contaminants are bonded to glass by a variety of mechanisms involving

- covalent, ionic and electrostatic forces.
- (v) both polar and non-polar solvent-solute interactions are required for effective removal of contaminants.
 - (vi) a relationship between wetting of a glass surface and cleaning has been established for long-chain aliphatic compounds.
 - (vii) this cleaning was only partial when both anionic and cationic derivatives were tested individually as surfactants (Table 1).

A MICELLAR REAGENT

The preliminary work defined some reagent specifications: acid cleaning alone was inadequate unless prolonged; base washes produced etching; simple non-ionic, anionic and cationic reagents were ineffective to a varying degree unless reinforced by thermal or sonic energy.

A complex reagent was therefore required, compatible with both aqueous and oleaginous systems, capable of dissociating contaminants affixed by all classes of bonding mechanism and free from corrosive effects upon the glass wall.

Micellar theory offered the required approach and enabled us to consider a further question: could the vial wall be protected against subsequent contamination by incorporating into the micelle a reagent itself capable of reacting covalently with the wall? Such reagents would be organosilicon derivatives or metallo-organic compounds.

The concept of protection of the vial wall

In principle, the idea is simple. A covalent derivative of the vial wall offers the opportunity for steering properties of the glass surface towards those desirable on both theoretical and practical grounds. Conversion of heterogeneous reacting sites in the glass to sites with homogeneous properties is possible in several ways. The problem is to decide which properties to emphasise. A wall with hydrophobic properties is obtained by either of two principal classes of reaction, illustrated in Fig. 3.

Scheme I (Fig. 3) indicates that in the simplest conversion an homogeneous set of hydroxyl groups is generated in the hydrated layer. The attendant problems of so doing have already been inferred. Scheme II shows the use of an organochromium

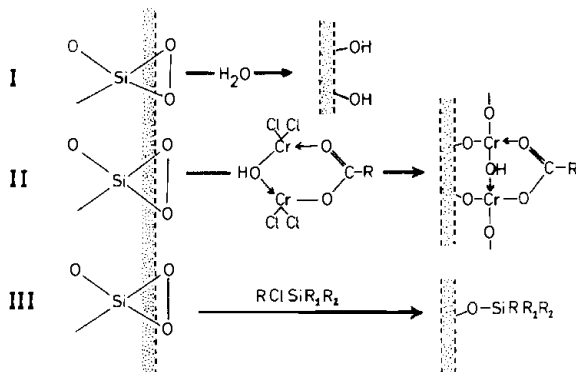


Fig. 3. Principal classes of derivatives of wall silicon atoms. These derivatives are referred to in the text.

derivative as an intermediate coupling agent. Methacrylic acid co-ordinated to chromium yields films with high flexural strength.¹ For our purpose, methacrylic acid was disadvantageous in view of its further reactivity, although published evidence confirms that the chromium-oxygen terminals become integral with the glass structure. Rather than undertake a programme based upon chromium co-ordinate chemistry, the organosilicon approach commended itself. Scheme III illustrates the manner in which a chlorosilane derivative reacts with the glass wall. In this way alkyl silicon polymers can be formed on the glass with covalent linkage. Such chemistry has a long history dating from Johannson's work in 1938 on the interaction of organochlorosilanes with glass for improving adhesion of lacquers.²

Silicon alkyl derivatives have the advantage of optical transparency at the required wavelengths and so seem ideal. Yet by emphasising the adhesive properties of such films, Johannson's work apparently contradicts the present purpose. Resolution of the paradox lies in the properties of the organic substituent. For example, aminopropylsilane and methylsilane derivatives exhibited different properties as shown in Fig. 4 and Table 3. In these experiments vials treated with different procedures (W1-S2) were lavaged with $^{14}\text{CO}_3^{2-}$, rinsed and counted after addition of micellar scintillator. Compounds W1-W3 are commercial laboratory washing detergents, which give considerable and apparently unpredictable scatter in their ability to elute carbonate. When the glass is treated with a methylsilane derivative (S1), excellent elution of carbonate occurs, with good clustering of individual results. As expected, introduction of an anion-exchange function, using a ω -aminopropylsilane, resulted in retention of some carbonate after rinsing (S2).

