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Workshop on Radioactive Contaminated Metallurgical Scrap

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Prague, Czech Republic, 26-28 May 1999

MEASUREMENT OF RADIOACTIVITY IN STEEL

(Prepared by H.-J.von Wachtendonk, S. Lüngen and N. Wilke, Thyssen Krupp Stahl AG, Germany)

This paper has been issued without formal editing by the secretariat.

Calibration Conditions

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Calibration source with certificate
- Date of reference
- Information of the nuclides
  - Energy
  - Half life of γ-nuclides present
  - Possibility of transition
  - Activity of γ-nuclides (with uncertainty range)

The gamma emissions should cover the expected range of gamma-energies.

Sample and calibration source must have the same geometry.

Each peak used for calibration should amount to > worr counts.

Introduction:

ThyssenKrupp Stahl is a big producer of steel in Germany. ThyssenKrupp Stahl recycles thousands of tons of steel scrap per month. With recycling the danger rises that a hidden radioactive source is introduced to the melting shop. Therefore, all incoming scrap is measured to detect those sources, but there remains a small risk that a weak or well hidden source passes this control. Therefore, the steel product is also measured to assure that the delivered steel is free of any artificial radioactivity.

Measurement of solid steel samples:

The Chemical Laboratories, department of production control, have overtaken the task to check each heat for radioactivity content. There are two reasons to perform this physical measurement in the chemical laboratory. The first reason is that the laboratory works 24 h per day, every day of the year. The second reason is that the steel sample analysis is performed by a fully automated system. The radioactivity measurement device is integrated into this automated system. So it is ensured that each heat is checked automatically. In figure 1 you can see the layout of one of the ThyssenKrupp Stahl production control laboratories.

The steel samples are taken by the steelworks personnel and sent to the laboratory by an automated tube system. All following steps are performed in automatic mode:

- Unpacking of sample
- Transport to grinder
- Grinding
- Transport to AE-spectrometer
- Spectrometric analysis
- Optionally: transport to x-ray spectrometer and measurement
- Transport to γ-ray detection unit
- γ-ray check
- Transport to marking device and sample storage

As the sample throughput is high (>10000 samples per month) the available measurement time is limited. As a consequence of this a precise nuclide specific measurement isn't possible as a multi channel analysis of the sample takes too much time, especially, when the radionuclide content is close to zero.

Therefore, we use a simple γ-ray spectrometer equipped with a NaI-detector as a single channel analyser with an upper limit of 1.33 MeV. The alarm threshold is calibrated with a pressed powder standard containing around 1 Bq/g Co-60 at the time of purchase. Whenever used the standard is corrected for decay. The production puts four requirements to the γ–measurement:

- It must be fast.
- It must be accurate.
- It must be sensitive.
- It must be cheap.

The sodium iodide detector used in this spectrometer is more sensitive (shows a higher efficiency) to radiation of lower energy than to higher energetic γ–emissions. The detector overestimates the lower energetic radiation compared to the 1.33 MeV radiation of Co-60.

