JOURNAL OF APPLIED ELECTROCHEMISTRY 10 (1980) 789–797

Wetting properties of the sulphur electrolyte in sodium–sulphur batteries[*]

G. J. JANZ, R. M. MURPHY[†], W. A. SPIAK

Cogswell Laboratory, Rensselaer Polytechnic Institute, Troy, New York 12181, USA

Received 6 February 1980

In the sulphur electrode of sodium–sulphur cells, carbon surfaces (generally as yarns or felts) are used as current collectors. During the charging cycle, oxidation of the molten polysulphides results in sulphur as a second (immiscible) phase. If sulphur has preferential wetting properties, an insulating sulphur layer may be established. A technique for wetting measurements with the molten sulphur electrolyte at ~ 330° C is described. The wetting properties of molten sulphur and molten polysulphides were investigated with both untreated and metal oxide treated carbon surfaces as substrates. Results are reported for felts with Fe, Ni, Co, Cr, Al (as oxides), and with H_2O, NaOH, Na_2SO_4 and $Na_2S_2O_3$ as impurities in the fluid electrolyte.

1. Introduction

The salient components of the sulphur electrode are a fluid electrolyte (molten Na_2S_x, or a mixture of two fluids, one polysulphide-rich and the other sulphur-rich, in the region of immiscibility) in contact with some form of carbon/graphite as the current collector. The latter is dispersed throughout the volume of the electrode compartment so that the molten electrolyte and carbon surfaces are in intimate contact throughout. The composition and temperature range of the fluid system [1, 2] in the sulphur electrode are illustrated in Fig. 1. It is apparent that, associated with the charge–discharge cycles, there is a disappearance of some phases and the appearance of others, and that this event occurs at the interface of the fluid and solids, where the solids are the electronic current collector (i.e., the carbon surface) and the ionic ceramic electrolyte (i.e., β- or β''-alumina). The physical properties of these fluids [3] in the molten polysulphide composition range are those of moderately well-behaved molten salts, i.e., good electrolytes. With the appearance of the sulphur-rich fluid phase in the composition range of liquid–liquid immiscibility, the fluid properties approach those of molten sulphur. Reference to the summary in Table 1 illustrates the magnitudes of such changes for the properties of density, conductivity, surface tension and viscosity.

From one viewpoint, the charge–discharge cycle can be likened to a 'withdrawing' of the carbon/graphite surface from one phase (e.g. molten sulphur) and an 'immersing' in the other liquid (e.g. molten polysulphide) and *vice versa*. If either of these fluids has pronounced preferential wetting properties for the carbon surfaces, a firmly held layer may be established. The presence of such preferential wetting by molten sulphur with the formation of an insulating liquid sulphur layer has been inferred from various electrochemical observations of the sulphur electrode during charge–discharge cycling [4–11]. Such considerations led to our undertaking wetting measurements. The pretreatment of the carbon surfaces, our techniques for surface treatment with metal oxides and some results for the wetting behaviour of molten polysulphides and sulphur have already been reported [12]. The present paper reports the results of an additional series of measurements to investigate factors contributing to the enhancement of polysulphide wetting through added metal

---

[*] Presented at the 30th meeting of the International Society of Electrochemistry at Trondheim, Norway, August 1979.
[†] Present address: Research Staff, Wilson Greatbatch Ltd., Clarence, N.Y. 14031, USA.

0021–891X/80/060789–09$02.90/0

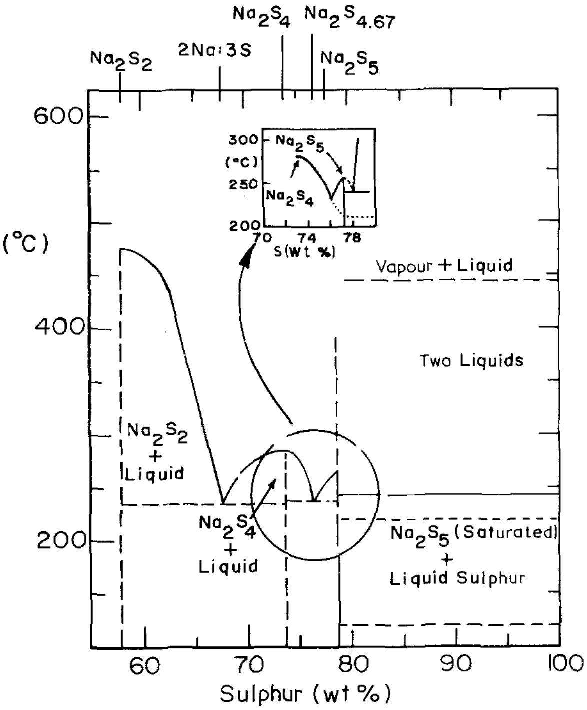

Fig. 1. Phase diagram of the Na–S system for the composition range: Na_2S_2 to 100% sulphur. Data from Rosen and Tegman [1] and Gupta and Tischer [2].

oxides. The extension of such measurements into the region of liquid–liquid immiscibility was also undertaken and this work is also reported here.