Having conferred hydrophobic properties upon the wall, it is important to confirm that the reagent is compatible with a range of scintillators (Table 3). In general, appropriate silicon derivatives of the vial wall do not affect the scintillation process. Indeed, a scintillator purchased from one source (Scintillator 2) started with a very high background count in the tritium channel which was diminished after derivatisation of the glass, though the precise reasons for this effect are unknown.

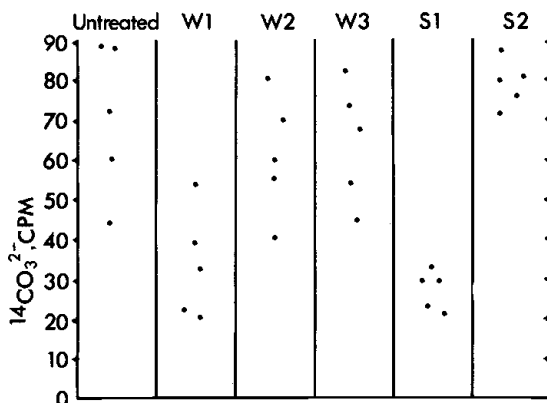


Fig. 4. Retention of carbonate by washed and derivatised standard glass vials. Vials were washed with a micellar reagent (W1), with commercial reagents (W2 and W3) or derivatised with a methylsilane (S1) or γ -aminopropylsilane (S2). Treated vials were rinsed under standard conditions with $^{14}\text{CO}_3^{2-}$, distilled water, and counted following addition of a micellar scintillator.

Table 3. Scintillator-wall interaction after hydrophobic wash.

Scintillator	PRE-WASH				POST-WASH			
	cpm in channel		Efficiency		cpm in channel		Efficiency	
	^3H	^{14}C	$^3\text{H} + ^{14}\text{C}$		^3H	^{14}C	$^3\text{H} + ^{14}\text{C}$	
1	19	12	29	41.7	20	14	34	40.8
2	115	13	128	44.9	63	13	86	40.6
3	22	14	36	43.3	24	10	36	43.7
4	19	12	31	43.3	19	8	28	44.1
5	27	14	41	41.6	33	9	45	41.5
S1	23	13	32	42.1	20	14	36	41.9
S2	22	12	29	40.9	24	10	33	42.0

Scintillators 1-5 refer to commercially available solutions purchased from standard suppliers, except 1, which was a gift. Vials were filled with scintillator and counted (PRE), contaminated with ^3H , washed with a micellar recycling reagent and counted in the channels shown, using ^3H -toluene as an internal standard. S1 and S2 are referred to in the legend to Fig. 4.

The silane reagent persisted through successive use cycles of the vial so the next problem to solve was the introduction of a chlorosilane derivative at the appropriate moment of the wash cycle. Micellar theory indicates that this should be possible provided critical points in the carriage and unwrapping of the package are identified so that sensitivity of the procedure to solution conditions can be minimised by an appropriate choice of chlorosilane derivative and carrier micellar molecules.

MICELLES AS REAGENT CARRIERS

Critical stages in carriage of the reagent to the vial wall become apparent from further theoretical considerations. In structural terms one can envisage linear micelles (Fig. 5) protecting their cargo of silane and delivering it by ordered or random interaction with the glass surface. In contrast, spherical micelles do not have the opportunity for multiple linear contact. Figure 6 illustrates the manner in which a spherical micelle could be constructed to deliver its cargo, either from a reservoir in the core of the micelle or from a coaxial domain in the ternary model. A further point, not illustrated in these models, is the need to interact with the contaminants in the wall prior to initiating deposition of the hydrophobic coat. To some extent this directs choice of the micelle type towards complicated structures so that the correct reaction sequence can be maintained.

