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Scientific Paper

HAZARDOUS AGENTS IN ANODE MANUFACTURE*

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The aim of this study was to assess to which extent the modernisation of an anode plant had reduced occupational chemical health hazards for jobs with the highest potential of exposure. Periodical measurements of dust and gases were performed at the same workplaces using the same methods, before and after modernisation. These measurements were compared with the recommended standards. Before modernisation the concentrations of total dust, carbon monoxide, carbon dioxide, sulphur dioxide, hydrogen fluoride, benzene, and phenol were above the recommended standards in 56.9 % (74/130) of the samples. After modernisation, only 12.3 % (21/171) of the samples were non-conforming. Before modernisation, workers were exposed to higher concentrations of all agents in all production sections. After modernisation, dust remained the primary pollutant in harmful concentrations in the anode baking furnace (GM=22.1 mg m^{-3}) and in the anode rodding room (GM=22.1 mg m^{-3}), hydrogen fluoride in the anode rodding room (GM=4.2 mg m^{-3}), and sulphur dioxide in all production sections. As plant modernisation has not completely resolved the exposure issue, stringent compliance to safety rules and regular medical checkups are necessary.

KEY WORDS: *aluminium production, benzene, carbon dioxide, carbon monoxide, dust, gases, hydrogen fluoride, occupational exposure, phenol, sulphur dioxide*

In the primary aluminium industry, series of cells called "pots" are used to reduce alumina (Al_2O_3) to aluminium by electrolysis. These pots may be of two types, Søderberg and prebaked. The main difference between them is in the way in which anodes are supported. In both technologies, the anode is a mixture of petroleum coke and coal tar pitch, mixed and pressed to form a semisolid paste. In potrooms using the Søderberg technology green anodes are continuously baked in uncovered pots and new fresh paste is regularly supplied during electrolysis. This involves exposure of potroom workers to very high coal tar concentrations in volatiles evaporated from the anode top. Prebaked anodes are produced outside the potroom in a separate section. In modern smelters, prebaking potrooms are preferred because of the lower levels of emission and because the prebake technology allows a more automated process with hoods covering the pot (1).

Workers are exposed to coal tar pitch volatiles and more specifically to polycyclic aromatic hydrocarbons (PAH) and to inorganic gases hydrogen fluoride, sulphur dioxide, carbon monoxide, and carbon dioxide (2, 3). At the same time, workers are exposed to hazardous physical agents: noise, magnetic fields, and radiation (4, 5).

The harmful effects of these pollutants can be seen in the lungs, on the skin, and in the central nervous system. Respiratory disorders were reported

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as early as 1936 by Frostad (6), who observed asthma attacks among Norwegian potroom workers. Combined exposure to dust and fluorides can lead to the development of the so called "potroom asthma" (7, 8). Dermatitis is common in the exposed workers, characterised by oedema, erythema and sometimes by skin erosions (9). Harmful effects on the central nervous system include behavioural disorders, tremors, difficulty moving and memory and concentration disorders (10, 11).

Production of aluminium and aluminium alloys in Aluminij d.d., Mostar, Bosnia and Herzegovina, takes place in several plants, the anode plant, electrolysis plant, casting, and the gas processing plant (Figure 1) (12). Aware that a 30-year-old technology could not compete with other aluminium manufacturers, the management of the company started to modernise all plants. The modernisation of the anode plant included computerised dosing of coal tar pitch, replacement of the fire lead system, and control of the anode baking process. For that purpose new chambers were built in the anode furnace, the crane for cleaning anode butts from electrolytic cells was modernised, and semi-automatic equipment was introduced for cleaning grey cast-iron. All sections of the plant are semi-automated and several free-steered diesel vehicles are used for material transportation. The factory has its own anode production with annual capacity of 130,000 tons of green anodes and 60,000 tons of baked anodes. Today, thanks to modern technology, Aluminij has become the largest and technologically most advanced aluminium manufacturer in the Southeast Europe, with annual production of 125,000 tons of high-quality aluminium, of up to 99.9 % purity (13).

Along with the care about the quality of products and profit, the management makes a great effort to protect workers' health and the environment. Measurements of hazardous chemical and physical agents in the work environment were performed regularly in accordance with national law and regulations (14, 15).

In this study, we compared the concentrations of hazardous chemicals before and after the modernisation of the anode plant. In addition, we categorised the production stages (jobs) in the anode plant with the highest potential for occupational exposure. These data can be used to improve safety at work.