2. Basic principles

Direct observations of the contact angle would be most informative. The properties of molten polysulphides and sulphur at 350° C, e.g., flammability, hygroscopicity, vapour pressure, colour (virtually black), make such direct observations difficult. An alternative approach would be to use the principle of capillary penetration, i.e., the substrate is dipped into the liquid and the wettability is observed by the wicking of the liquid.

The pore size distribution of the individual fibres has been reported elsewhere [12]; the diameter range for 90% pore space is ~ 0.01–0.05 μm. The fibres in this matrix, individually and collectively, are illustrated in Fig. 2 at magnifications of 3000× and 60×, respectively. Assuming that the voids in the closely woven graphite matrix may in the first approximation be considered as a collection of 'capillaries', then the volume of the liquid wicked is given by

$$v_e = N\pi r^2 kh \tag{1}$$

Table 1. Physical property data for molten Na_2S_5, sulphur and sulphur-rich (polysulphide saturated) liquids. Temperature ~ 350° C; data from [3]

	Viscosity (cp)	*Density* (g cm^{-3})	*Electrical conductance* (Ω^{-1} cm^{-1})	*Surface tension* (dyne cm^{-1})
Na_2S_5	20	1.86	0.4	114
Sulphur	500	1.67	1×10^{-8}† 1×10^{-12}‡	45
Sulphur saturated with Na_2S_5			1×10^{-7}	

† Reagent-grade sublimed sulphur (as received).

‡ The same, but further chemically treated to reduce (organic) impurity content.

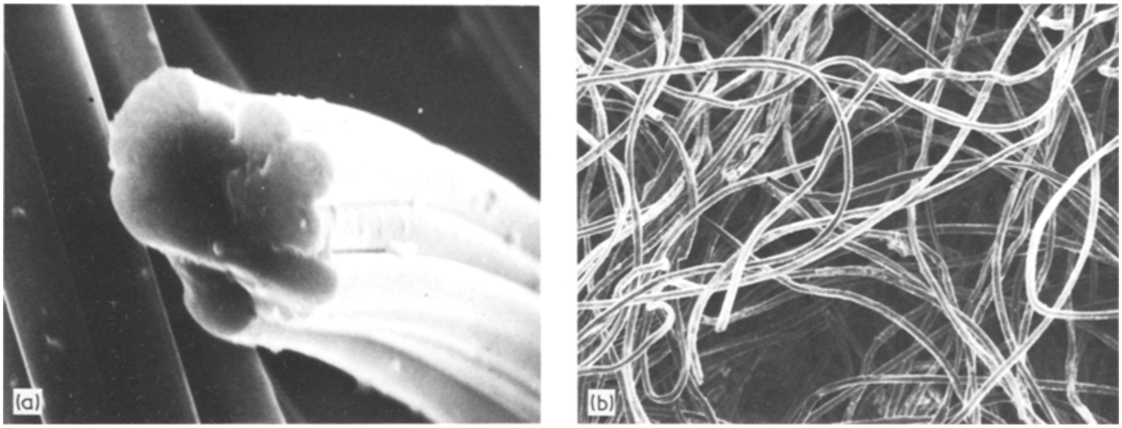

Fig. 2. SEM photomicrographs of WDF graphite felts. (a) Surface of individual fibres of the felt matrix (3000 ×); (b) the fibres of the matrix (60×).

where N is the number of 'capillaries', k the tortuosity factor of the wick substrate and h the climb height (i.e. equilibrium height). Combining this with the definition of the adhesion tension

$$\gamma \cos \phi = h\rho g r/2 \qquad (2)$$

it follows that the adhesion tension ratio can be gained from measurements of the equilibrium heights and/or the weights w of the fluids wicked; i.e., for a common surface

$$\frac{(\gamma \cos \phi)_{\text{sulphur}}}{(\gamma \cos \phi)_{\text{polysulphide}}} = \frac{(h\rho)_{\text{sulphur}}}{(h\rho)_{\text{polysulphide}}} = \frac{(w)_{\text{sulphur}}}{(w)_{\text{polysulphide}}} \qquad (3)$$

