CORROSION AND THE PROTECTION OF THE ENVIRONMENT*

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UDC 620.197

The author discusses data on the emission of biologically hazardous and corrosion-active substances (SO_2, dusts containing heavy metals, etc.) into the environment. It is emphasized that the cost of preventing emission should be compared with the economies similarly obtained in a number of fields — reduction in losses due to illness, loss of timber and crops, and damage to metallic and nonmetallic materials, in particular those obtained from scarce and nonrenewable raw materials.

In the industrially developed countries, the losses due to corrosion and the costs of protection against it constitute up to 3.5% of the gross national product [1]. The effectiveness of the increased application of up-to-date methods of protection from corrosion has greatly reduced the rising emission of aggressive gases and effluents. For this reason, since the start of the current century the loss of steel due to corrosion has been reduced to a roughly constant level of about 30% of the annual output [2]. In most industrially developed states the proportion of atmospheric corrosion in the total corrosion losses is at least 80% [3].

Atmospheric corrosion, which is largely due to the relative humidity of the air (>60%), is increased by the presence of acid gases and dust if the latter contains metals or has strong reducing properties.

Increased atmospheric corrosion is due to the emission of the same gases which are harmful to men, animals, and plants. Prevention of the emission of such gases will lead not only to preservation of the living world, reduction of losses due to illness, losses of crops,etc., but also to reduction in the loss of metal.

Therefore, in economic investigations the cost of prevention of emission must be compared with the economy achieved in all these fields. We may add that the metal lost by corrosion is an irreplaceable raw material, the reserves of which cannot be regarded as inexhaustible. In this connection we must note the value of secondary raw material (unit mass of utilized scrap metal replaces 1.3 units of mass of primary raw material [4]). In the industrially developed countries the distribution of emission of gases, smoke, and dust between different branches of industry is approximately reflected by the data in [5]: thermal electric power stations, 58%; nonferrous metallurgy, 22%; ferrous metallurgy, 16%; other branches (chemicals etc.), 4%.

Of course, these averaged data do not at all represent the local emission in the neighborhoods of the various industries. On the other hand, we must not underestimate the emission from dwelling houses. In large towns with large numbers of houses with heating stoves, in the winter the emission of sulfur dioxide from the houses may be greater than that from industrial premises.

The composition of the emissions can be estimated on the basis of approximate data for the Federal German Republic (1969) [6] in millions of tons per year: carbon monoxide 8; sulfur dioxide 4; nitrogen oxides 2; hydrocarbons 2; dust 4.

From the viewpoint of corrosion of metals, the most dangerous of these are sulfur dioxide and oxides of nitrogen, and also dust, especially if it contains noble metal impurities such as copper, mercury, etc., or sulfides and other substances acting as strong reducing agents. There are also other harmful substances — waste from chemical and related industries, such as

*Report presented to the Third Conference of Specialists of Member-Nations of the Council of Mutual Economic Aid [COMECON] on the problem of "Devising Methods of Protecting Metals from Corrosion," Warsaw, April 1980.

Scientific-Research Institute, Meinsberg, German Democratic Republic. Translated from Zashchita Metallov, Vol. 17, No. 1, pp. 3-10, January-February, 1981. Original article submitted August 5, 1980.

hydrogen sulfide (from synthetic fiber production), hydrogen fluoride (from aluminum and fluorspar processing), hydrogen chloride (from the production of potash fertilizers and other products such as polyvinyl chloride), chlorine (from plants for electrolysis of alkali metal chlorides), etc. [7]. In general, such substances are only of local importance in the neighborhoods of the corresponding industries and in the plants themselves. However, owing to the very high aggressiveness of these wastes, e.g., chlorine and hydrogen chloride or fluoride, their corrosive action in these zones is very strong and costly protective measures must be taken.

On the other hand, the harmful and in particular corrosive action of sulfur dioxide is spread out over wide territories of the industrially developed countries, since most of its emission is due to burning of fossil fuels. Therefore the dangerous SO_2 concentration (0.15 mg/m^3 for prolonged exposure) is never interrupted except in purely agricultural localities.