Unlike the enzymologist we at least have the opportunity wilfully to design a reaction mechanism so that the problems can be clarified by considering the kinetics of a hydrophobic deposition sequence. Figure 7 shows a micellogenic molecule (A) coaxially protecting a core reagent (B) in solution (S) within a boundary (G). In this model, there are three abortive dead-end complexes, AG, AB and BS, the desired complex being BG. Assuming that G represents a single site capable of reacting with A, B and S, then BG could be formed in a linear displacement sequence of the type shown in Fig. 7. The forward velocity, V_5 , becomes critical in relation to the rate of reaction of B with S. If the latter greatly exceeds the former, then inadequate amounts of the desired complex BG will be formed.

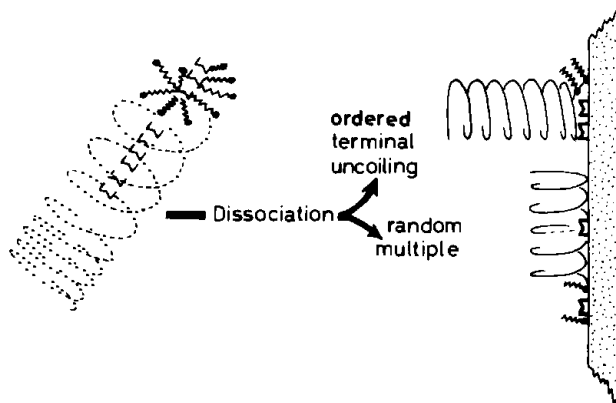


Fig. 5. Models of linear micelles acting as reagent carriers. Σ = a derivatising reagent; as with Fig. 1, the shaded area refers to the vial wall.

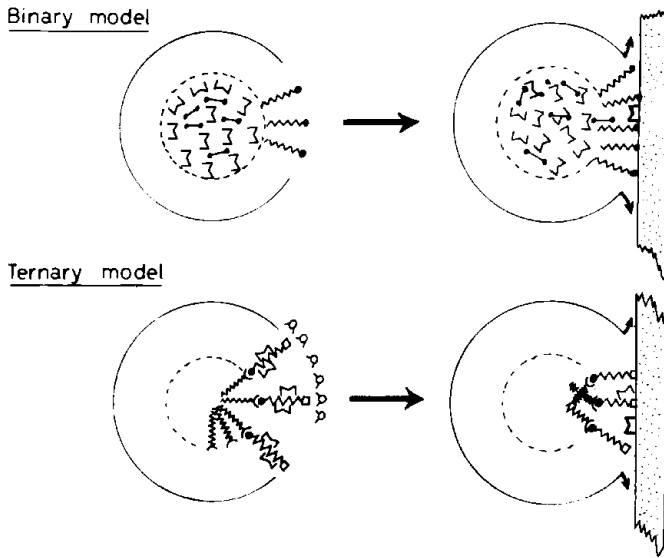


Fig. 6. Models of spherical micelles acting as reagent carriers. For simplicity the micellar and reagent molecules are depicted in a similar way to those in Fig. 5.

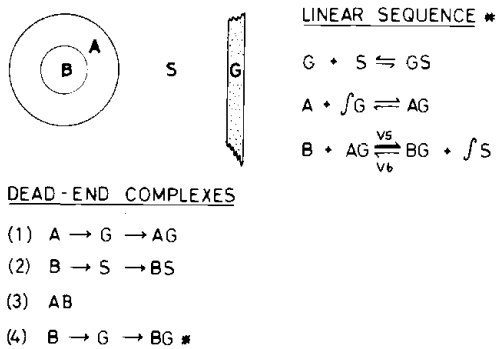


Fig. 7. Kinetic models of reaction between a micellogenic molecule (A), core reagent (B) and glass substrate (G) in a solution (S). Unwanted and wanted (*) dead-end complexes are shown together with a linear (*) sequence leading to the wanted complex.

Another restraint imposed by the model is the reaction sequence, whereby if A and S interact with G the resultant complexes will dissociate in the presence of unreacted B. This sequence has the advantage that A and S could perhaps be used to fulfil the other function required of the reagent, namely that of facilitating removal of unwanted contaminants.

A FUNCTIONAL COMPLEX

Formulation of a micellar complex depended upon resolution of two central issues: satisfactory detergency of contaminants and ability to deliver a reactive molecule to exposed residues on the glass wall. The former was solved by studies outlined previously whilst the latter proved to be especially complicated by virtue of the factors mentioned in the previous section. By finding that polyglycol ethers formed complexes with silane reagents, a skeleton became available upon which a complex micellar reagent could be constructed with the desired properties.

