WORKER EXPOSURES TO INHALABLE AND TOTAL AEROSOL DURING NICKEL ALLOY PRODUCTION

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Abstract—This paper describes a study that was carried out at a North American nickel alloy production facility to compare the levels of personal exposures to inhalable and total nickel-containing aerosols. It is part of a large body of work aimed at assessing the impact of introducing new personal sampling instrumentation with performance consistent with the latest criteria proposed by the International Standards Organization (ISO), the Comite Européen Normalisation (CEN) and the American Conference of Governmental Industrial Hygienists (ACGIH). Side-by-side sampling using the 37-mm filter holder (for total aerosol) and the so-called IOM inhalable aerosol sampler was conducted for the personal exposures of workers in a range of workplaces throughout the facility. The results showed that inhalable aerosol exposure levels—for both overall aerosol and for total nickel—were consistently and significantly higher than the corresponding total aerosol levels. Weighted least-squares linear regression yielded factors ranging from about 1.3 to 2.4 for overall dust and from about 1.5 to 3.5 for nickel. Inspection of the statistical distribution of the exposures for the whole plant suggested that it was log-normal. Copyright © 1996 British Occupational Hygiene Society.

INTRODUCTION

The International Committee on Nickel Carcinogenesis in Man (ICNCM) under the chairmanship of Sir Richard Doll set out to investigate the specific forms and airborne exposure levels of nickel that might be associated with various forms of cancer. The research involved a detailed study of mortality as a function of estimated exposures over a period spanning the years from 1902 to 1987 (Doll et al., 1990). It was concluded that: (a) there was no evidence of carcinogenic risk associated with exposures to metallic nickel; and (b) respiratory cancer risks were related mainly to exposure to water soluble nickel forms at concentrations in excess of 1 mg Ni m$^{-3}$ and to exposure to less soluble forms at concentrations greater than 10 mg Ni m$^{-3}$.

Owing to uncertainties and gaps in the historical exposure data, however, it was not possible to develop quantitative dose-specific models of risk.

The current threshold limit value (TLV) recommended by the American Conference of Governmental Industrial Hygienists for soluble forms of nickel is 0.1 mg Ni m$^{-3}$ (concentration in terms of total aerosol) compared with 1 mg Ni m$^{-3}$ for insoluble forms and metallic nickel (ACGIH, 1995), but these values are under consideration for change to the lower level of 0.05 mg Ni m$^{-3}$ for all forms existing as aerosol. Although the ACGIH-TLVs are not regulatory standards per se, they are influential in the setting of formal standards in many countries. Meanwhile, in the

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U.K., nickel exposures are regulated by the Health and Safety Executive (HSE, 1993a) in terms of a maximum exposure limit of 0.1 mg Ni m$^{-3}$ (concentration in terms of the inhalable aerosol fraction) for soluble forms and 0.5 mg Ni m$^{-3}$ for insoluble forms.

Discussion during the past decade on a particle size-selective, health-based rationale for aerosol exposure assessment has led to substantial international agreement by the International Standards Organization (ISO) (1992), the Comité Européen Normalisation (CEN) (1992) and ACGIH (1994) on a unified approach. In particular, the inhalable fraction—representing particles which are capable of entering the body through the nose and/or mouth during breathing—is considered more appropriate than the total aerosol approach which is the basis of past and current practice in most countries. Where increased risks of respiratory cancer appeared in nickel refineries, both sinonasal and tracheobronchial cancers were detected. Therefore, it is considered that the inhalable fraction is the most relevant index of nickel exposure.

Nickel alloy production is an important part of the nickel using industry, producing high-quality alloys characterized by high strength, resistance to extremes of heat and mechanical stress, and long-term resistance to corrosion. The nickel content of such alloys ranges from 20 to 90%, depending on the customer needs. This paper considers exposure to airborne nickel in this industry sector, in particular the impact of adopting the inhalability criterion as the basis of occupational exposure standards (that is, switching from one based on total aerosol). It is based on experimental results that were obtained during a study carried out at a large nickel alloy production facility in North America in the summer of 1993.

METHODS

Choice of worksites and workers

The research was carried out at seven processes located in a number of buildings within the nickel alloy facility. Brief descriptions of these processes are provided in Table 1. The workers themselves were selected for participation in the study as randomly as possible with the assistance of the professional industrial hygienist permanently located at the facility.