The annual world emission of SO_2 is 140-150 million tons, corresponding to over 70 million tons of sulfur. Over 93% of this belongs to the northern hemisphere of the earth [5]. The annual consumption of sulfur (for producing sulfuric acid and other sulfur compounds) is scarcely half of this amount. When sulfur-containing fuels such as coal and gas are used to produce energy, 80-90% of the emission is associated with their combustion [8]. In the German Democratic Republic the annual emission of sulfur dioxide is 5-6 million tons [9]; much of the SO_2 (about 5%) is oxidized to sulfuric acid, which is very toxic and corrosive. In southern Scandinavia the amount of sulfuric acid separating from the precipitation is about 1 $g/m^2 \cdot yr$; sometimes 0.1-0.2 g of H_2SO_4 per square meter separates from a single snow or rain shower [10]. However, the sulfur dioxide itself causes marked damage. Of course, the SO_2 content of the air varies over a wide range according to the sources of the emission; e.g., in industrial regions with large towns it reaches over 1 mg/m^3 and sometimes over 2 mg/m^3, but in territories with low population density, it is only about 1% of this value [11].

Owing to the global scale of SO_2 emission, its dangerous consequences have been studied very closely. Before turning to the corrosive action of SO_2 we may make a few remarks on its biological hazard. Although people actually start to choke only when the SO_2 content of the air reaches 15 mg/m^3 (100 times the concentration permitted for long-term exposure), in large towns and industrial regions even a concentration of over 1 mg/m^3, if exposure is prolonged and is combined with dust and other factors, can lead to bronchitis; it is estimated that in the USA about two billion dollars are lost every year from this disease owing to reduced productivity and absence from work through sickness. Growing crops and coniferous trees are especially sensitive to sulfur dioxide; in this case its harmful action is felt even at concentrations of 0.25 mg/m^3, and, for example, in the German Democratic Republic it leads to annual losses of timber totaling 30-50 million marks. It is to be remembered that today timber is becoming increasingly important as a fuel and raw material which is renewable with the aid of solar energy.

The corrosion losses of iron and steel rise sharply at SO_2 contents of over 1 mg/m^3, particularly when the relative air humidity is over 60%. In a rural atmosphere where the SO_2 content is about 0.06 mg/m^3 (approximately corresponding to separation of less than 20 mg SO_2 per square meter per day at the surface), the corrosion losses do not exceed 400 $g/m^2 \cdot yr$, whereas in an industrial atmosphere (over 40 mg SO_2 per square meter per day) they exceed 700 $g/m^2 \cdot yr$ [12]. As well as large steel surfaces subject to atmospheric corrosion, importance attaches to corrosion of steel structures used in electricity transmission lines. The conductors themselves are also subjected to aggravated corrosion under the influence of SO_2 [13].

According to Barton [14], the critical SO_2 load which can aggravate the corrosion of steel is 6-10 $g/m^2 \cdot yr$; for copper and zinc the value is about twice as great. However, in Paris after the start of the use of sulfur-containing fuels it was found that the rate of corrosion of galvanized roofs doubled. Detailed investigations revealed that hot-galvanized surfaces in industrial centers corrode 10 times faster than in rural sites [15]. The corrosion of aluminum is also accelerated by SO_2, though not so much as for steel and zinc.

The corrosion losses due to SO_2 can undoubtedly be reduced by desulfurization of fuels or by removing SO_2 from flue gases. At one chemical plant in the German Democratic Republic, the use of low-sulfur natural gas reduced the SO_2 emission by 3500 tons/yr. At another plant the introduction of apparatus for desulfurization of the flue gases reduced the SO_2 emission by 20,000 tons/yr [16]. In Czechoslovakia the replacement of high-sulfur brown coal by oil reduced the SO_2 emission by 16,000 tons/yr [17].

Many methods have been developed for desulfurizing oil and coal and for removing SO_2 from flue gases; although the necessary capital outlay is high, the cost of desulfurization increases the cost of energy only by 0.3-0.5 East German pfennigs (0.12-0.2 kopecks) per kilowatt-hour. Considering the various losses due to SO_2 and the high cost of additional corrosion protection, from the viewpoint of the economy as a whole this increase in the cost of energy seems perfectly justified.