Such a complex has the desired properties as seen from the following studies. Figure 8 shows for two scintillators that the background count (A1, B1) of new vials clusters at a satisfactory low value after a single wash. The efficiency of these vials is at the expected value for tritium (A2, B2). After rewashing, the background (A3, B3) and efficiency (A4, B4) are essentially unchanged. During these experiments a group of vials was isolated with a moderately high background count (C1) but essentially normal efficiency (C2). After wash-recycling, the background fell to the expected value (C3) and the efficiency remained at the previous value. The reason for these unused vials clustering originally at the relatively high background value is not known.

To examine the comparative effectiveness of the complex reagent, groups of 16 vials were selected for a range of background counts from 10–70 counts min^{-1} in the tritium window (Fig. 9). These vials were recounted after washing with a recycling reagent (W1) and with standard commercial laboratory glassware washing preparations (W2 and W3). Although all reagents tended to reduce the background count, the

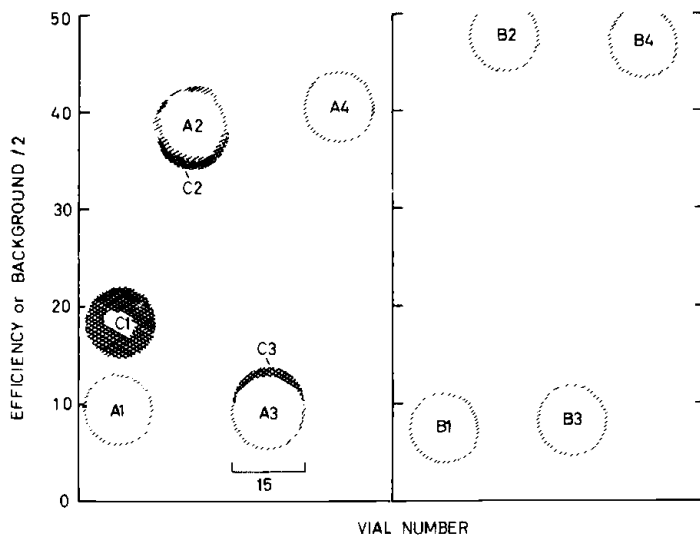


Fig. 8. Clustering of background counts and efficiency of washed vials. A batch of standard glass was selected for background counts in the region of 10 counts min^{-1} (A and B) or 18 counts min^{-1} (C) in the ^3H -window, treated as described in the text with a silane complex during washing, and finally assayed with a micellar scintillator (A and C) or hydrophobic, xylene-based scintillator (B).

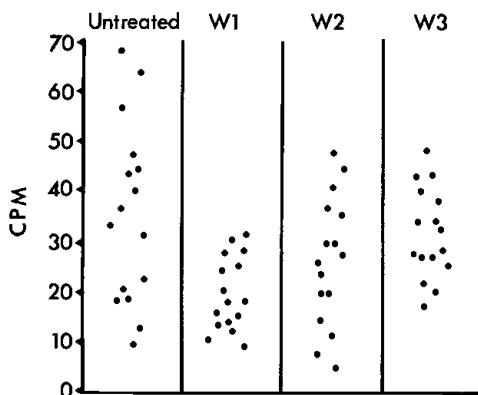


Fig. 9. Reduction of intrinsic ^3H -background of used vials by washing. Each point represents one used vial. Vials were assayed with a micellar scintillator after extensive water rinsing (untreated), washing with a micellar washing complex (W1) and standard laboratory washing solutions (W2 and W3).

recycling yielded the lowest counts and the most compact clustering. These properties are conserved over at least a tenfold range of reagent concentration during washing (Fig. 10), where it is also seen that the kinetics of this process are rapid at all concentrations of the reagent. It would be expected that erosion profiles of the vials show minimal erosion by a recycling reagent of this type. This is confirmed in Fig. 2, where groups of vials were soaked for up to 250 h. Vials soaked in sodium hydroxide show intense erosion, those soaked in a standard laboratory glassware washing detergent also show a surprising amount of erosion. In contrast, those soaked in a recycling reagent (NE555) show little change, perhaps a slight gain in