As an extension, quantitative insights may be gained for the same fluid in contact with two substrates if the capillarity and tortuosity of the fibrous materials do not differ greatly. Thus the adhesion tension ratio for two such substrates and the same fluid common to both is

$$\frac{(\gamma \cos \phi)_{\text{fm}_1}}{(\gamma \cos \phi)_{\text{fm}_2}} = \frac{(h\rho)_{\text{fm}_1}}{(h\rho)_{\text{fm}_2}} = \frac{(w)_{\text{fm}_1}}{(w)_{\text{fm}_2}}. \qquad (4)$$

Here fm_1 and fm_2 are two fibrous materials with closely similar distributions of capillary radii and tortuosity factors and the h, ρ, and w are as before.

Studies of the adhesion tension ratio may thus be used for basic measurements of the relative affinities of the fluids for the same surface and also in comparisons of the bare (untreated) carbon felts with the same substrates altered through surface treatments (e.g., with surface-deposited metal oxides).

3. Experimental methods

3.1. *Wicking techniques*

For measurements with one fluid, the following procedure was used. The carbon/graphite felt wick and the material to be wicked (polysulphide or sulphur) were loaded into two tubular assemblies (Figs. 3a and b) which were mated to a vacuum manifold through conventional cone/socket vacuum stop-cock assemblies modified for suspension of the wick with fine platinum wire (Fig. 3d). These 'twin' tubular assemblies are closely matched in length, so that the wick and polysulphide mating can be achieved with assurance that the wick position is well defined (e.g., close to full depth of the vessel). To make this mating possible without contact with the other reactant, the melt was 'flowed out' and solidified in a position as illustrated in Fig. 3c. The indentations in the wall of the vessel assisted in positioning the wick and in keeping the fluid and wick from contact during the re-melt. After mating, this system was re-evacuated and the polysulphide (or sulphur) was heated carefully with a portable furnace until the wick and fluid were in contact in the bottom of the vessel. The assembly was then isolated (under vacuum) and transferred to the wetting/wicking furnace. The tubular wicking vessels were sufficiently long for the cone/socket assembly to extend well above the hot zone.

On completion of the wicking period, the assembly was brought to atmospheric pressure so that the wick could be withdrawn for quantitative weight analysis. The wick transfers in the preceding steps were made under research-grade argon (99.999% purity) at atmospheric pressure.

To extend the preceding techniques to measurements in the two-phase liquid region, the procedure was modified as illustrated in Figs. 3d* and e. Because of the markedly differing physical properties (Table 1) and melting temperatures, the two fluids (sulphur-rich and polysulphide-rich immiscible liquids) cannot readily be 'flowed-out' (as in the single-fluid mating step). The wick is thus suspended above the melt, immersed and withdrawn using a windlass improvised with a horizontal cone and socket, fine Pt wire and a glass-rod clamp (Figs. 3f–h). The glass-rod clamp has the further advantage of providing the weight required to 'sink' the (low-density) graphite felts and to maintain the felts in well-defined (extended) configurations. The follow-up analyses are much the same as for the preceding single-fluid measurements.

3.2. *Carbons*

WDF graphite felt (Union Carbide Corporation) was used as the basic substrate for the series of surfaces for the wettability measurements. Metal oxides as surface deposits were applied by the thermal decomposition of metal nitrates and thermal bake-out techniques, as described elsewhere in detail [12]. To investigate the various morphologies of alumina, the final thermal bake-out was modified to complete the preparation at the following temperatures: 350° C, 500° C, 600° C, 700° C, 850° C, 900° C and 1100° C. The surfaces were characterized by scanning electron microscopy (SEM) and X-ray fluorescence spectroscopy and the porosities and pore spectra by mercury intrusion porosimetry, much as in the preceding studies [12].