As well as corrosion of metals, sulfur dioxide causes rapid deterioration of building materials such as concrete, marble, and sandstone. In Great Britain, the yearly losses due to SO_2 damage, including corrosion losses, are estimated at £150,000,000. This gas causes paper and other cellulose products to become weak and brittle.

Gaseous industrial wastes which differ in toxicity and ability to accelerate the corrosion process include hydrogen sulfide. This compound, which is at least 100 times more toxic than sulfur dioxide, is emitted by chemical plants producing sulfate cellulose and synthetic fibers and by metallurgical plants in the granulation of slags. Owing to its horrible smell, on the whole more care is taken to prevent pollution of the atmosphere by hydrogen sulfide than by sulfur dioxide.

Hydrogen sulfide not only aggravates the corrosion of steel and promotes dangerous stress corrosion cracking (mainly in the absence of oxygen), but is also a strong corrosion agent for copper and silver. Formation of poorly conducting sulfide layers on surfaces of these metals may lead to loss of electrical contact in electrical apparatus, switches, etc. The losses thus caused in electrical and electronic plant and equipment are an order of magnitude greater than the corrosion losses of metal. In our microelectronics age, damage to contacts by corrosion layers is so great that there is an exponential growth in the consumption of noble metals — Au, Pt, Rh, etc. Oxides of nitrogen are emitted by the chemical and mining industries and also by power plants, but mainly by automobiles and trucks (in the Federal German Republic, about 2,000,000 tons/yr). They are not only biologically very dangerous (the limiting tolerable concentration is 0.04 mg/m^3), but also accelerate corrosion, mainly of nonferrous metals, since they form nitric acid.

These harmful substances exert corrosion action not only from the gas phase but also when dissolved in water. The corrosive action of absolutely dry gases, including that on steel, is very slight. But at a relative humidity above 60% the metal surface becomes covered at least by an adsorbed film of water, so that electrochemical corrosion mechanisms are realized, in which water itself plays an important part.

Hydrogen chloride is emitted by potash fertilizer factories, chemical chlorinating plants, and garbage incinerators in which polyvinyl chloride production wastes are burnt. As well as raising the acidity and hence the toxicity of the water, hydrogen chloride causes strong corrosion of steel and even of corrosion-resistant chromium–nickel steels, because pitting corrosion of steels of this type develops in hydrogen chloride solutions. We may note that localized corrosion can arise even when the chloride content is about 2 mg/liter.

The possibility of increasing corrosion hazard by certain measures to protect the environment, such as burning waste, was recently very convincingly demonstrated [18].

On the other hand, during atmospheric corrosion a thin condensed film of moisture on steel can absorb appreciable amounts of HCl or NaCl, so that a high concentration of electrolyte may be created on the metal surface even when the air contains only small amounts of these substances.

The emission of highly toxic hydrogen fluoride is only of local importance, since this gas is emitted in biologically dangerous concentrations only by specialized production processes (electrolytic production of aluminum from cryolite and processing of phosphorites or fluorspar). However, hydrogen fluoride causes strong corrosion of metals which are very resistant to the usual chemicals (acids) and to atmospheric corrosion, e.g., titanium, zirconium, and aluminum. The annual emission of hydrogen fluoride in the Federal German Republic is estimated to be 200 tons. In this case also, measures to protect the environment in which a secondary raw material is also used may accelerate corrosion on account of enhanced addition of fluxes in processing aluminum scrap and release to the atmosphere of the combustion products and chloride and fluorode contaminants in the aluminum swarf [19].

All industrial plants, especially thermal power stations, and also metallurgical works, emit large amounts of solid wastes in the form of dust together with the gaseous waste. For

this reason the dust emission has rapidly risen since the turn of the century. The dust content of the air today is 20% higher than at the beginning of the century [20]. The rates of sedimentation of many dusts are so slow that they are carried away from the plants to the furthest corners of the earth, e.g., the Antarctic.