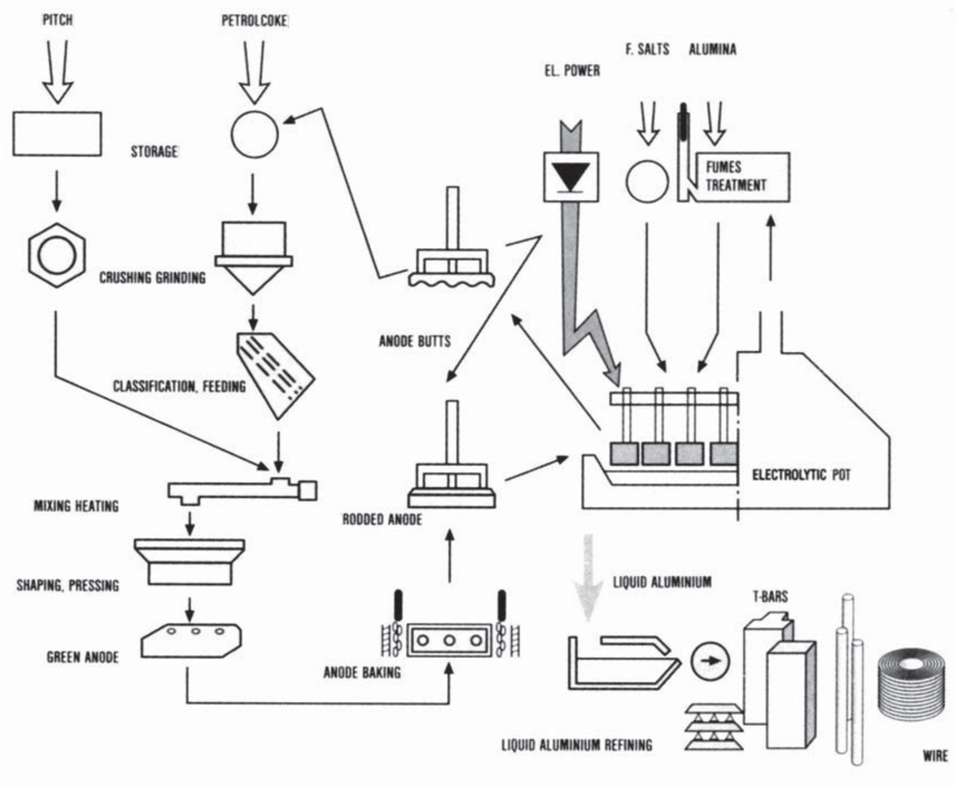

Figure 1 *Aluminium production flow chart in Aluminij d.d. Mostar (adjusted from reference 12)*

MATERIALS AND METHODS

Plant description

Anode production is an important part of aluminium production. In Aluminij d.d. Mostar anodes for 254 pots, used for the reduction of alumina, are produced in a nearby carbon electrode section of the anode plant. In the green mill section of the anode plant, crushed petroleum coke and recycled anode butts are mixed with liquid pitch to form anode paste and then are compacted into big green blocks. Crude anodes are moved to the oven section where they are baked in deep brick-lined pits at around 1,100 °C for 21 days. About 216 prebaked anodes are produced daily in two wide baking rooms. The baking calcines the binding pitch and ensures that the anodes have good thermal and electrical conductivity. After the baking, the anodes are rodded with cast iron and connected to aluminium rods in the rodding room. As finished products, the anodes are stored or moved to the potrooms for positioning into the pots. Twenty prebaked anodes are placed in each pot every 14 to 28 days. The exhausted anodes are removed from the pots, crushed to granules in the rodding section, and sent back to the green mill section for recycling.

Measurements

Mandatory periodical measurements of chemical agents in the anode plant were made in 1988, before its modernisation, and in 2004, after modernisation. Samples were collected during 6-hour shifts over five days in the presence of workers. Means of three measurements were taken as true measurement values. In both study periods, measurements were taken at the same workplaces using the same methods. The results were then compared to recommended standards (16, 17). The effect of modernization on the concentrations of chemical substances was estimated comparing the measurements from both periods.

Dust

Dust in the working environment was collected using an aerosol monitor device (Model 8520, Dust Trak, TSI Incorporated, Shoreview, MN, USA) and measured as the concentration of total dust particles. Dust samples were collected during 6-hour shifts over five days in the presence of workers. The mean concentration (mg m^{-3}) was compared with the maximum allowed concentration (MAC) (18).