3.3. *Sulphur, polysulphides*

The measurements of sulphur wetting were made with reagent-grade (sublimed) sulphur; except for 'predrying' (at 10^{-4} torr dynamic vacuum and with temperatures increasing from 25° C to ~ 150° C), the sulphur was used 'as received'. The poly-

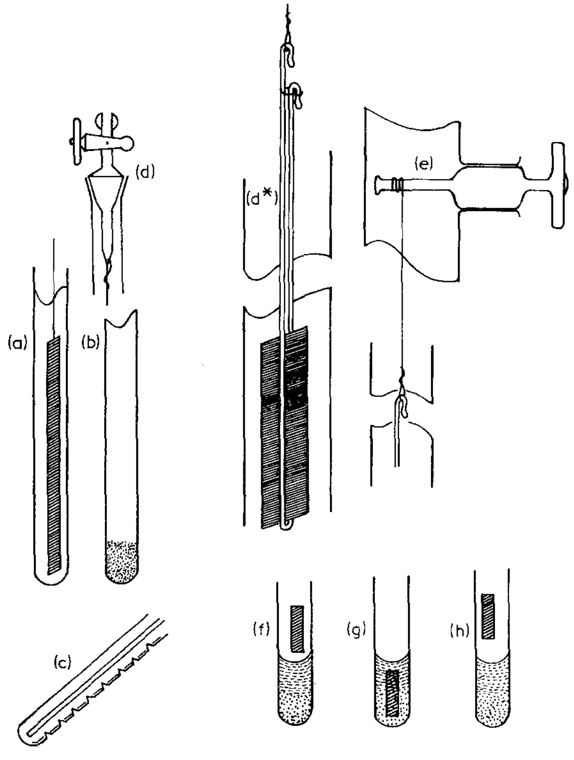

Fig. 3. Wetting and wicking: experimental assemblies. (a) and (b) 'twin' tubular assemblies for wick and fluid pretreatment prior to mating as shown in (c); (d) shows details of the wick suspension and vacuum connection 'header' for these tubular vessels; (d*) and (e) show the glass clamp for the felt and the windlass-type device built into the 'headers' for the two-liquid phase studies; (f), (g) and (h) illustrate the initial, immersed, and final positions of the wicks during such measurements.

sulphide was a product of approximate trisulphide composition, Na_2S_3, and was prepared by the direct reaction of elemental high-purity sodium and sulphur under toluene reflux as described by Cleaver *et al.* [13]. It was filtered and washed with toluene; the excess toluene was removed under vacuum at room temperature and the molten polysulphide was filtered through a pyrex glass frit and stored in hermetically sealed glass ampoules until required for use. For measurements with the two immiscible liquids, the fluids were prepared from the trisulphide composition with additional sulphur, in amounts sufficient to yield the required quantities. The 'two-phase liquid' system was then prepared by heating this mixture for a prolonged period (with periodic vigorous manual shakings); these mixtures were prepared in the vessels to be used for the subsequent wetting/wicking measurements so that no additional transfers were required.

4. Measurements

A series of measurements was undertaken to investigate the wetting properties of molten polysulphides (i.e., Na_2S_3), molten sulphur, and molten Na_2S_5/sulphur (two immiscible liquids in contact as saturated solutions) using both the bare graphite felts and the same substrate treated with various metals (as oxides). The systems were maintained at 330–350° C for the wetting observations; with molten sulphur the measurements were not too satisfactorily accomplished at these temperatures. Owing to the free volume extending above the hot zone the sulphur would 'climb' out to lower temperature regions and it was found necessary to 'chase-back' the sulphur into the hot zone repeatedly during the course of an experiment. This problem was not encountered with the polysulphides; the molten polysulphides remained in the bottom of the wicking vessel so that the measurements were accomplished in the hot zone throughout each experiment.

The wetting/wicking results were normalized to a standard felt size (28 cm x 0.5 cm) for quantitative comparisons. The wetting properties of sulphur, polysulphide, and [Na_2S_5 (sulphur saturated)–sulphur (polysulphide saturated)], i.e. two immiscible liquids in equilibrium, were investigated relative to the following surfaces: bare (untreated) graphite felts and graphite felts treated with alumina and oxides of Fe, Cr, Ni, Co, and selected mixtures, e.g., alumina–Fe oxide. Measurements were extended to investigate the influence on the wetting properties of trace impurities such as H_2O, Na_2SO_4, etc. in the molten electrolyte. These results are also reported here.

5. Results and discussion

The results for the bare felt surfaces are summarized in Table 2. Compared with molten sulphur, the polysulphides appear relatively nonwetting. Using the adhesion tension criterion (Equation 3) and the averaged results, the adhesion tension ratio is found to be: sulphur/polysulphide $= \sim 27$.

