Corrosion management in bleach plant by electrochemical protection

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ABSTRACT

Pulp and Paper Industry experiences intensive and diversified types of corrosion. As such it has often remained at the fore-front of corrosion management technology. Among its various sections bleach section is observed to be among the more corrosion affected sections. This is basically due to lower pH, presence of oxidising and chlorine containing chemicals in its liquor media. Observance of pollution control and energy saving procedures has further increased the corrosivity of these media. This in turn has resulted into replacement of conventional materials of construction by upgraded but costlier materials. Alternatively other protection procedures have also been under consideration. Present paper discusses the condition of Indian mills in the present scenario and also the likely impact of bleaching practices which may be adopted in future. In the more aggressive conditions, electrochemical (anodic) protection turns out to be more cost-effective. As such the paper also discusses the principal of anodic protection, how it can be used in bleach washers and what practical aspects one should take care of ? Considering case histories, attempt has baen made to show the beneficial aspects of applying the protection system. Lastly a brief idea has baen given about the experiments that are being planned in connection with the development of such a protection system,

1. Introduction :

Pulp and Paper Industry is severely affected by corrosion in its various section. The attack on construction materials is intense as well as of diversified type. The severity of attack can be gauged from the Table 1.

Table 1 : Annual Corrosion Cost experienced by Pulp and Paper Industry (1-5).

Country	Year	Annual Corrosion Cost	
		Per ton	Total (in millions)
USA	1971	$ 5.00	$ 190.00
	1978	$ 13.60	$ 1400.00
	1984	$ 5.00-$13.6	$ 525-$1170
Canada	1978	$ 8.35	$ 270
	1989	—do—	$ 300
India	1985	Rs. 100.00 ($ 7.70)	Rs. 160 ($ 12.3)

Due to above reason, Pulp and Paper Industry is often at the forefront of corrosion control technology, as appears from following facts (1,2) :

i. Paper industry is one of the first to utilise Molybdenum bearing Stainless-Steel more than 60 years ago.

ii. It is also the among the few industries, which uses stainless steel quite widely (95% of the machines in a pulp and paper mill is made up of Stainless-Steel) (6).

iii. Wide acceptance of FRP constructed process machinery components in this industry.

It has been estimated that, with the existing knowledge of corrosion, around 17% of the expenditures incurred due to corrosion effects (≈ Rs 3 Crore per year in case of Indian industry) can be recovered back by avoiding the losses (3)

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Corrosion in a Pulp & Paper mill can be divided into four sections. Corrosion in (i) Digester House (ii) Recovery Section (iii) Bleaching Section and (iv) Paper Machine. Of these, carrosion in bleach section is observed to be most expensive. A study conducted in 1975 by a United States Pulp & Paper company showed that corrosion in bleaching process costs approximately $ 4.25 per ton of paper produced. Thus 50% of the total corrosion cost spent in producing bleached paper is consumed by bleaching section (5,7) alone.

In order to combat corrosion, in bleach section, several surveys and a resurvey (8—12) have been conducted to assess the severity of problem, its correlation with process parameters etc. It was also observed that equipments involved in handling and washing of stock between successive bleaching operations were more prone to corrosion attack than others. As such emphasis was given to corrosion problems faced by washers, vat, related pipe-lines and other components. Later mill based tests were also conducted on bleached pulp washer (13-22) with a view to find and suggest suitable construction materials for process machinery. Evidently, perhaps, no other section of paper industry has drawn attention so widely as this section of bleach plant in solving its corrosion problems. Consequently all discussion henceforth, relates to bleached pulp washers and related machinery components.

The above mentioned surveys and mill based tests also indicate that corrosivity of bleach plants in developed countries mills has increased over past about 20 years—the time when mills started using filtrate recycling. This became necessary in view of the stringent Government laws to protect environment by not allowing to spew 'garbage' into the environment and secondly also for saving energy. These measures have led to closed up washwater systems which changed the process parameters to lower pH, higher temperature, Cl^- ion levels and residual Cl_2 concentration (10, 11, 16, 23, 24). All these factors increase corrosivity of bleach plant media. This is also reflected in the trend that conventionally used construction materials e.g. SS-316 and 317 have now been replaced by higher Mo,Cr and Ni containing stainless steels and Ni based alloys for fabricating various components of chlorination (C) and chlorine dioxide (D) stages of bleaching (8—11, 13, 16, 18, 19). Clearly, the practices adopted in present day bleach plants have made this section much more corrosive than what it was earlier. The situation obviously warrants observing, suitable corrosion management procedures in order to economise paper making.