Automatic Laboratory

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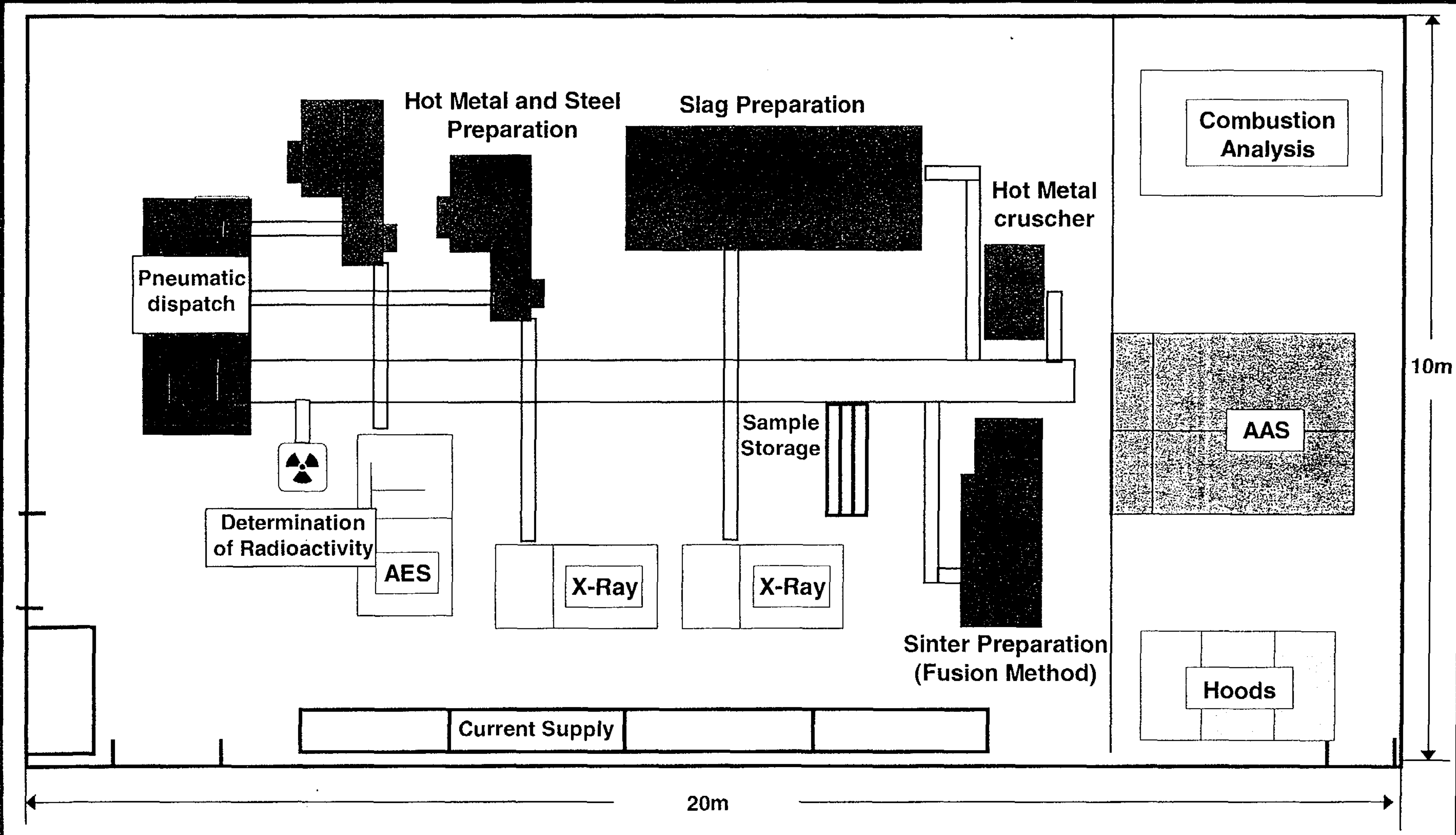

Measurement of slag samples:

Slag samples of blast furnace or converter process are not measured routinely. They show a higher radioactivity due to the enrichment of natural occurring nuclides like uranium and thorium during the production process. These elements are introduced by the process materials as minor component of iron ore, coke and coal used for heating and reduction. So they are unavoidable. During the production process the equilibrium between mother nuclide and daughter nuclides is disturbed as many of the daughter nuclides evaporate at the high temperatures applied during the process. Nevertheless, figures 5 and 6 show that many of the daughter nuclides can be detected when measuring γ–emissions. A quantification is difficult as similar standards are not available. Procedures for wet chemical isolation and analysis are under development.

Another possibility is the measurement of uranium and thorium by spectroscopic methods like ICP-AES or ICP-MS. In the case of uranium one can get additional information about the isotopic composition when using ICP-MS. Uranium of natural origin and enriched or depleted uranium can be distinguished. To measure other radioactive nuclides by these means is only possible when large amounts are present, normally only natural occurring nuclides. For the expected low content of artificial nuclides these methods aren't sensitive enough.