If the dust from electric power stations contains only insoluble salts such as calcium sulfate or carbonate, its sedimentation on the surface of iron is relatively safe. But soluble salts — sodium sulfate or chloride — dissolve in the film of moisture on the surface, increasing its electrical conductivity and thus the corrosion rate. If aggressive ions such as chloride ions are then formed, the corrosion danger, primarily that of pitting, becomes marked. If the dust contains carbon or other reducing substances, it can lead to reduction of the passivating layers on iron or chromium–nickel steels. Therefore, the metal becomes at least locally damaged by corrosion, and its own type of pitting arises.

Deposition of dust on steel, aluminum, or zinc is particularly dangerous if it contains more noble metals such as copper, mercury, etc., or their compounds. In this case, in particular, on account of contact deposition of the more noble metals from the solution, contact cells are formed on iron or aluminum articles, leading to enhanced corrosion.

As discovered in the USA, about 5000 tons of mercury are emitted every year with the light ashes from burning coal (about half the world production of this metal). This is not only dangerous to health and to nature in general, but presents a hazard of increased corrosion. While fallout of metallic mercury (probably present in dust) is relatively harmless to humans (it is organomercury compounds such as $HgCH_3$ which are highly toxic), on less noble metals (Fe, Zn, Al) it forms contact cells leading to rapid corrosion.

According to Swedish data [21] it is not enough to forbid the use of fungicides containing mercury. Removal of mercury-containing dust, which forms highly toxic compounds in living organisms, is necessary in the interests of conservation of the environment and of protection from corrosion.

Lead is another metal which is emitted into the atmosphere in huge amounts in dust (mainly as oxide) and greatly increases the atmospheric corrosion of iron, aluminum, and zinc. Most of its emission is in exhaust gases from automobiles and trucks. It is added to the fuel in the form of the antiknock agent tetraethyl lead. In the USA alone, 150,000 tons of lead are emitted into the environment in this way, and in the Federal German Republic 30,000-40,000 tons. Lead can also cause contact corrosion of the automobiles themselves, especially in the exhaust system [22], and in all structures of less noble metals. It is noteworthy that the toxicity of lead absorbed by organisms, especially children, has until recently been greatly underestimated, and as a result we now need to reduce the permissible lead content. In Great Britain with its heavy road traffic the lead content of the air is 0.5-10 $\mu g/m^3$. However, to avoid toxic action, especially on children, it should not exceed 0.7 $\mu g/m^3$ [24]. For this reason there is a pressing need to replace tetraethyl lead with other antiknock agents, not least because of the loss of irreplaceable raw materials. It is thought that world lead reserves amount to 93,000,000 tons (1970). Up to 1970, 3.4 million tons of lead have been produced per year; thus, at a constant rate of production the reserves will be exhausted even before 2000 A.D. [23].

The marked differences often observed between the corrosion behavior in a real atmosphere (e.g., in towns and industrial zones) and that in the laboratory may perhaps be due to the fact that in the laboratory no account is taken of contamination of the air by metals.

Aggressive components in the atmosphere (and above we are far from having mentioned them all) act particularly strongly when the air is moist, and of course this cannot be controlled in the open air. In principle, metals can be protected from aggressive components by organic coatings which themselves must be resistant to the atmosphere, its pollutants, and radiation. In many countries, up to 70-85% of the iron and steel is protected from atmospheric corrosion in this way (and also some aluminum and zinc). However, paints and varnishes give coatings which must be renewed every few years, since they are permeable to water, leading to rusting under the coating.

Regardless of the coating deposition technology, most of the solvents (hydrocarbons, chlorohydrocarbons, etc.) go into the atmosphere. Owing to the short service life of the organic protective coatings, not only are large amounts of valuable solvents lost, but the environment is polluted by the highly toxic vapors, which may give rise to nervous complaints, cancers, etc.