2. Corrosion Management Procedures :

In principle, there are three ways in which ill effects of corrosion can be managed :

- 2.1 Selection of suitable materials.
- 2.2 Providing suitable protective barrier to surface likely to experience corrosion.
- 2.3 Electrochemical Protection.

Infact, the most cost-effective alternative could be to avoid the use of corrosive chemicals and process conditions. However, bleaching operation is such that harsh process conditions can't be avoided.

2.1 Material Selection :

Suitable material selection involves intelligent use of metals as well as non-metals including polymer composites. It consists in comparing, for the purpose of selecting appropriate material, corrosion resistance, mechanical strength, fabrication, cost and availability of candidate materials This is done so as to form a given process equipment or component with a material which turns out to be cost-effective considering its life to be 10 years or so. Efforts for selecting appropriate materials has led to development of many new alloys e.g. 317 for bleach plants in early fifties (16), alloys 86 for suction rolls in eightees (6) and many other specialised stainless-steels and nickel based alloys. Increased corrosivity in past decade and half has led to use of these newer developed, more exotic alloys. In some mills bleach pulp washers for C and D stages have been suggested to be constructed of Titanium 25) and non-metallics (26-29) as they are considered more resistant in media even more corrosive than anticipated in bleach plants. However, the 'exotic' materials are very costly whereas improper design and fabrication in case of non-metallics have led to premature failures of components/equipment (30). Both the observations have forced the workers to look to other alternatives for corrosion management in more cost-effective way.

2.2 Protective Barrier:

The second alternative involves in providing a protective barrier to metallic surface thereby avoiding its contact with corrosive media. Such a situation can be realised in one of the following ways—(i) by coating the metallic surface, (ii) by lining the surface with a more corrosion resistant material which could be a metal e.g. Ni or its alloy, Titanium etc. or a non-metal e.g. FRP, Polyvinyledene fluoride (Kynar) etc. (27, 28; 30, 31). A general problem of application of coating or lining is that it involves complex procedures e.g. surface preparation, removal of even traces of air between liner and vessel wall etc. It requires highly skilled workmanship and hence increases cost of coating/lining Further because of different thermal expansion coefficients of polymers and metals, durability of steel-to-resin bond is doubtful specially at temperatures beyond ≈ 50°C. This is more so in the case of washers which are moving equipment.

2.3 Electrochemical Protection :

Electrochemical protection is the third alternative, applied to reduce the ill effects of corrosion. There are two ways of protecting the system by this method—(a) Cathodic protection, (b) Anodic protection.

(a) Cathodic protection: In this method, the material/equipment that is to be protected is made cathode either by 'impressed' negative potential from external battery, or by the use of 'sacrificial' anode. Cathodic protection is being used for about half a century for protecting underground pipe-lines or structures specially around sea-shores. It has been established as an effective, economic and proven tool over the years in many industries e.g. petrolium, chemical industries etc. In paper industry, it finds applications for underground supply lines, internal surfaces of water storage tanks, effluent clarifiers etc. (32, 33).

(b) Anodic Protection : This is achieved by making the material/equipment as anode. This is done by applying positive potential to the material that is to be protected, so that it passivates. Naturally this method is applicable only for those materials which experience passivation' during anodic polarisation. Anodic protection is much recent alternative than cathodic type, conceived only in late fifties (34). However, due to its much less operational cost, simplicity of operation and its acceptibility to already existing systems, this technique is expected to be accepted by industries in coming years, specially by those which encounter very aggressive media. It is already being used to protect equipment used to store and handle sulfuric acid, in fertiliser industy etc. (34). In pulp and paper industry, this technique has been accepted even more than cathodic protection. Thus anodic protection systems have been developed and are in use for protecting digesters, recausticizing storage tanks and unit clarifiers and bleach plant washers (1). Our concern at present is to develope an anodic protection system for bleach plant washers. This is because bleach plant washers are most corrrosive and corrosivity in them is expected to increase further in future due to filtrate recycling and anodic protection technique is expected to be cost-effective in comparison to other corrosion therefore, has been given about the theoretical background of the technique, its over all performance in bleach plant washers and need for developing such system for bleach plants in Indian mills. Lastly a brief idea has been given about the future plans on the experiments to conduct in the endeavour of developing such a system.