A third method for detecting radioactive nuclides in slag is α–spectrometry. This method is time consuming and costly as a representative part of the sample must be dissolved completely, the α–active nuclides must be separated (e.g. by electrodeposition) and measured in an α–spectrometer. Such a device is available at ThyssenKrupp Stahl, but methods are still under development. The method shall be applied if it is assumed that α–active nuclides may be present not showing characteristic γ–emissions (e.g. Am-241, Pu-isotopes). The measurement may be necessary to calculate the radiation dose to steelworkers who were exposed to α–active species containing dust.

Conclusions:

At ThyssenKrupp Stahl several simple γ–detectors are in use to check each heat rapidly for the absence of artificial radioactive nuclides. In the case of detecting a radioactive steel sample a high resolution γ–spectrometer is at our disposal to be able to evaluate which nuclides are present and in which amount. Methods are partially developed. The lack is the absence of appropriate standard material (steel standards containing Co-60 and/or other nuclides).
Conventional spectrometry (ICP-AES, ICP-MS) can be applied to some natural radioactive nuclides if they are present in the order of 0.1 to 1 ppm by weight.
Additionally, α–active species can be measured when the other methods can't be applied (concentration below detection limit of ICP-spectrometry and no appropriate γ–emissions). Sample preparation and measurement methods are under development.
It isn't planned to check for pure β–emitting nuclides because the effort for isolation and identification is too high.

Blast Furnace Slags

Time of measurement		1h		2h		4h		8h		16h	
	Total activity	0,45		0,58		0,94		0,60		0,74	
Nuclids	Energy [keV]	Identificated Nuclids / Securitiy of Identifcation [%]									
Ac-228	89,95								**41,7**	X	**61,5**
	93,35							X		X	
	209,28							X		X	
	338,32							X		X	
	463,00									X	
	794,70									X	
	911,60							X		X	
	969,11							X		X	
Bi-212	727,17									X	**60,5**
Bi-214	609,31	X	**38,8**	X	**35,4**	X	**54,4**	X	**64,7**	X	**76,7**
	768,36									X	
	934,06									X	
	1120,29	X				X		X		X	
	1238,11							X		X	
	1729,60							X		X	
	1764,49			X		X		X		X	
Cd-109	88,03									X	**95,2**
K-40	1460,81			X	**95,0**	X	**99,1**	X	**98,5**	X	**96,9**
Pb-212	74,81	X	**55,4**	X	**77,6**	X	**77,8**	X	**87,8**	X	**90,8**
	77,11			X		X		X		X	
	87,20							X		X	
	89,80									X	
	238,63	X		X		X		X		X	
Pb-214	74,81			X	**90,6**	X	**90,4**	X	**96,4**	X	**98,0**
	77,11			X		X		X		X	
	87,20							X		X	
	89,80									X	
	241,98			X		X		X		X	
	295,21			X		X		X		X	
	351,92			X		X		X		X	
Ra-226	186,21	X	**99,7**			X	**99,5**				
U-235	89,96		**47,1**		**52,0**		**47,4**		**51,6**	X	**53,0**
	93,35			X				X		X	
	185,71	X		X		X		X		X	

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Nuclides detected in blast furnace slags

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THE SCRAP SITUATION AFTER THE BOSNIAN WAR: ITS SOURCES, QUALITY, REGULATION AND CONTROL

(Prepared by Mr. P. Derviš, Bosnian Foundrymen's Association, Bosnia and Herzegovina)

Summary

Before the war, the Bosnian foundry and steel industry were supplied with scrap partly from domestic sources but mainly from foreign market. The annual steel production before the war recorded 130,000 tonnes of casting and 2,000.000 tonnes of crude steel. Most of the scrap was imported to secure and stabilize the production. During and after the war, despite a significant loss of production, efforts have been made to return to the normal production level in many ways.

In the wake of the war, there has been a growing concern over the import of radioactive contaminated metallurgical scrap or low quality raw materials which are uncontrolled or of unidentified sources. In this regard, it is urgently required to establish an effective system to prevent from, to detect and to control the flow of the radioactive contaminated metallurgical scrap.

The system should be established in such a way that all sorts of radioactive metallurgical elements should be controlled and prevented from use in all the metallurgical manufacturing processes, ferrous and non-ferrous alike.

GE.98-33010

The coverage of control should start from the border or (air) port checkpoint where the flow of the scrap begins to the final steel product. The control system should take a form of internationally common and acceptable standards and regulations. Equipments and measurement techniques should also be internationally common.