Table 3 gives the results for a series of exploratory measurements with treated graphite felt surfaces and molten polysulphide. Co, Cr, and Al (as oxides) enhance the wetting properties, Al_2O_3 having the most pronounced effect. Use of a mixture of Al and Fe (as oxides), with Fe in trace amounts, does not promote additional wettability; the differences in the amounts wicked as ascribed to insufficient contact (immersion) of the wick in the measurement with 'Al only' surface additive. When the area of the treated surface of this wick was decreased by re-deposition of carbon, the wettability decreased. This surface is shown in Fig. 4 at 1240x, and the polysulphide wicking in Table 3, last entry.

The wetting properties of alumina-treated surfaces were investigated by using a series of graphite

Table 2. Polysulphide and sulphur wicking: bare graphite felt surface

*Polysulphide wicked** ($g\,g^{-1}$ felt)		*Sulphur wicked** ($g\,g^{-1}$ felt)
(a) 0.36	(f) 0.29	(i) 9.4
(b) 0.36	(g) 0.37	(j) 11.4
(c) 0.30	(h) 0.29	(k) 8.4
(d) 0.29		
(e) 0.60		
Average ~ 0.36		Average ~ 9.7

* Results normalized to wick dimensions of 28 cm × 1 cm × 0.5 cm; polysulphide at 330–350° C; sulphur at ~ 330–350° C except for (j) for which the hot zone was decreased to ~ 200° C.

Table 3. Polysulphide wicking: treated felt surfaces

As oxides	*Wt%*	*Polysulphide wicked** ($g\,g^{-1}$ felt)
Al	~ 16	5.8
Ni	~ 15	0.36
Cr	~ 18	4.1
Co	~ 17	2.6
Al + Fe	~ 13	7.2
Al + Co	~ 18	1.5
[Al + Fe]C	[13]35	2.2

* Wick dimensions: 28 cm × 1 cm × 0.5 cm; temperatures: 330–350° C.

Table 4. Polysulphide wicking: alumina-treated surfaces

Alumina (wt%)	*Polysulphide wicked** ($g\,g^{-1}$ felt)
~ 2	1.1
~ 6	8.2
~ 16	5.8
~ 50	8.5

* Wick dimensions: 28 cm × 1 cm × 0.5 cm; ~ 10–11cm fully penetrated with polysulphide; maximum height wicked ~ 28 cm.

felts with increasingly loaded fibres. The results are shown in Table 4. It is seen that the wettability of such graphite surfaces is significantly enhanced by rather modest amounts of alumina (i.e., ~ 6–16%); the bulk of the polysulphide was found in the first 10–11 cm of the felt wick, but partial filling of the wicks was found to much greater heights, (e.g., to the total wick length of 28 cm in the present case).

The thermally induced decomposition of aluminium nitrate, and transformations at increasing temperatures [14–17] are summarized schematically in Fig. 5. A series of treated felt surfaces was prepared [12] starting with approximately 48–53 wt% alumina-loaded graphite felt, with the final bake-out temperature in the Asbot treatment being adjusted to 380° C, 500° C, 600° C, 700° C, 850° C, 900° C and 1100° C. The amounts of polysulphide wicked by each of these felts was approximately the saturation amount, i.e., ~ 7.3–9.4 $g\,g^{-1}$ felt wick (normalized to 28 cm x 1 cm x 0.5 cm wick size). The wicked polysulphide distributions for this series are summarized as bar graphs in Fig. 6. While there are differences, one may conclude that in the first approximation the various treatments lead to surfaces of approximately equal activity.

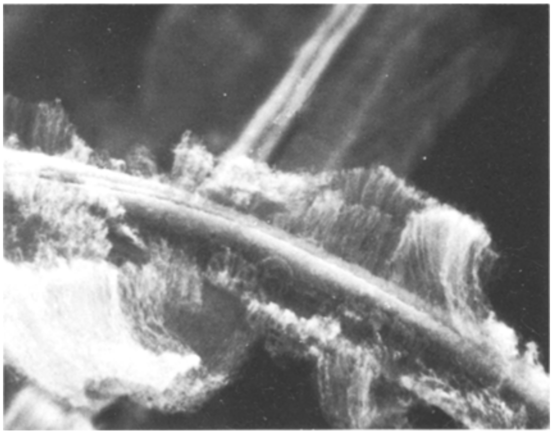

Fig. 4. SEM photomicrograph of a WDF graphite felt surface treated with Al and Fe (as oxides) and with re-deposition of carbon to decrease the treated surface. The re-deposited carbon in the Al–Fe (oxide) treated fibres is seen as plumes.