3. Principle of Anodic Protection :

Any metal (stainless steels, nickel based alloys etc.) in a liquid environment (bleach liquor) can be considered as equivalent to an electrical cell (or battery) having two electrodes anode and cathode (Fig. 1). Due to inhomogenities existing in a material, some parts of this material may act as anode and others as cathode. In corrosion terminology, anode experiences corrosion (dissolution of material) and cathode remain unaffected. Corrosion proceeds only when anode and cathode are in electrical contact because that leads to flow of electrical current. Where as ions flow through the liquid medium, electrons flow through electrical conducting medium [metal itself in Fig. 1 (a) or connecting wire in Fig 1 (b)], there by completing the electrical circuit. The measure of electrical current thus is also a measure of rate with which metal is corroding. One observes that all the condilions required for flow of current [Fig. 1 (b)] are met in Fig. 1 (a), where a rotary drum has been shown in contact with bleach liquor, and hence the material of drum corrodes.

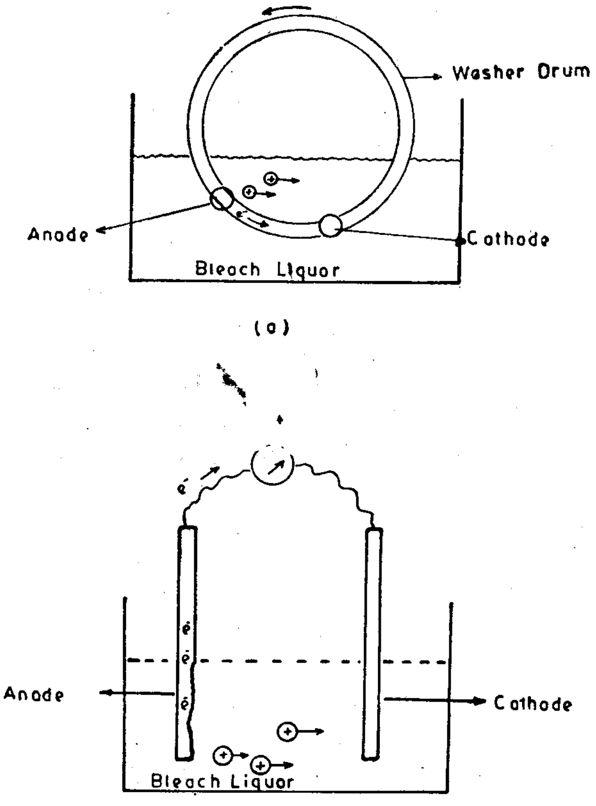


Fig.1: SIMILARITY BETWEEN WASHER IN BLEACH LIQUOR AND EQUIVALANT ELECTRICAL CELL.

In order to reduce or eliminate corrosion, the material drum may be made cathode of electrical cell (cathodic protection), because 'cathode' in corrosion electrical cell remains unaffected. Other way could be that drum material is made 'anode' of corrosion cell (anodic protection), by impressing positive potential from an external battery (anodic polarisation). But how come a material will be protected by making it as anode since anode experiences corrosion ? The trick is passivation behaviour', shown by stainless steels, nickel based alloys. When a positive potential is applied over a metal, its corrosion rate varies as shown in Fig 2. Since corrosion rate is proportional to current flowing in corrosion cell, we are considering current axis basically equivalent to corrosion rate axis. The starting potential is 'O', on increasing potential up to A (active region), corrosion increases. Beyond A, on further increasing potential, corrosion decreases drastically, The reduced corrosion rate could be as less as one twentieth of the rates when potentials are in active region and is presumed to be due to formation of protective (passive) film over matallic surface. Between B and C, corrosion rate remains almost independent of applied potential. This region is called as 'passive' region. On increasing the potential beyond C, the protective film breaks, corrosion rate increases and the material starts experiencing localised corrosion e.g. pitting, crevice corrosion etc. This region is called as 'transpassive region'. This type of behaviour of a material, shown in Fig. 2, is know as 'anodic polarisation behaviour', because in this the metal is made anode of the electrical cell. In anodic protection, it is therefore tried to keep the potential of metal, that is to be protected, in 'passive' region (region 'BC' in Fig. 2) with the help of external source of electricity (a D C. battery or rectifier).