Therefore, care must be taken to recover the solvents by using appropriate suction and condensation equipment, especially in view of the increasing use of the toxic chlorohydrocarbons as solvents. In this field there are also considerable additional losses of products obtained from mineral raw materials. In many countries these losses amount to thousands of tons per year. Of course, recovery of the solvents requires heavy costs [25], especially if the protective layers are coated on structures in the open air. Therefore, corrosion protection by means of such coatings, which must often also be applied in 4-5 coats, is not only laborious but also dangerous to health and expensive from the viewpont of raw material reserves, and this is often left out of consideration in economic calculations. Therefore in any case, we must prefer a coating deposition technology not requiring solvents; such coatings are less permeable to water and give better corrosion protection. If in any highly developed industrial country practically only paints and varnishes are used as protective coatings, the emission of solvent vapors will be 4-5 kg per head of the population per annum. If in technical processes they emit to the atmosphere gases which are biologically and medicinally hazardous and which destroy materials, one must ask whether it is necessary to alter the production technology so as to eliminate them. If this is impossible, one must always reduce emission to a minimum by such technical means as absorbers, filters, etc. The necessary costs must be compared with the biological damage, especially the health hazard, and with the losses of materials both in the emission itself and on account of corrosion due to the emitted substances. Only in this way can we find the economically optimal solution to the protection of the environment from air pollution.

If in the present-day industrial atmosphere the corrosion losses are in all about 50% higher than in a rural atmosphere, this is naturally accompanied by a corresponding increase in stoppages, repair work, and other losses. In the future the increase of industrialization will increase the metal area requiring protection in the developing countries also, and since the corrosion rate rapidly rises with the temperature and air humidity, these countries must reckon on high corrosion losses. On the other hand, in all the industrial countries the increasing role of chemical processes in the national economy will lead to increased contamination of the atmosphere by corrosion-active gases. Therefore, in addition to organic and inorganic protective coatings, we must pay special attention to the development and adoption of low-alloy steels which "protect themselves" from corrosion owing to the formation of passive layers.

In the German Democratic Republic, steels of type KTS (with small amounts of copper and chromium) without paint are already recommended in an industrial atmosphere containing more than 1 mg/m^3 of SO_2 as being better than ordinary structural steel [26]. However, it remains an open question to what extent these steels will be able to retain their stability in the face of the rising aggressiveness of the atmosphere. Therefore, we must do all we can at least to prevent further increases in the emission of aggressive gases.

The use of high-alloy steels resistant to aggressive chemical reagents in chemical engineering will not be discussed here. We shll merely remark that the position with respect to raw materials used for the production of nickel and other alloying elements makes it necessary to use ceramic materials with the appropriate mechanical and thermal properties.

The contamination of rivers and lakes with effluent from industry, dwellings, and agriculture is in many cases threatening the very existence of life in the water. This threat even exists for the world ocean, e.g., in the Baltic sea, as a result of transport of pollutants by rivers, deposition of solid wastes, and the annual spillage of 5-10 million tons of oil from tankers, marine oil rigs, etc. Far from negligible is the pollution of water due to anticorrosion measures, viz., the deposition of galvanic coatings. The effluents from galvanizing plants are a source of high toxicity and of corrosion-active compounds of heavy metals (Ag, Cu, Pb) and such very toxic anions as CN^-.

Serious attempts have already been made (and they must be even greater in the future) to reduce the cyanide pollution of the environment by the use of cyanide-free electrolytes. Of course, this is possible only on a limited scale, and we must remember that the complex formers CN^-, EDTA, etc. are often dangerous not on account of their own toxicity but rather because they prevent the separation of heavy metals during purification of effluent, so that these metals get into the water. From this viewpoint, replacement of cyanide by other strong complex formers cannot be effective [27-29]. It must be remembered that even traces of salts of noble metals, particularly Cu, Hg, and Pb, which are in any case highly toxic and accumulate in plants and fish, are deposited (by cementation) on steel, zinc, or aluminum, and

cause very dangerous contact corrosion, presenting a hazard to ships and structures in the water. There are no reliable data on the additional corrosion losses arising in this way. However, it is thought that 5000 tons of mercury get into the world's seas and rivers every year. The upper reaches of the Rhine alone received 85 tons of this metal in 1970, in the form of compounds; to this must be added 1500 tons of lead per year [30]. The amount of copper must be of the same order. If we consider that the world resources of these metals are small, and that the recovery and processing of their ores require large amounts of energy, and if we remember the destructive action of these metals on life in the water, even without quantitative data on the additional losses due to corrosion, we must make every effort to remove them from the effluents; there are many technical possibilities for this purpose — precipitation, ion exchange, absorbents, and closed-cycle (effluent-free) processes.