Aerosol sampling

For the purpose of aerosol sampling, the inhalable fraction is defined quantitatively in terms of a curve relating the aspiration efficiency of the human head (or inhalability) as a function of particle aerodynamic diameter ($d_{ae}$). For ranges of conditions corresponding to workplaces this convention—as uniformly adopted by ISO, CEN and ACGIH—is described by

$$I = 0.5 \{1 + \exp(-0.06 \, d_{ae})\}$$

for $d_{ae}$ up to and including at least 100 $\mu$m. A range of existing personal samplers intended for sampling total aerosol have been tested in wind tunnels and shown not adequately to match the new inhalability criterion (Mark and Vincent, 1986; Vincent and Mark, 1990). Most notable is the fact that the closed-face 37-mm filter cassette,
Table 1. Summary of processes and the total numbers of paired samples taken at the nickel alloy production facility

<table>
<thead>
<tr>
<th>Process</th>
<th>Activity</th>
<th>No. of worker pairs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting/pouring shops</td>
<td>Melting in electric arc and gas furnaces, addition of metals to produce specific alloys</td>
<td>7</td>
</tr>
<tr>
<td>VIM/VAM</td>
<td>Vacuum induction melting and vacuum arc melting to produce alloys in highly controlled environments</td>
<td>7</td>
</tr>
<tr>
<td>Primary mill/grinding</td>
<td>Preparation of ingots by removing scale and impurities prior to hot rolling</td>
<td>7</td>
</tr>
<tr>
<td>Primary mill/hot rolling</td>
<td>Reheating and reduction of large ingots into strips and sheets</td>
<td>6</td>
</tr>
<tr>
<td>Bar/wire production</td>
<td>Reheating and reduction of ingots into specific bar and wire forms</td>
<td>7</td>
</tr>
<tr>
<td>Strip mill</td>
<td>Successive heat and chemical treatments to remove surface scale</td>
<td>8</td>
</tr>
<tr>
<td>Cold drawing</td>
<td>Cold working of rods, bars, pipes, etc., for annealing</td>
<td>5</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>47</td>
</tr>
</tbody>
</table>

described in the NIOSH *Manual of Analytical Methods* (NIOSH, 1994) and widely used throughout North America and elsewhere, clearly undersamples with respect to the inhalable fraction (Mark *et al.*, 1994). Even the seven-hole sampler, which is specifically identified as suitable for the inhalable fraction in the HSE document 'General methods for the gravimetric determination of respirable and total inhalable dust' (MDHS14) (HSE, 1993b), has been shown in such tests to consistently undersample with respect to inhalable aerosol. At present, the 2 l. min⁻¹ Institute of Occupational Medicine (IOM) inhalable aerosol sampler appears to be the only sampling instrument available specifically for collecting the inhalable fraction.* This instrument has been described extensively in the literature (Mark and Vincent, 1986; and others). Both the IOM and 37-mm samplers were used in the present study, with the objective of determining: (a) the level of exposure obtained by each method ($E_{IOM}$ and $E_{37}$, respectively); and (b) the comparison $E_{IOM}$ vs $E_{37}$ for each identified job or task classification. For each worker participating in the study, one of each type of sampler was worn side-by-side, randomized as far as possible with respect to the left or right side.

**Analysis of samples**

For all the samples taken, quantitative analysis involved determination of: (a) the mass of overall particulate matter collected; and (b) the mass of nickel collected. In addition, for about 10% of samples collected, more detailed analysis was carried out for nickel speciation, and full details of those results are described elsewhere (along with those for other nickel industry sectors) (Vincent *et al.*, 1995). The analyses for the mass of collected particulate matter was conducted on-site. For the 37-mm sampler, gravimetric assessment of the collected aerosol involved weighing the filter.

*Very recently, however, a range of personal samplers has been tested in a large wind tunnel study carried out in Britain under the auspices of the European Commission, and the results have identified more than one other device which may be suitable for collecting the inhalable fraction (see Kenny, 1995).
before and after sampling, with the difference providing the mass of sampled overall aerosol which deposited onto the filter. No account was taken of internal wall deposits. This is the most common mode of use for this sampler. For the IOM personal inhalable aerosol sampler, the whole cassette was weighed, so that internal wall losses are explicitly included. To reduce errors associated with moisture adsorption, all samples were conditioned prior to weighing by placing them in a dessicator overnight. The weighings were performed using an electronic balance (Model RC210P, Sartorius, Goettingen, Germany). After weighing, the filter in each 37-mm cassette was sent to an industrial hygiene analytical laboratory for quantitation. For the IOM sampler, the process involved an extra stage since, as already mentioned, the inhalable fraction for this instrument is taken to be the entire catch of aerosol collected inside the cassette. For this, therefore, the filter was removed and placed in a test tube. Then the inside surfaces of the cassette were wiped out carefully with an alcohol-impregnated second filter which was then added to the first filter already in the test tube. Tests have shown that one such wipe effectively removes most of the collected material. The quantitative analyses for nickel were all performed by inductively-coupled plasma-atomic emission spectroscopy (ICP-AES).