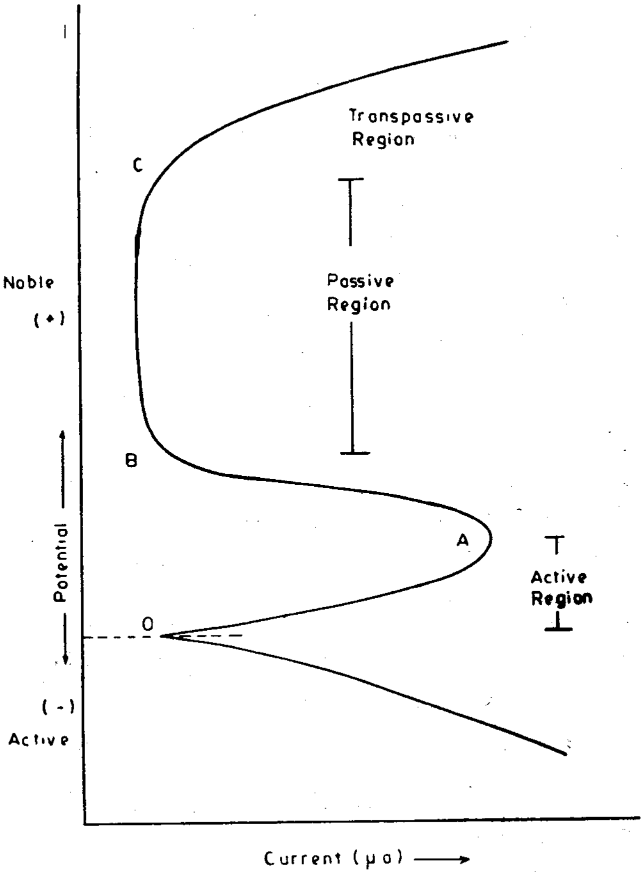


FIG 2 · ANODIC POLARISATION BEHAVIOUR OF STEEL

In order to know as to how much potential is to be applied and how much current will be required before the material passivates, it is necessary to know anodic polarisation behaviour of the material in the corrosive environment. For our case, it is, therefore, necessary to know this behaviour for stainless steels in liquors prevailing in bleach section namely C-vat filtrate and ClO_2 solutions (filtrate of chlorine dioxide stage) Since hypochlorites are not that aggressive as these two solutions are, hypochlorites are not that aggresive as these two solutions are, discussion has not been given about passivation behaviour of steels in hypo-vat filtrates. To understand the requirement of potential and current for passivating steels in Cl_2 and ClO_2 solutions, we can consider the examples of earlier developed protection systems for C and D-stage washers (35–38).