The weight changes of the treated felts were followed by batch weighings in a separate series of measurements. Relative to the treated felt (after 380° C bake-out), the additional weight losses were approximately: 500° C, ~ 4.7%; 600° C, ~ 6.4%; 700° C, ~ 5.0%; 800° C, ~ 7.5%; 850° C, ~ 5.9%, 920° C, ~ 4.3%; 1100° C, ~ 4.9%. The felt surfaces prepared at 380° C and 500° C were quite sensitive to atmospheric moisture (i.e., they exhibited hydroscopic properties). The nature of the volatiles was not investigated except to note that both H_2O and nitrogen oxides were released as the bake-out temperature was increased to about 600° C. No clear trend is apparent in the weight losses and it appears that the decomposition processes are more or less complete at ~ 600° C. The increase of the final Asbot temperature to 1100° C shows no marked improvement to the polysulphide wettability of the treated surfaces. The possible improvements to the alumina/graphite bonding by the use of these higher temperatures were not investigated.

The results of measurements with the felt wicks totally immersed in the molten fluids (two liquids immiscible phase composition range of the Na/S system) are summarized in Table 5. With untreated

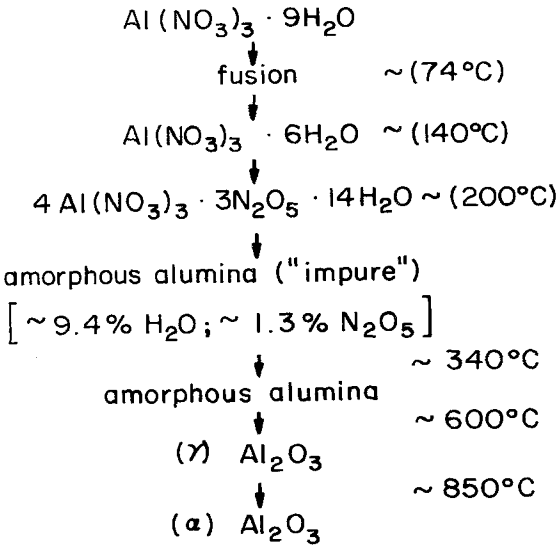

Fig. 5. Thermally induced decomposition of aluminium nitrate and transformation with decreasing temperature.

or bare WDF felts, and heavily loaded (alumina treated) felts, it is seen that preferential penetration of the graphite felt matrix by sulphur and polysulphide is obtained; with a moderate loading, under such a competitive situation, approximately equal penetration of the felt by the two fluids was obtained. This is expressed quantitatively in the adhesion tension ratios.

It is of interest to compare the adhesion tension ratio for the bare felt surfaces found in competitive wetting (Table 5), and from use of single fluids (Table 2), i.e., 27 and 40. Apparently the single-fluid measurements, which are experimentally less difficult, may be used to gain insights into the competitive situation in the region of liquid–liquid immiscibility.

Some wicking measurements were undertaken with exposure of the polysulphide to atmospheric (room) conditions during the transfers prior to wicking, and with H_2O, NaOH and a mixture of Na_2SO_4 and $Na_2S_2O_3$ added in small amounts to the polysulphide. Measurements were made with

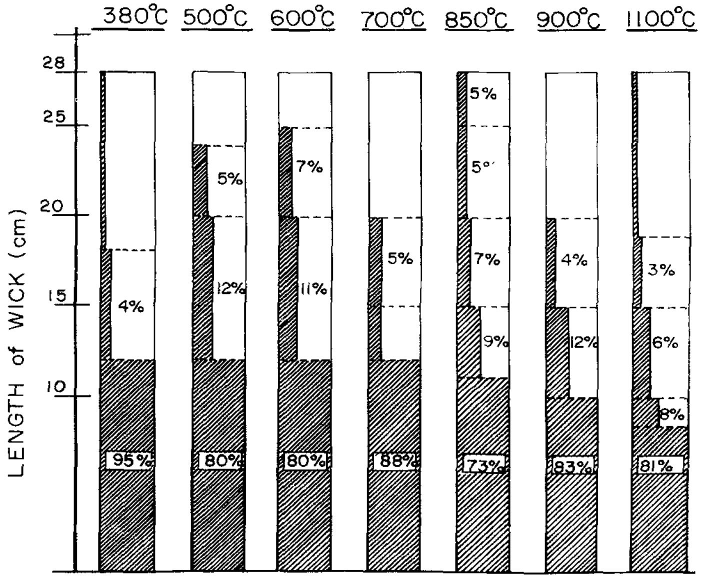

Fig. 6. Bar graph summary of the wicked polysulphide (weight per cent) as a function of the length of wick extending above the molten polysulphide fluid level in the wicking assembly. The approximate amounts wicked are shown qualitatively as the filled areas and quantitatively as per cent of the polysulphide. Partial penetration to the full 28 cm length was observed in some instances.