**Analysis of data**

The comparison of the data for the two samplers was performed using weighted least-squares linear regression techniques aimed at identifying differences between the working groups for given worksites and processes. In particular, we analysed the results in terms of the relation

\[ E_{\text{IOM}} = S \cdot E_{37}, \]

where \( E_{\text{IOM}} \) is the exposure to inhalable aerosol obtained using the IOM sampler and \( E_{37} \) is the exposure to total aerosol obtained using the 37-mm sampler, and where \( S \) is a coefficient which describes the observed relationship—assumed to be linear—between the two measures of exposure. Fuller details of the statistical methods—including the identification and treatment of outliers—are described elsewhere (Tsai et al., 1996b) and are not repeated here.

**RESULTS**

The results of the comparisons of worker-paired inhalable and total aerosol exposure measurements are presented in Fig. 1 in the form of plots of \( \Delta E_{\text{IOM}} \) vs \( \Delta E_{37} \) (for overall dust) and \( \text{Ni} E_{\text{IOM}} \) vs \( \text{Ni} E_{37} \) (for nickel content). Here, each worksite is identified by the symbol used. The results are plotted on log axes in order to best portray the data over the full ranges of exposure concentrations. On these axes, the fitted relationship appears as a line which is parallel to, but displaced from, the 1 : 1 line. Therefore, the fitted slope, \( S \), may be read off the graph from the magnitude of the displacement from the 1 : 1 line. The line drawn on each graph is the one obtained by weighted least-squares (WLS) regression after the removal of clear outliers meeting experimental, statistical and scientific criteria (Tsai et al., 1996b). There were just three such outliers. Table 2 summarizes the results of the regression analysis for each worksite, where the ones in parentheses were obtained with the outliers included.
Exposure during nickel alloy production

Fig. 1. Comparison between $E_{10\text{OM}}$ and $E_{17}$ for the nickel alloy production plant studied for (a) overall dust and (b) airborne nickel. The straight lines are the fitted relationships obtained by weighted least-squares linear regression. The calculated slope ($S \pm 95\%$ confidence interval), the number of sample pairs ($n$, excluding the outliers) and $R^2$ are: $S = 1.94 \pm 0.24$, $n = 45$, $R^2 = 0.86$ for overall dust (two outliers excluded); $S = 2.29 \pm 0.39$, $n = 46$, $R^2 = 0.76$ for nickel (one outlier excluded). $O$, melting/pouring; $\triangle$, bar/wire production; $O$, primary mill/rolling; $\square$, primary mill/grinding; $\bigtriangleup$, VIM/VAM; $\bigcirc$, strip mill; $\bigodot$, cold drawing. (Note: the asterisks denote data points that are considered to be outliers under the scientific and statistical conditions defined by Tsai et al., 1995.)
Table 2. Summary of results of the analyses of data for the alloy production plant studied, showing the slope (±95% confidence interval) of the relationship $E_{IOM}$ vs $E_{37}$. (Note: results in parentheses are those for analyses carried out before the outliers were removed)