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MEASUREMENT OF RADIOACTIVITY IN STEEL

(Pre[illegible]ed by H.-J. Wachtendonk, S. Lüngen and N. Wilke,
Thyssen Krupp Stahl AG, Germany)

Summary

Even after the control of scrap deliveries, there remains a small risk that the radioactive contaminated scrap passes the detecting devices. Therefore, the chemical laboratory takes a role to measure each heat for the absence of artificial radioactive nuclides with a g-spectrometer equipped with Nal-detector. As the measurement must be performed in sequence with the steel production process, the allowable time for the measurement is quite limited. On the other hand, there could be still some possibility that background radiation might be present as the samples may contain some natural radioactivity. The task is how to differentiate the nature of radioactivity between naturally remaining radioactivity within safe limit and artificial nuclides present in the sample at a low level even though a very small amount of radioactivity could be detected in short time in both cases.

We have set the alarm limit to 0.1 Bq/g for Co-60 as indicating nuclide. This limit is set more than 4 s (s=standard deviation) from the average background radiation. Therefore, false alarms are quite improbable.

GE.98-33014

Strategy

The NaI-γ-spectrometer performs a gross-γ-measurement but it can not differentiate the nature of the nuclides present. If the alarm limit is hurt, the sample is measured on a high resolution of γ-spectrometer with Ge-detector for identification of the γ-emitting nuclides.

Calibration

Even though no appropriate international standards are adapted and no commercial measuring equipments are commercially available, the desired standard should contain Co-60 in the order of 1 to 100 Bq/g. The presence of other γ-emitting nuclides is desirable. In the Workshop we will present how to surmount this difficulty.

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DETECTING THE PRESENCE OF ABNORMAL RADIOACTIVITY IN SCRAP USING THE STATISTICAL METHOD

Prepared by G. Baillet, USINOR, France

Summary

The radiological protection criteria recommended for recycling of metals (as in the paper "Radiation protection 89") cannot be used when scrap is checked on arrival at steel plants. In the event of an incident, neither the nature of the radioelements that may be present in the scrap, nor their level of activity, nor their physical form, are known.

In practice abnormal radioactivity in scrap is detected by comparison with ambient radioactivity. However, ambient radioactivity cannot be regarded as a threshold of acceptability which applies to all products. Its level varies substantially from one place to another. All products display natural radioactivity: its level varies greatly, but in some cases it significantly supplements ambient radioactivity, though this does not mean that the products must be considered dangerous (the classic example is that of some granites and some refractory materials).

In our arrival checks on scrap-carrying vehicles (lorries and wagons) using gantries, we focus on changes in the measured ambient radioactivity, expressed in impulses per second, which arise from the presence of the vehicle between the sensor and the ambient radioactivity. For each vehicle, this shielding effect is expressed in terms of the ratio between the level measured in the presence of the vehicle and the level measured immediately before its arrival. The result is therefore a dimensionless number.

GE.98-33018 (E)

We carried out a statistical analysis of the results of lorry checks at three sites where the checking equipment is identical, but the natural ambient radioactivity levels very different. We observed that the distributions of the values of this ratio were identical for all the sites, and relate very well to a Gaussian distribution with a mean value of 0.71 and a standard deviation of 0.06. Hence these values are characteristic of the dispersion of the shielding effect of the population of "scrap-carrying lorries checked with a specific type of checking equipment" irrespective of the location of the check. It is true that there is a degree of regularity in the volume and tonnage of the lorries used for carrying scrap, but in view of the variety in the content and types of the scrap, a broader dispersion was expected.

It was then possible to identify a warning threshold on the basis of the value of this ratio by using the classical rules of statistical process control: the threshold is equal to the mean increased by five standard deviations, corresponding to a generally accepted compromise between the rate of non-detection of an actual anomaly and the false alarm rate. This is the value which has been applied to all our steel plant sites, which all have the same checking equipment.

Where different equipment is to be used, all that will be necessary will be to recalculate the mean and standard deviation during a prior observation exercise on a significant number of normal vehicles before pursuing the same line of reasoning. Indeed, this method is not restricted to scrap and may be applied wherever checking is carried out in relatively repetitive and reproducible conditions, especially where gantries are used.