Gases

Gas concentrations were measured using a universal device for detecting and measuring the emission and diffusion of gases in the workplace atmosphere MIRAN SapphIRE-100/100c (Foxboro Co., Foxboro, MA, USA). The gases measured were carbon monoxide, carbon dioxide, sulphur dioxide, hydrogen fluoride, benzene, and phenol. At least three measurements were made at different locations plant locations. The mean concentrations (mg m^{-3}) were compared with MAC (18).

Statistical methods

The chi-square test and Fisher's exact test were used to test the differences between measurements before and after the plant was modernised. The level of $P<0.05$ was considered statistically significant. Geometric mean (GM) was calculated for the log normal distribution of the measurements. All statistical analyses were performed with Statistical Package for Social Science (SPSS) 11.

RESULTS

Harmful substances were detected and their concentration determined in 130 samples before and in 171 samples after the modernisation of the anode plant. Table 1 shows the concentrations of total dust, carbon monoxide, carbon dioxide, sulphur dioxide, hydrogen fluoride, phenol, and benzene before and after modernisation.

Total dust, carbon monoxide, carbon dioxide, sulphur dioxide, hydrogen fluoride, benzene, and phenol before modernisation exceeded MAC (18) in 56.9 % (74/130) of samples. Concentrations of total dust (GM=26.4 mg m^{-3}) exceeded the recommended values in 73.1 % (19/26) of samples (Table 1). In five samples this excess was as high as eight times the threshold concentration. Sulphur dioxide (GM=17.2 mg m^{-3}), hydrogen fluoride (GM=2.2 mg m^{-3}), carbon dioxide (GM=5,385.3 mg m^{-3}) and carbon monoxide (GM=23.3 mg m^{-3}) exceeded MAC in 71.4 %, 46.7 %, 50.0 %, and 43.5 % of the samples, respectively (Table 1).

Measurements of the same harmful substances after modernisation showed significantly lower concentrations in 12.3 % (21/171) of the samples. Significantly lower were the concentration of total dust ($P<0.041$), carbon dioxide ($P<0.001$), carbon

Table 1 *Concentrations of environmental pollutants in the anode plant*

Hazardous agent	Before modernisation N	GM/ mg m^{-3}	Range/ mg m^{-3}	>MAC/ %	After modernisation N	GM/ mg m^{-3}	Range/ mg m^{-3}	>MAC/ %	MAC/ mg m^{-3}	Test of sig.	P value
Total dust	26	26.4	3.2-128.7	73.1	14	14.6	2.7-50.2	35.7	15	X^2=5.29	0.041
Carbon dioxide	22	5385	91.5-20075	50.0	19	238	199.5-3950	0	9000	X^2=12.98	<0.001
Carbon monoxide	23	23.3	0.58-75.4	43.5	17	0.2	0.1-12.3	0	50	*	0.002
Sulphur dioxide	21	17.2	0.3-53.2	71.4	14	7.4	0.5-38.0	35.7	4	X^2=4.38	0.037
Hydrogen fluoride	15	2.2	0.7-10.0	46.7	16	0.3	0.1-5.3	12.5	2.5	*	0.054
Benzene	12	28.5	3.6-52.3	58.3	11	0.6	0.2-3.6	0	15	*	0.005
Phenol	11	5.3	0.4-23.1	45.5	11	0.5	0.07-16.1	9.1	1.2	*	0.149

N - total number of samples;
GM - geometric mean;
MAC - maximum allowable concentration;
>MAC / % - percentage of determinations exceeding the maximum allowed concentration
**Fisher's exact test*

Table 2 *Concentrations of environmental pollutants in the anode plant by production stages*