Table 5. Polysulphide, sulphur competitive wetting with totally immersed felts. Wick dimensions: 7 cm × 1.6 cm × 0.5 cm

		Weight (g)	*Adhesion tension ratio* (sulphur: polysulphide)
Bare felt	Sulphur	28	40
	Polysulphide	0.7	
Alumina-treated felt (50 wt%)	Sulphur	1.9	0.1
	Polysulphide	21.2	
Alumina-treated felt (17 wt%)	Sulphur	14.3	1.3
	Polysulphide	10.6	

both bare and alumina-treated felts. The results are given in Table 6. Based on this limited series of measurements, it is apparent that the polysulphide wettability of the bare felt surfaces is significantly decreased (cf. Table 2, 0.36 g g^{-1} felt), and that H_2O and [sulphates and thiosulphates] also have adverse effects on the alumina-treated felt surfaces. Except for these exploratory series of measurements, the influence of various additives as impurities in the polysulphides was not investigated further in the present work.

In the Absot treatment of the graphite felts, a concentrated HNO_3 wash is used to pretreat the felts, the last traces of which are removed thermally (~ 500° C) under dynamic vacuum. It was observed that the activity of a moderately loaded felt (~ 6% alumina) decreased by ~ 86% in polysulphide wettability if the HNO_3 step was bypassed. With bare felt surfaces, the decrease noted was ~ 50%. By contrast, the sulphur wetting properties appeared unchanged. Nitric acid data reported elsewhere [18, 19] show that acid strengths greater than 40% are required for significant surface oxidation and that chemical activation of the carbon/graphite surfaces is promoted by such treatments. The untreated surfaces are essentially neutral and possess low surface areas; oxidative treatments, to increase both the concentration of chemically active acid groups (to promote metal oxide–fibre interactions) and the surface area, appear to be beneficial in the preparation of felts treated with metal oxide for improved polysulphide wettabilities.

Table 6. Polysulphide wicking: trace impurities

Additive to polysulphide		*Polysulphide wicked*[a] (g g^{-1} felt)	
Impurity	*Amount* (wt%)	*Bare felt*	*Alumina-treated felt*[b]
O_2, H_2O	c	d	6.9
H_2O	~ 2	0.15	2.6
NaOH	~ 1.3	0.05	7.5
$\begin{bmatrix} Na_2SO_4 \\ Na_2S_2O_3 \end{bmatrix}$	each ~ 1	0.05	1.0

[a] Wick dimensions: 28 cm × 1 cm × 0.5 cm; temperature: 330–350° C.
[b] Approximately 14 wt% alumina.
[c] Room atmosphere exposure of polysulphide during loading of wicking vessel (~ 5 min) prior to evacuation and premelting.
[d] Not investigated.

The role of sulphur purity was examined through a series of experiments in which three different grades of sulphur were used for the preparation of the polysulphide (by the toluene technique [13]). The sulphurs were: reagent-grade (sublimed) sulphur, the same quality subjected to further rigorous sublimation and the same quality purified by refluxing with MgO, after Bacon and Fanelli [20]. The comparative properties [3] of these qualities of sulphur show that the electrical conductivity decreases from 1×10^{-8} ohm^{-1} cm^{-1} to approximately 1×10^{-12} ohm^{-1} cm^{-1} with the MgO purification techniques. The wetting qualities of the polysulphides (prepared from these three grades of sulphur) were found to compare closely, i.e., 8.2, 9.3, and 9.0 g g^{-1} felt. The felt wicks for these series of measurements were modestly loaded felts (~ 15% alumina) at temperatures of 330–350° C.