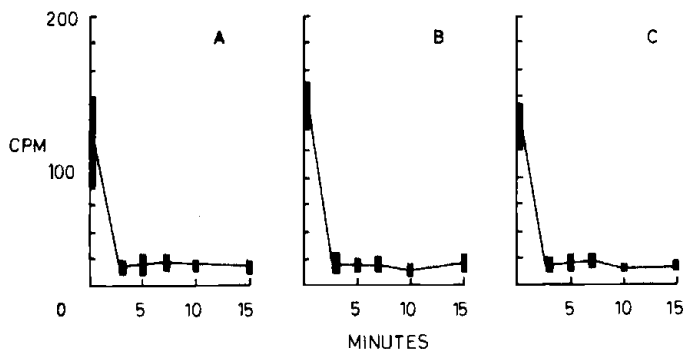


Fig. 10. Compact clustering of ^3H -background counts over a tenfold range of concentration of a micellar wash reagent. Used vials were washed in batches, for the times shown, in 1% (w/v;A), 5% (w/v;B) and 10% (w/v;C) aqueous solutions of a polyethersilane micellar complex washing reagent. Undried vials were assayed with a micellar scintillator in the ^3H -channel.

mass, by the end of the experiment. Encouraged by these findings we have been able to expand the properties of these micellar vial recycling reagents by formulation, along the lines described, of polynomial micelles containing several classes of molecule each conferring a specific function upon the micelle for use in separate phases of a single washing cycle. This not only permits specific elution of an isotope from a vial, but also enables desirable mechanical properties to be conferred upon the glass, such as scratch resistance.

ACKNOWLEDGEMENTS

We express gratitude to colleagues who have willingly and generously supplied many of the molecules studied in this programme. Nuclear Enterprises Ltd. are gratefully thanked for support of the programme.

REFERENCES

1. J. V. P. Torrey, *Modern Plastics* **30**, 154 (1952).
2. E. G. Rochow, *An Introduction to the Chemistry of Silicones*, 2nd Edition, Chapman and Hall, London, 1951.

DISCUSSION

C. McEvoy: My research shows that efficient rinsing in cold tap water, using a device of my own invention, is a sufficient method of decontaminating most vials. Are you suggesting that this scintillator should be used because of its easy decontamination in all cases or only in specific applications in which radionuclides are known to be strongly adsorbed onto the vial wall?

N. G. L. Harding: In our carbonate elution assay we have shown that elution of carbonate is variable, particularly in etched vials. With 'fresh' vials, elution is also variable, although with properly washed vials I find that a distilled water rinse will elute carbonate. The type of scintillator is also important, for we find that a vial used with a micellar scintillator is easier to wash than when used with non-micellar types, particularly if proteinaceous samples are used. Finally, chemical modification of the vial wall enables us to have a set of vials in the laboratory with known properties of a uniform minimal capacity for isotope adsorption.

B. W. Fox: We use a commercial sonicator successfully in routine washing of vials -- with minimal incidence of contamination.

N. G. L. Harding: I agree that this is an extremely effective procedure for washing a vial, particularly if one sonicates in the presence of isopropanol as we show in the paper. However, in a multi-isotope, multi-procedure laboratory, the problem can be complicated, particularly if users presoak vials before sonic washing. Large sonicators are expensive and so we decided to develop a washing procedure, essentially independent of a supply of sonic and thermal energy. At the same time the opportunity presented itself for coating a vial wall to obtain an homogeneous set of vials with the desired properties.

B. Bakay: We use oven-cleaned vials satisfactorily in our laboratory.

N. G. L. Harding: I agree that oven cleaning is effective for removing organic isotopes though I am not sure of the long-term effects upon recycling, and the procedure is expensive to run so one might do equally well to purchase new vials.

B. W. Fox: This procedure only applies to organically found isotopes, e.g. ^3H and carbon.

B. Bakay: Yes.

N. G. L. Harding: Metal residues will combine with heated glass.