This method cannot be used to measure in becquerels the exogenous activity added to the load, still less to determine the nature of the radioelement present. In a particular configuration it simply allows us to say that beyond a warning threshold defined in this manner, there is a high probability that an anomaly is present: it will then be necessary to isolate the vehicle and undertake additional investigations using specialists and special equipment so that, taking all necessary precautions, the presence of hazardous products can be confirmed and appropriate steps taken.

By defining a reference population for a given product, this experimental method also makes it unnecessary to search needlessly for absolute zero radioactivity, which might be a temptation with increasingly high-performance equipment. With this method, the use of such equipment will only enhance the sensitivity with which anomalies can be detected in terms of the reference population.

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FINANCIAL CONSEQUENCES OF ILLICIT MOVEMENTS OF METALLIC SUBSTANCES CONTAMINATED BY RADIOACTIVITY

Prepared by J.-P. Montmayeul, Commissariat à l'Energie Atomique, France

Summary

It is increasingly frequent for States to have to deal with illicit movements of metallic substances contaminated by radioactivity. Steps taken in the areas of safety and health protection necessarily have financial implications. Except in cases of special urgency, a financial evaluation is vital before such decisions are taken. Specific actions must be initiated. Aside from action by the industries directly involved in self-regulation procedures, checks must be imposed in cases of fraudulent trafficking which has no connection with fair commercial activity. Customs administrations may take specific steps to restore order to legitimate markets. International organizations have a special role to play in disseminating information and promoting international cooperation.

The paper outlines the financial impact of fraudulent trafficking, and methods of ensuring that those responsible for such activities bear the financial costs incurred. It underlines the roles that can be played by those involved in the traffic in contaminated products.

Keywords

Free trade, self-regulation, health protection, financial evaluation, re-export, customs, international cooperation.

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A METHOD OF ASSESSING THE PERFORMANCE OF PORTAL MONITORS TO ESTIMATE THE SYSTEM DETECTION CAPABILITY

(Prepared by J. R. Cox and A. Stoian, Exploranium G.S. Limited, Canada)

Summary

Over the last few years various groups have carried out tests of Portal Monitor systems most notably the SMA (Steel Manufacturers Association) test programs in the USA in 1995 and 1996 and Prague tests (run by the Czech Meterological Institute) in 1996. While various test methods have been used, the main focus of these tests was to compare the performance of various systems as a measure of performance. While this is essential in order to give an independent view of manufacturers claims, it gives no ABSOLUTE information about system performance. Also any performance limitations imposed on an actual installed system may limit system actual performance, but this would be impossible to compute from the published test data.

In 1997 Exploranium participated in a series of tests run by the Industrial Association of Brescia, Italy which attempted to assess system performance in absolute terms.

From our experience at the SMA, Prague and now the Brescia tests, Exploranium has combined the ideas of all these tests and added some new ideas to develop a new testing program that can be used to quantify any system performance.

In our opinion, an ideal test must contain the following elements:

- be easy to carry out in as short a time as practical;
- be carried out in a realistic steel plant environment for credibility;
- use a generally available scrap that permits repeatable results;
- be capable of adjustment for different truck sizes;
- be capable of adjustment for different scrap types;
- be capable of adjustment for variable backgrounds;
- use readily available test sources; and
- **give quantitative measure of detection capability**

A test has been developed and a preliminary testing program carried out. The following describes this test and give some actual test results.

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EXPERIENCES WITHIN BRITISH STEEL SINCE 1989

(Prepared by D.S. Harvey, British Steel, United Kingdom)

Summary

Monitoring of scrap delivered to British Steel began in 1989, and was extended to all sites by 1992. The quantity of scrap monitored at steelplants of British Steel, and associated companies in the United Kingdom, is in excess of 2 million tonnes/annum. The number of detections of radioactive material which triggered alarms at these sites had reached 35 September 1998, with additional detections being made at the sites of suppliers. Investigation of the causes of the alarms has revealed some consistent features. Most of the alarms are caused by natural radioactivity, few have been caused by man made radioactivity than that arising in the United Kingdom. The highest levels of radioactivity found have all been in imported scrap. Because stainless steel scrap is a particular source of risk suppliers have been asked to instal monitors to check for radioactivity. As a consequence most of the material is checked by the supplier, and by the steelplant, before it is used. The confidence that radioactive material will be detected has been substantially increased as a result.