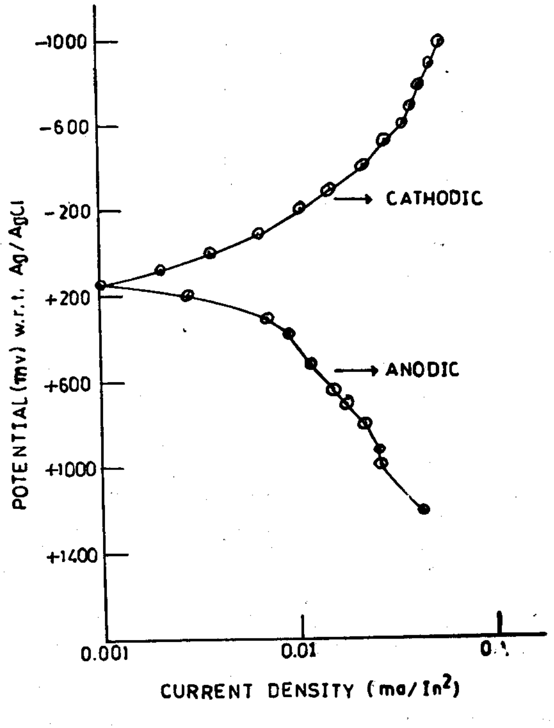


FIG. 3 - POLARISATION CURVE OF SS-317 IN Cl_2 SOLUTION

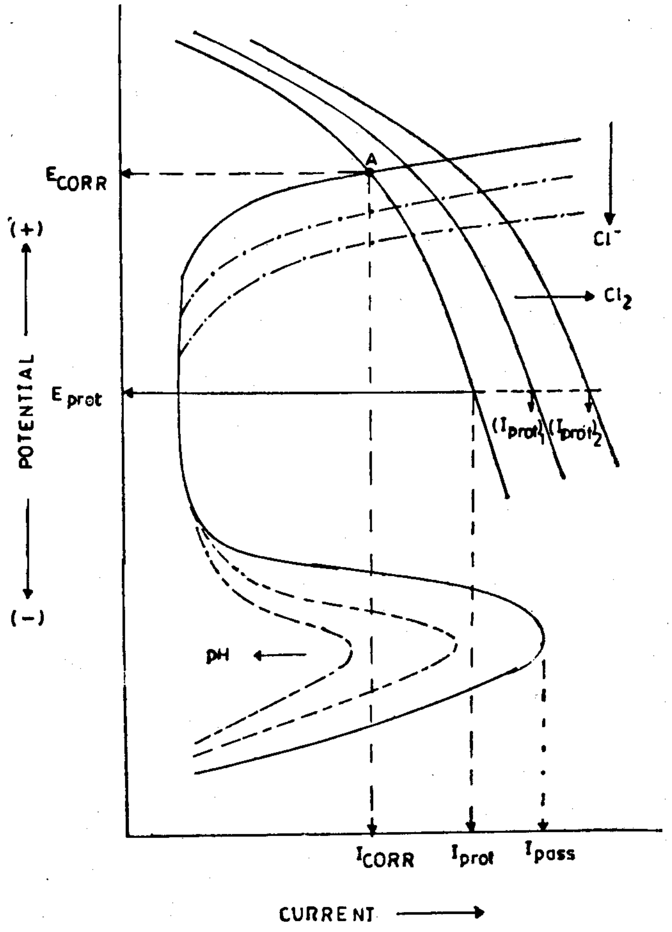


FIG. 4 - ANODIC AND CATHODIC POLARISATION CURVE

Fig. 3. shows polaristation curve of stainless steel (SS–317) in Cl_2 bleach solution at 60°C. This curve shows corrosion potential (potential of washer material i.e. SS–317 in Cl_2 solution in normal state of washer) as +200 mV (with respect to Ag/AgCl reference electrode). This curve can be seen (Fig 4) as built of

(i) the anodic polarisation curve due to following anodic reaction taking place at stainless– steel

$$Fe \longrightarrow Fe^{2+} + 2e^- \qquad ...(1)$$

(ii) cathodic polarisation curve due to following cathodic reaction

$$Cl_2 + 2e^- \longrightarrow 2Cl^- \qquad ...(2)$$

Corrosion potential corresponds to point 'A' in Fig. 4 Naturally in normal state, the washer material of chlorination vat will be in transpassive region (Fig. 2) and as such it experiences pitting and crevice corrosion both. This is a fact observed experimentally during mill tests (21,22). Fig. 4 also show corrosion rate of washer in normal state (this will correspond to I_{corr} of (Fig 4). In order that, washer material operates in passive state, the potential of washer (or corrosion potential) has to be changed to some value lying well within passivation region (say B in Fig. 4). This potential is more negative than e_{corr} (corrosion potential). In other words, the washer is to be polarised cathodically such that washer potential becomes I_{port}, (Fig 4). Current required for attaining e_{prot} is e_{prot} (Fig. 4.). In case of SS-317 in Cl_2 water (Fig. 3) (35) corrosion halted when potential was maintained 1200 mV above the corrosion potential (i.e. at-1000 mV with respect to Ag/AgCl.) Table-1 (37) shows the C-stage washer's corrosion potential and the potential at which washer was maintained in different mills. In each case, the washer was polarised cathodically by a few hundred millivolts.