A commercially available polysulphide product,

Na_2S_4 (Alfa-Ventron) was considered as a possible reference material for the present series of wetting measurements, much as in the preceding communication [1]. The commercial product was exhaustively washed with toluene (sulphur extraction) and pre-dried at 150° C under vacuum prior to wettability tests. Using alumina-treated felts, it was found that the wetting properties of this polysulphide were not sufficiently reproducible and did not rise to levels noted with the polysulphides directly synthesised by the Na/S reaction under toluene. Examination of the melting processes (visually) showed the toluene-washed commercial material still retained sulphur; under dynamic vacuum this was evidenced as a black liquid froth at 95–150° C; after this had run its course, the solid residue at 150° C was green and could be heated *in vacuo* to melting temperatures; approximately 1–3 wt% sulphur sublimed during this period. By contrast, the laboratory synthesis from Na/S (under toluene) melted smoothly under comparable conditions, with no 'frothing' (95–150° C) and without sublimation of sulphur. The earlier practice of using the commercial material for wettability measurements [12] was discontinued in the present work in favour of the 'in-house' laboratory preparations with concomitant greater quality control.

In situ tests in sodium–sulphur cells fell outside the scope of the present investigations and remain an area for further work. Based on graphite wetting studies in molten halides [21], one would predict that the wetting behaviour as a function of the composition of the melt phase would be closely similar both in the presence and absence of d.c. anodic polarization. It is of interest to note that two patents appeared [22, 23] during the course of these investigations. Both report improvements in charge–discharge cycling of sodium–sulphur cells and use alumina-treated graphite surfaces as part of the sulphur electrode.

Acknowledgements

This work was made possible in part by support received from the National Science Foundation and the U.S. Department of Energy. The participation of G. M. Volker and G. Miller in the preparative synthesis of the polysulphides and analytical aspects of these studies is acknowledged with pleasure.

References

[1] E. Rosen and R. Tegman, *Chemica Scripta* **2** (1972) 221.
[2] N. K. Gupta and R. P. Tischer, *J. Electrochem. Soc.* **119** (1972) 1033.
[3] 'Physical Properties Data Compilation Relevant to Energy Storage. II. Molten Salts Data on Single and Multi-Component Salt Systems', U.S. Dept. Commerce, National Bureau of Standards, NSRDS-NBS-61, Part II, April (1979).
[4] R. D. Armstrong, T. Dickinson and M. Reid, *Electrochim. Acta* **20** (1975) 709.
[5] D. A. Aikens, K. W. Fung, F. A. Ludwig and R. P. Tischer, 'Extended Abstracts', *The Electrochemical Society Spring Meeting, Toronto, Canada* (1975) p. 881.
[6] R. D. Armstrong, T. Dickinson and M. Reid, *Electrochim. Acta* **21** (1976) 935.
[7] F. A. Ludwig, *Electrochem. Eng.* **10** (1977) 391.
[8] M. P. J. Brennan, *Electrochim. Acta* **22** (1977) 279, 609.
[9] *Idem, J. Electrochem. Soc.* **125** (1978) 705.
[10] R. D. Armstrong and M. Reid, *Electrochim. Acta* **22** (1977) 1332.
[11] D. A. Aikens, in 'The Sulfur Electrode' (edited by R. Tischer) Academic Press, New York (1979).
[12] G. J. Janz and R. M. Murphy, *J. Electrochem. Soc.* **125** (1978) 1605.
[13] B. Cleaver, A. J. Davies and M. D. Hames, *Electrochim. Acta* **18** (1973) 719.
[14] K. H. Stern, *J. Phys. Chem. Ref. Data* **1** (1972) 747.
[15] K. Funaki and Y. Shimizu, *Kogyo Kagaku, Zasshi* **62** (1959) 788.
[16] A. M. Kalinina and E. A. Porai-Koshito, *Dokl. Akad. Nauk. SSSR* **114** (1957) 365.
[17] K. Nikolic and K. Velasevec, *Acta Pharm. Jugosl.* **10** (1960) 131.
[18] J. W. Herrick, 'Proc. 2nd. Ann. Technical Conf. Reinforced Plastics/Composites Division', Soc. Plastics Ind. Inc., Section 16-A, (1968) p. 1.
[19] J. C. Goan and J. P. Prosen, *ASTM Special Publ. 452* ASTM, Philadelphia, Pa. 19103, USA (1969) pp. 3–26.
[20] R. F. Bacon and R. Fanelli, *Ind. Eng. Chem.* **34** (1942) 1043.
[21] E. A. Zhemchuzhina, in 'Surface Phenomena In Metallurgical Processes', (edited by A. I. Belyaev) Metallurgizdat, Moscow (1963); English Translation Consultants Bureau, New York (1965) pp. 70–101.
[22] F. A. Ludwig, U.S. Patent 4 084 041; assigned to Ford Motor Co. (11 April 1978).
[23] I. Wynn Jones and G. Robinson, Br. Patent 1 528 672; assigned to Chloride Silent Power Ltd., (18 October 1978).