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CRITERIA FOR THE CHOICE OF A MONITORING EQUIPMENT

(Prepared by C. U. Wieters, Thyssen Krupp Stahl, Germany)

Summary

The user of a monitor unit for the detection of radioactive nuclides in scrap has to study the conditions for his special installation. It is very important to verify the way and the transport units for the scrap. For example, scrap delivery by rail shows little variation of transport unit size, way and position of the wagon with the disadvantage of big scrap cargoes. Lorries have a large variation in size and it si very difficult to have good measuring condition (position, speed etc.). Only the size is normally a little bit smaller than by rail.

The next is to find the best location on the site for the logistic and measuring conditions. It must be assured, that all the delivered scrap passed by this point with a documentation. The optimum of detection and verification of the measurements has to be prepared.

The criteria for the sensitivity of the detectors to the ionising radiation must be found. In our company we declare a shielded radioactive Cs-137 source with a high activity, normally used in radiometric measuring units, having a dose rate at the surface near 7,5 pSv/h. This metal bloc has to be found in all position of the scrap cargo.

The other criteria for the choice of the monitor unit are the maintenance, the control of function, sensitivity and the price.

The first installation of the monitor unit has to be controlled by a specialist with tests. Also it is recommended to install a documentation of all measured cargoes. With this documentation it will be assured, that all cargoes are controlled and it is easier to clarify the reasons of malfunction like vibration, other sources (ceramic, radiometric sources, etc.).

On our site, we installed finally two types of detectors, first in 1995 for the scrap delivered by rail working in the standing position of the wagon on the wight bridge. Therefore a small CsJ detector, who is very sensitive to gamma rays, were installed with the best positions around the scrap-wagon. Second, for the delivery by road through only one entry to our plant, used big plastic detectors which assured also a good result of detection in a slow motion of the scrap.

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PLASMARC TECHNOLOGY FOR THE TREATMENT OF METALLIC RADWASTE

(Prepared by H. Weigel Mannesmann Demag, Germany and
W. Hoffelner, MGC Plasma, Switzerland)

Summary

The Plasmarc®-incineration and melting technology is suitable for processing radioactive wastes arising from the fields of medicine, industry and research, and from the operation and maintenance of nuclear power plants. Combustible wastes can be thermally decomposed and metals melted in the same facility together, and the incineration products and metals are thus turned into a form suitable for disposal in one step.

In secondary metallurgy the Plasmarec® technology can be used for melting scrap metal and recovering usable metals from metalliferous wastes, particularly composites of different metals and ceramics and metals and plastics. In the case of special wastes, it is possible to thermally decompose otherwise problematic residues in an oxygen free atmosphere at high temperatures.

Example Zwielag facility (Radwaste Facility, Switzerland)

Material construction in the incineration mode could be in 200-litre standard drums with a total weight can be up to 300 kilograms. If an average processing efficiency of 200 kilograms of mixed waste per hours is assumed.

Melting

In the melting mode for metals, the drums coming from the storage rack are place in the slowly rotating furnace using a grabbing device. Because of the low speed of rotation, the central outlet is initially blocked with a stopper. The drums, with contents, are then molten in the plasma arc. As soon as there is a melted mass, the speed of rotation of the furnace is increased until there is no material outflow when the stopper is removed. The stopped is then removed and the rotation speed of rotation is reduced once again to allow the melt to flow out, exactly as in the incineration mode.

Mixing

In the mixing mode, metallic/nonmetallic mixtures (e.g. reinforced concrete) can be processed. The meltable components are melted and the organic components are thermally decomposed. Because of differences in density, the anorganic residues float on the surface of the molten metal and can be vitrified using additives.

These different operating modes of the Plasmarc furnace allow various types of wastes to be convened into products suitable for disposal in a repository (metal, glass) without sorting and in a single step. These ensures that all the activity which can be retained is contained in the moulded products. Since no dispersible ash is formed, the entire process is virtually dust-free.