4. Anodic Protection System :

Fig. 5 shows a bleach pulp washer with installed anodic protection system. It shows a single anode mounted in the base of tile-lined vat (such vats exist normally in Indian mills) and connected to positive (+ive) terminal (anode) of regulated voltage DC-rectifier. The negative (—ive) terminal (cathode) of this power supply is connected to the washer drum through a rotating mercury contact. The voltage that is to be impressed on washer, is measured between cathode side of rectifier and saturated calomel reference electrode (SCE) lying in vat. The measurement is done using a high impedence voltmeter. The voltage may have to be adjusted manually time and again due to change in protection potential, which occurs due to variation in residual Cl_2 concentration during process. This change could be as much as ≈ 500 mV. Now-a-days one makes use of potentiostat, a device which does these operations automatically.

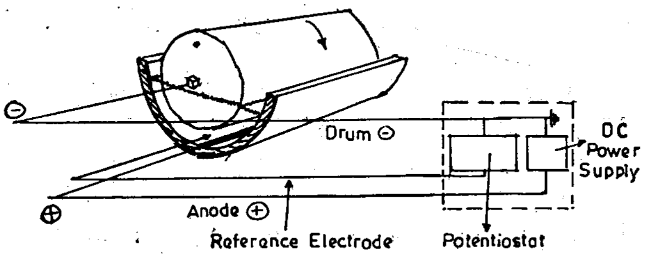
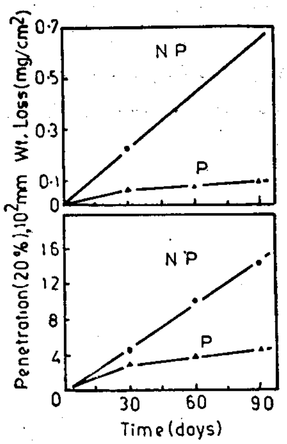


FIG. 5 - ELECTROCHEMICAL PROTECTION OF WASHER

5. Effectiveness of Protection System :

The effectiveness of such a protection system can be judged from an example of chlorine washerwhose vat liquor has 0.15 gm/litre residual Cl_2 (39). The corrosion potential of washer is observed to be + 200 mV (with respect to SCE), a potential sufficient for washer to experience crevice corrosion and pitting both. When it is protected initially by applying—700 to—800 mV, one observes, inhibition of localised corrosion on washer, but it observes general corrosion (washer is in 'activation' range). As such corrosion rate is found to increase. The potetial of washer is further adjusted to—500 mV and then protection optimised by adjusting potential to -550 mV (with respect to SCE). Results of the corrosion loss and penetration depth due to localised corrosion are compared. in Fig (6), in the case of protected and unprotected coupons which were exposed for 30,60 and 90 days. These results clearly establish the effectiveness of protection system.



N P–Not Protected
P –Protected

FIG 6 - COMPARISON OF CORROSION IN PROTECTED ,. UNPROJECTED COUPONS

In order to further establish its effectiveness, corrosion tests were performed in C— and D—stage washers of several mills, on protected and unprotected coupons covering a protection period upto 5½ years (40). It was observed that, on an average, the protected coupons lost ≈ 1/5 of the weight lost by unprotected coupons. It can further be seen that an alloy with protection improved its performance to that of an upgraded (but unprotected) alloy- Thus SS-3.6L and 317L, with protection, perform equivalent to 904L and 254 SMO respectively. (Under identical conditions, an unprotected SS-317L coupen lost ≈ 5 times more weight than that of 254 SMO). In other words, one can expect to continue to use SS-316L and 317L, but with protection system, in chlorination washer with increased corrosivity instead of having a washer of upgraded alloy e g. 904L, 254 SMO etc. which are much costlier than SS-316L and 317L.