Many examples of such methods have been described [29, 31-34], including those for other branches of industry, such as the use of ion exchange to obtain the scarce metals Ni and Cd from effluents from the plants for purifying furnace gases from lead works [35] or regeneration of Zn from effluents of synthetic fiber production [36].

With effluents from towns, chemical and mining enterprises, the water receives alkali and alkaline earth metal salts, which are almost nontoxic, but owing to their large amounts may disturb the biological equilibrium; in most cases they are chlorides, which sharply increase the corrosive aggressiveness of the water.

According to rough estimates [37], in the Federal German Republic the rivers receive 9-10 million tons of such salts per year, including about 5 million tons from mining enterprises and 3 million tons from the chemical industry.

The potash fertilizer industry, especially in the German Democratic Republic, produces a continually rising amount of magnesium chloride, which can shift the pH of water into a biologically hazardous region and can also stimulate corrosion. Since the pH has a decisive influence on biological processes in waters, in the German Democratic Republic all the effluents before arrival at the outlets are treated to establish a safe pH, which is also safe from the corrosion aspect. However, the increased chloride content itself aggravates corrosion, primarily pitting, in unprotected metal. This corrosion can be reduced by electrochemical protection, at least in the case of iron, as done in seagoing ships on a large scale. However, for structures in rivers and lakes and river boats, little is done in this respect, and in the case of a large number of relatively small objects the costs would be very high.

Thus, the close relation between corrosion and pollution of the environment requires general consideration from the technical and economic viewpoints.

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PROTECTION OF STEEL STRUCTURES FROM CORROSION*

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UDC 620.197

Paints and varnishes do not give anticorrosion protection of steel structures in an aggressive atmosphere. The authors assess the economic advantages and prospects for using low-alloy steels with enhanced resistance to atmospheric corrosion, electrolytic aluminum coatings, hot galvanizing, etc. Further improvements in protection will require differentiated selection of methods; account must be taken of a) the correctly determined complex of corrosion and industrial load, b) the standard conditions and times of renewal of the protection for all parts and units of a structure, and c) the overall costs over the whole service life. The authors discuss particular ways of realizing these principles and the problems awaiting solution.

The production of steel structures is a field in which a correct and well-differentiated approach to the choice of anticorrosion protection with the aid of all the means available for this purpose can greatly reduce the losses. However, the importance of improving the protection of steel structures from atmospheric corrosion is largely governed by three factors: the huge number of structures in use, the necessity of ensuring long service lives, and the high cost of replacing short-lived paint coats.

As an example, we may cite certain data from our experience in Czechoslovakia. At the end of 1975 the area of steel structures was about 140,000,000 m^2 and their mass was 6100 tons. In 1977, 340,000 tons of steel structures were produced. Many of these new structures were protected by paint coats. About 12% of the structures are at present protected by metallization with aluminum, 12% by hot galvanizing, and less than 2% are made from low-alloy steels with enhanced resistance to atmospheric corrosion. Since over 50% of the steel structures in use in Czechoslovakia are subject to the action of an atmosphere with degree 4 or 5 of aggressiveness, the paint coats presently in predominant use are economically ineffective, because they give satisfactory resistance only in an atmosphere with a degree of aggressiveness not over 2.

The paint coats on steel structures used in an aggressive atmosphere cannot at present be restored in the required amounts and with the required quality, owing to a shortage of labor.

*Report presented to the Third Conference of Specialists of Member-Nations of the Council of Mutual Economic (COMECON) on the problem of "Devising Methods of Protecting Metals from Corrosion," Warsaw, April, 1980.

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