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PREVENTION OF RADIOACTIVITY IN STEEL SCRAP NECESSITY OF AN INTERNATIONAL CO-OPERATION BETWEEN INDUSTRY AND GOVERNMENT

(Prepared by G. Amedro, Eurofer)

Summary

European steel companies will not melt radioactive contaminated scrap, even if it would be allowed to do so by legislation and proposed clearance levels.

Scrap delivering companies as well as steel producing companies are well armed with scrap charge control.

Additional control during steelmaking is given by analysing a crude steel sample of each heat.

Regulations regarding health criteria are available on national and European level (RP43 and RP89) but are not usable in practice for the control of scrap.

Problems which are at present left to be solved by each site equipped with a means of detection cover principally the following areas:

- Definition and detection of abnormal radioactivity without identification of the radioactive element;
- Common definition of alarm thresholds by suppliers and customers;
- Emergency isolation measures for suspect vehicles (e.g. immediate return or quarantine), notably involving illicit material from a third country;
- Identification, isolation, handling and destination of discovered radioactive products;
- Financing of associated costs;
- Public relations;
- Preventative actions.

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It seems to be necessary to produce a benchmark for the activity level in finished steel to determine the actual level of "normal radioactivity" in the European steel pool.

A comparison of the actual state of the art in the European steel industry due to the measures already taken would be useful.

The present situation can only be efficiently improved by action in common:

- Industry has made large investments in detection equipment and in staff training. It has now practical experience;

- Additional regulatory provisions and appropriate logistical means are now awaited from government, i.e. concerning technical know-how and expertise, properly adapted equipment and in certain cases financial intervention.

European directives should not constitute an impediment and should, if necessary, be adapted to needs.

When the situation is clarified, future regular checks will be an indicator of effectiveness of scrap entry control and legal regulations to avoid the spread of activity.

Certifying the quality management systems of scrap delivering companies is a further way to assure that they are able to follow their declarations not to deliver any contaminated steel scrap.

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FULLY AUTOMATED GAMMA SPECTROMETRY GAUGE OBSERVING POSSIBLE RADIOACTIVE CONTAMINATION OF MELTING SHOP SAMPLES

(Prepared by J. Kroos, G. Westkaemper, Salzgitter AG and
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Summary

In recent years, there has been a growing concern in the steel industry about the possible radioactive contamination of steel scrap ever since the first shutdown of the radioactive contaminated plant. We can easily foresee the increased exposure of steel market and steel mill as well to the risk of radioactive contamination.

Therefore, it is required to establish the internationally harmonized and thus, internationally acceptable set of standards to regulate the limit of radioactive presence in the scrap, taking into consideration of tolerable level of natural radioactivity which could be present in the scrap.

By the theoretical reasoning, contamination of steel products can occur in the liquidifying process. Therefore, only the specific activity with respect to the mass [Bq/g] is of interest. The German Radiation Protection Regulation (Para. 4, line 1) allows the handling of materials with a specific activity of less than 100 Bq/g for man-made radio nuclides and 500 Bq/g for natural radio nuclides by law. On the other hand, the German Radiation Protection Commission e.g. recommends that all materials with a specific activity of less than 0.1 Bq/g can be allowed to release without any restrictions. Although these recommendations do not contain the phrase of legal enforcement, it mentions a guideline for steel production.

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At Salzgitter AG several monitoring systems have been installed to check the scrap transport by rail and by car. At the moment, the scrap transport by ship is reloaded onto wagons for monitoring afterwards. In the future, a detection system will be mounted onto a crane for a direct check on scrap upon the departure of ship.

Furthermore, at Salzgitter AG Central Chemical Laboratory, a fully automated gamma spectrometry gauge is installed in order to observe a possible radioactive contamination of the products. The gamma spectrometer is integrated into the automated OE spectrometry line for testing melting shop samples after performing the OE spectrometry. With this technique the specific activity of selected nuclides and dose rate will be determined. The activity observation is part of the release procedure. The corresponding measurement data are stored in a database for quality management reasons.