6. Influence of Process Parameters :

The degree of protection, provided on some setting of potential and current, by this system to washers is highly dependent upon process parameters namely residual Cl_2, Cl− and pH. This dependence can be understood with the help of Fig. 4. One observe that with increase in Cl_2, corrosion rate of material increases. Corrosion potential also increases to more positive values, at which initiation of localised attack is much more readily than when Cl_2 concentrations are less. Under this situation, cathodic polarisation to a larger degree will be required and it will also require larger protection current.

With increase in Cl−ion concentration, the passivation range decreases. As such, one requires very good potential control system (potentiostat) so that during protection, the potential neither transfer to active region nor to transpassive region. Also, tendency to experience localised corrosion will increase hence larger protection current and polarisation potentials will be required.

With lowering of pH, distinctive between active and passive region becomes more and more, since the current value required to start passivation (Ipass) increases. As such this type of protection system becomes more effective for lower pH solution rather than for higher pH solutions.

7. Practical Aspects related to Application of Protection System :

In order that potential of washer always remains in passive region, the rectifier must be potentiostatically controlled. Potentiostatic control is also necessary as the current density demand, to keep washer at a certain potential, varies within wide limits. This is because one observes quite significant variation in residual Cl_2 (0-150 ppm, upper limit could be few thousand ppm sometimes) in washer, while in practice, which in turn governs current density required for protection. Thus design of protection system is not based on means value of residual chlorine, but on its maximum and minimum values.

Protection criteria can not be chosen simply on the basis of anodic polarisation behaviour but long time (upto at least a week) exposure at constant potential is also necessary. This is basically because it takes some time before pitting and crevice corrosion initiate. So it is quite possible that crevice corrosion might initiate even at a potential, which from anodic polarisation behaviours appears as protection potential (38).

Values of currents needed to obtain and maintain protection can be estimated from polarisation curve. These currents are time dependent and are usually larger than those determined in the plant. In one case, current densities needed to obtain protection dropped from 0.29 to 0.02 Amp/ft^2 when time to reach Protection potential was increased from 5 min to 30 min. As a principle, the current density required to obtain protection regulates the size of power supply chosen for installation. Similarly, current to maintain protection also decrease with increase in time to reach protection potential. Thus currents needed to maintain passivity, obtained from polarisation curve, are usually higher than required, and thus include an inherent safety factor (41).

Another factor that can be determined from laboratory investigations is retention of passive film without current flow, In some cases it is observed that, once the system has been shifted into passive region, it remains stable for some time (for days in some cases) with no current flow. This type of behaviour allows use of less complicated and thus more economical and on-off type of potential control. This type of situation is however, less likely to observe in case of Cl_2 water because of the presence of Cl− ion (41), which breaks passive film.

8. Economics and Commercialization :

To estimate benefits of electrochemical protection, a detailed calculation of the economics of using the protection system on washers has been made (39,40). (Details of calculation are shown as Appendix—A). Based on the following assumptions that :

i. Normal life-expectancy of washers is 4 years.

ii. Cost of 317L washer (excluding vat, drive) is US $ 520,000/-.

iii. Installed cost of Electrochemical protection system is US $ 55,000/- .

iv. Mill throughout is 250,000 tones per year, and washer size is 4m dia × 9m.

the calculation indicates that if washer's life is doubled i e. from 4 to 8 years, the payback period is about 1 year and if the life is quadrupled i.e. from 4 to 16 years, payback period is even less than 1 year.

It is worth mentioning here that the above calculation has not considered saving in chemical costs due to reduced consumption of SO_2 or NaOH used as antichlores (b) saving in maintenance and down-time costs. These costs could be very substantial and could easily overshadow the capital cost saving as described above.

In another study (42), comparison of cost incurred in forming tanks using differently available protection procedures has been done. One observes that anodically protected mild steel tank is cheapest in comparison to that of stainless steel and of glass lined mild steel. The same study also reveals that protection systems are more economical to use on larger tanks rather than smaller tanks as has been shown in following Table.