Production stage/ Hazardous agent	Before modernisation N	GM/ mg m^{-3}	Range/ mg m^{-3}	>MAC/ %	After modernisation N	GM/ mg m^{-3}	Range/ mg m^{-3}	>MAC/ %
Green Mill								
Total dust	10	16.0	6.5-31.3	60.0	4	5.2	2.7-7.9	0.0
Carbon monoxide	8	23.7	0.6-72.7	50.0	7	0.2	0.6-40.6	0.0
Carbon dioxide	8	10346	3618-19746	75.0	7	1039	194-3888	0.0
Sulphur dioxide	9	18.5	8.1-53.2	66.7	5	6.9	0.8-23.7	20.0
Hydrogen fluoride	4	1.7	1.4-1.9	0.0	5	0.08	0.01-0.9	0.0
Benzene	4	10.2	3.6-25.4	0.0	4	0.3	0.03-2.1	0.0
Phenol	4	8.1	4.0-23.1	50.0	4	1.0	0.1-11.7	25.0
Anode baking furnace								
Total dust	6	27.6	14.3-56.4	83.3	4	22.1	7.6-43.8	75.0
Carbon monoxide	8	13.1	5.8-58.6	37.5	8	0.1	0.01-0.2	0.0
Carbon dioxide	8	1861	18-13410	25.0	8	64.2	0.2-688	0,0
Sulphur dioxide	6	11.1	2.1-38.0	66.7	5	6.6	0.3-38.0	40.0
Hydrogen fluoride	6	1.6	0.7-3.0	33.3	8	0.3	0.1-1.6	0.0
Benzene	5	47.3	38.0-52.3	80.0	5	0.6	0.03-2.0	0.0
Phenol	3	12.6	8.8-16.5	66.7	5	0.4	0.04-0.4	0.0
Anode rodding room								
Total dust	10	42.2	12.5-128.7	80.0	6	22.1	6.3-50.2	33.3
Carbon monoxide	7	44.4	12.3-73.7	42.9	2	4.5	1.6-12.3	0.0
Carbon dioxide	6	8750	2754-18062	50.0	4	235	214-280	0.0
Sulphur dioxide	6	23.2	10.1-44.4	83.3	4	12.7	8.4-19.9	50.0
Hydrogen fluoride	5	3.7	2.1-10.0	100	3	4.2	3.2-5.3	66.7
Benzene	3	48.5	45.7-50.8	100	3	38.0	37.3-38.0	0.0
Phenol	4	4.1	3.3-5.12	25.0	2	0.4	0.3-0.4	0.0

N - total number of samples;
GM - geometric mean;
MAC - maximum allowable concentration;
>MAC / % - percentage of determinations exceeding the maximum allowed concentration
**Fisher's exact test*

monoxide ($P<0.002$), sulphur dioxide ($P=0.037$), and benzene ($P<0.005$). The concentration of respirable dust, which was not measured before modernisation, exceeded the recommended value in 11.59 % (8/69) of the samples. In four samples the concentration was six or eight times higher than the threshold limit value of 5 mg m^{-3}. Concentrations of hydrogen fluoride (GM=0.313 mg m^{-3}) and phenol (GM=0.5 mg m^{-3}) dropped to below the recommended values (Table 1).

Table 2 shows the concentrations of harmful substances by production stages. Before modernisation, workers were exposed to high concentrations of dust, carbon monoxide, carbon dioxide, sulphur dioxide, and phenol throughout the production stages. During anode mixing and shaping in the green mill the concentrations of these harmful substances exceeded MAC in 51.1 % (24/47) of the samples. Hydrogen fluoride and benzene were found in concentrations lower than recommended. Total dust (GM=16.0 mg m^{-3}), sulphur dioxide (GM=18.5 mg m^{-3}), carbon monoxide (GM=23.7 mg m^{-3}), and carbon dioxide (GM=10,346 mg m^{-3}) were above MAC in more than half the samples, and sulphur dioxide exceeded MAC about 5 times.

In the anode baking furnace, high concentrations of all harmful chemicals were measured in 52.4 % (22/42) of the samples. Total dust (GM=27.6 mg m^{-3}) and benzene (GM=47.3 mg m^{-3}) exceeded MAC in over 80 % of samples. In the rodding room, where the baked anodes are fixed with a steel stub, the concentration of pollutants was above MAC in 68.3 % (28/41) of the samples. High concentrations of hydrogen fluoride (GM=3.7 mg m^{-3}) and benzene (GM=48.5 mg m^{-3}) were measured in all samples (Table 2).

After the modernisation, the concentrations of harmful substances dropped in all the anode plant sections. Sulphur still exceeded the recommended value in all sections, while dust remained higher in the anode baking furnace (GM=22.1 mg m^{-3}) and the rodding room (GM=22.1 mg m^{-3}) in 75.0 % and 33.3 % of the samples, respectively. Hydrogen fluoride (GM=4.2 mg m^{-3}) still exceeded MAC in the rodding room in 66.7 % of the samples.

DISCUSSION

After the modernisation of the anode plant, the level of workers' exposure to hazardous substances dropped significantly in all its sections. Our results suggest that the working conditions in the anode plant today correspond to those in Norwegian aluminium plants (19). The atmospheric concentration of chemical contaminants varies between plants, and usually depends on the technology used (4).

Measurements taken before the anode plant was modernised revealed the presence of dust with coal tar pitch and carbon in all anode production stages in concentrations up to eight times higher than the allowed maximum. After modernisation, dust concentration dropped significantly, but is still harmful to worker health.