Capacity (Litres)	Cost (£)		
	Anodically Prot. Mild	Glass lined Mild Steel	Stainless Steel
30,000	2,000	2,500	3,500
450,000	6,500	10,500	17,000

The presently discussed protection system for bleach plant washers has found wide acceptability in North American mills. This is basically because—

(a) This alternative is more economical to use in comparison to other alternatives e.g. use of lined (nonmetal/metal) steels, upgradaded steels or nickel based alloys.

(b) Comparatively low cost of electrical protection system.

(c) Ease of retrofitting to an existing washer. Most of the systems have been installed to existing washers.

Wider acceptibility of this system in the North American Paper Industry can be gauged from Table 2.

TABLE—2

Adaption of Electrochemical Protection Systems in Bleach Plant Washers (12,37,40)

Year	Number of Installed Systems
1978	First System
1981	24
1982	>50
1985	≈ 36% of total chlorination washers.

9. Need of Electrochemical Protection System for Indian Mills :

Presently the electrochemical protection systems are non-existant in Indian mills. Are bleach plant washers in these mills not so corrosive as to warrant application of any protection procedures ? If at all there is any such need, perhaps electrochemical protection system will be worth trying because of the advantages associated with them, as discussed in earlier sections.

Corrosion tests in two of the larger Indian Paper Mills have been conducted in their bleach plants (21,22) Aims of these tests were to measure nature and degree of corrosion, corrosivity of liquid media and performance of materials. Materials selected for the test were austenitic stainless steel 304, 304L, 316L and 321 type. The selected materials are among the most commonly used materials for manufacturing process equipments. Some of the important findings of tests were—

i. In both the mills, all materials were very severely affected by pitting and crevice corrosion in chlorination washer. Thus even 316L is not suitable to use in making process equipments e.g. washer drum, wire cloth, pipes, pumps, valves etc for this stage.

ii. Hypochlorite washers showed moderate or practically no localised corrosion. However, crevice corrosion was severe in certain cases e.g. for all materials in hypochlorite 1 washer (wet/dry environment—similar to the one as experienced by washer drum), for 316L, 321 in liquid phase of hypochlorite washers of one of the mills.

iii. One of the mills showed comparatively much higher degree of corrosion attack. In this mill the process parameters of washer filtrate of all stages showed lower pH, higher residual Cl_2 and Cl^- content, all of which together help in deteriorating the environment for materials (22). Hence none of the tested materials is suitable to use where they could be exposed to gaseous phase e.g. shower pipe, upper part of washer and vat.

iv. The corrosivity of these plants was not as bad as of their counterparts in North American and Scandinavian mills. This is perhaps mainly due the fact that Indian mills practically do not practice filtrate recycling, whereas the western mills are almost 'closed'.

So one observes that at least in chlorination washer, the environments are so aggressive that SS-316L can not be considered as material of construction. And this is the condition when Indian mills are not practising filtrate recycling. Conditions will deteriorate further when recycling is adopted in order to control pollution and save energy and water. Similarly, chlorine dioxide is also likely to be adopted as one of the stages in bleaching sequence. Chlorine dioxide washer is equally corrosive as chlorination washer is In other words, in Indian mills also one has to think in terms of adopting some corrosion control strategy presently for chlorination washer and later for chlorine dioxide washer also. Among the available alternatives, anodic protection appears to be most cost-effective. Since use of upgraded alloy and that of non-metal lined alloy is a costlier proposition,

10. Future Plans:

Considering the above, we at the Institute have planned electrochemical polarisation measurements to be done on steels and nickel based alloys in bleach liquors in the laboratory conditions. Later these experiments will be performed in earlier studied mills because information about the corrosivity of their bleach plants is known to us. In order to establish cost effectiveness, mill tests like the ones performed earlier (21, 22) will also be conducted on upgraded alloys e.g stainless steels 317L, 904L, 254MO etc. and Ni based alloys e.g. Hastelloy C, Incoly, Inconel etc. These are among the candidate materials of construction for bleach plants.

Another important aspects of development of electrochemical protection system is that operational parameters of this system are very much affected by process conditions which vary significantly from mill to mill. As such, mill tests in several mills will be required to perform so as to ensure wider adaptability of the protection system. It is hoped that mills will come forward with all possible help in this endeavour.

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