In addition to the dust, sulphur dioxide, carbon monoxide, and carbon dioxide are emitted, especially from the pitch during the long period required for calcining the blocks. These gases are very dangerous to the environment and are recognised as major hazards to worker health. Before modernisation, high levels of these pollutants were measured at almost every workplace. With modernised technological process and use of coke with low sulphur percentage, sulphur dioxide concentrations dropped, but could not be completely eliminated because of the nature of the anode production process (20). Carbon monoxide and carbon dioxide concentrations dropped to a minimum, and work conditions that meet modern standards of aluminium production have been achieved (10).

Pollutants emitted from the bake furnace include polycyclic organic matter and other hydrocarbons that develop from heating and carbonising the paste binder pitch in the ring furnace. The pitch contains polycyclic organic matter that is recovered as tar from coking. During this process, about 80 % of all volatile compounds evaporates. Workers in the baking and rodding rooms were exposed to relatively higher coal tar pitch volatile concentrations than workers in the green mill. Our results are not comparable with Canadian studies where these concentrations were the highest in the green mill during mixing and shaping of the anode paste (21).

The level of PAH mainly depends on the quality of anodes and production technology. When prebaked anodes are produced, PAH emission equals 0.05 kg t^{-1} while when Sóderberg anodes are used, this emission rises to 25 kg t^{-1}. Over the recent years, PAH levels have dropped significantly. The new anode prebaking technology used by Aluminij Mostar has reduced the PAH emission to less than 0.01 kg t^{-1} (22).

After modernisation, high concentrations of hydrogen fluoride have remained an issue in the rodding room. Fluorides emitted from the furnace originate from exhausted anodes, removed from pots. The most common device used to control the emission of hydrogen fluoride and PAHs from anode baking furnaces is the dry alumina scrubber system. Likewise, high concentrations of hydrogen fluoride were present in all Aluminij Mostar plants (23). Operating procedures and work practices can have a direct effect on emission control. The quantities and composition of emission are strongly influenced by operating conditions, temperature, degree of automation, method of crust–breaking, and housekeeping. The experience and the motivation of the workers and their way of handling materials and equipment may also be of importance (22).

In anode manufacturing shops exhaust ventilation equipment with filters should be installed. Enclosure of pitch and carbon grinding equipment further effectively minimises exposure to heated pitches and carbon dusts. According to the work safety guidelines, workers have to wear personal protective equipment as well as respiratory protection from harmful gases and aerosols (14). Regular checks of atmospheric dust concentrations should be followed up by medical checkups when necessary.

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Sažetak

ŠTETNI AGENSI PRI PROIZVODNJI ANODA

Cilj je rada procijeniti učinak modernizacije tehnološkog procesa u Tvornici anoda na prisutnost i razinu koncentracije prašine i plinova štetnih za zdravlje radnika u radnom okolišu, kao i na poslove s velikim potencijalom za izloženost zaposlenih. U tu svrhu uspoređivani su rezultati obveznih periodičkih mjerenja kemijskih čimbenika provedeni prije i nakon modernizacije. Mjerenja su provedena na istim radnim mjestima i istim metodama tijekom radnih smjena i uspoređeni sa sadašnjim nacionalnim Standardom. Prije modernizacije, koncentracije ukupne prašine i plinova: ugljikova(II) oksida, ugljikova(IV) oksida, sumporova(IV) oksida, fluorovodika, benzena i fenola prelazile su preporučene vrijednosti u 56,9 % uzoraka, a nakon modernizacije u 12,3 % (21/171) uzoraka. Prije modernizacije radnici su istodobno na velikom broju radnih mjesta svih odjela bili izloženi prekomjernim koncentracijama štetnih kemijskih čimbenika. Nakon modernizacije prašina je i dalje prisutna u visokim koncentracijama pri pečenju anoda (GM=22,1 mg m^{-3}), kao i pri zalijevanju anoda (GM=22,1 mg m^{-3}), a geometrijska sredina koncentracije fluorovodika pri zalijevanju anoda iznosi 4,2 mg m^{-3}, dok je sumporov(IV) oksid prisutan u svim fazama proizvodnje anoda u koncentracijama štetnim za zdravlje radnika. Modernizacijom tehnološkog procesa smanjene su prisutnost i koncentracije kemijskih čimbenika u radnom okolišu. Međutim, izloženost prašini, sumporovu(IV) oksidu i fluorovodiku samo je djelomično smanjena.

KLJUČNE RIJEČI: *benzen, fenol, fluorovodik, plinovi, prašina, profesionalna izloženost, proizvodnja aluminija, sumporov(IV) oksid, ugljikov(II) oksid, ugljikov(IV) oksid*

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