<table>
<thead>
<tr>
<th>Worksite</th>
<th>Regression results ($S = E_{IOM}/E_{37}$, no intercept)</th>
<th>Overall aerosol</th>
<th>Total nickel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting/pouring</td>
<td>2.05 ± 0.67, n = 7, $R^2 = 0.91$</td>
<td>2.14 ± 0.54, n = 7, $R^2 = 0.94$</td>
<td></td>
</tr>
<tr>
<td>Bar/wire production</td>
<td>2.22 ± 1.17, n = 6, $R^2 = 0.83$</td>
<td>1.48 ± 0.17, n = 7, $R^2 = 0.99$</td>
<td></td>
</tr>
<tr>
<td>Primary mill/rolling</td>
<td>1.57 ± 1.09, n = 5, $R^2 = 0.80$</td>
<td>2.13 ± 0.59, n = 5, $R^2 = 0.96$</td>
<td></td>
</tr>
<tr>
<td>Primary mill/grinding</td>
<td>1.84 ± 0.67, n = 7, $R^2 = 0.88$</td>
<td>1.69 ± 0.53, n = 7, $R^2 = 0.91$</td>
<td></td>
</tr>
<tr>
<td>VIM/VAM</td>
<td>1.34 ± 0.28, n = 7, $R^2 = 0.96$</td>
<td>2.36 ± 0.91, n = 7, $R^2 = 0.87$</td>
<td></td>
</tr>
<tr>
<td>Strip mill</td>
<td>2.40 ± 0.73, n = 8, $R^2 = 0.73$</td>
<td>3.55 ± 1.92, n = 8, $R^2 = 0.73$</td>
<td></td>
</tr>
<tr>
<td>Cold drawing</td>
<td>2.03 ± 0.75, n = 5, $R^2 = 0.93$</td>
<td>2.49 ± 1.86, n = 5, $R^2 = 0.77$</td>
<td></td>
</tr>
<tr>
<td>Whole process</td>
<td>1.94 ± 0.24, n = 45, $R^2 = 0.86$</td>
<td>2.29 ± 0.39, n = 46, $R^2 = 0.76$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(1.87 ± 0.25, n = 47, $R^2 = 0.83$)</td>
<td>(2.56 ± 0.67, n = 47, $R^2 = 0.33$)</td>
<td></td>
</tr>
</tbody>
</table>

and are shown for the purpose of illustrating the effects of those three outliers. The results show that the exposure based on the inhalable fraction ($E_{IOM}$) consistently exceeds that based on total aerosol ($E_{37}$) by factors ranging from about 1.3 to 2.4 for overall dust and from about 1.5 to 3.5 for nickel.

**DISCUSSION**

The main feature of the inter-sampler comparison results is that the ratio $S$—equal to the ratio $E_{IOM}/E_{37}$, as given by Equation (2)—is consistently greater than unity for worker exposures in all parts of the nickel alloy facility. This is entirely consistent with what is known from wind tunnel experiments about the aspiration efficiencies of the two samplers (Mark and Vincent, 1986; Mark et al., 1994) and from aerosol sampling theory (Vincent, 1989; Tsai et al., 1996a), as well as the fact that, as mentioned earlier for the 37-mm sampler, only the filter is analysed (so that internal wall losses are not accounted for).

Estimates of the multiplicative bias of the 37-mm sampler's ability to determine the inhalable fraction, given by $S$ in Equation (2), are provided for the individual worksites in Table 2. These estimates differ from worksite to worksite, but are based on quite small numbers of measurements. The imprecision of the estimates (as reflected in the 95% confidence intervals) is of sufficient magnitude to preclude a quantitative determination of systematic differences in $S$ across worksites.

It is of interest to examine the combined data for the whole plant. When this is done, the cumulative distributions for $D_{IOM}$, $D_{37}$, $Ni_{IOM}$ and $Ni_{37}$, respectively, are as shown in Fig. 2. Inspection of these four data sets was carried out using both the Lilliefors (Leidel and Busch, 1985) and ratiometric (Waters et al., 1991) tests for log-normality, and were found to meet the conditions for log-normality. Although this finding does not strictly permit us to assume that a single $S$ value can be found for the whole plant, it is nevertheless of some interest from the pragmatic occupational hygiene viewpoint to attempt to derive such a single value for $S$. With this in mind, for the whole alloy production facility, the merged data provide (see also Table 2)
Fig. 2. Distributions of exposures to overall dust and nickel in the nickel alloy plant studied, as measured using both IOM sampler and the 37-mm sampler. The individual data points are plotted as cumulative distributions on log-probability axes, where the straightline tendency suggests log-normality.

\[ S_{\text{plant}} = 2.29 \pm 0.39 \text{(standard error)} \]  \hspace{1cm} (3)

which should be used with caution only for the particular facility studied.

**CONCLUDING REMARKS**

It has been shown that, for the nickel alloy production plant studied, the IOM inhalable aerosol sampler consistently collects more dust and nickel than the 37-mm total aerosol sampler. This follows the trend that has been found in other industries (as summarized by Vincent, 1995) and elsewhere in the nickel producing and using industries (Tsai *et al.*, 1995, 1996b). This finding is important since, whilst the 37-mm sampler is currently the most widely used sampler for occupational nickel and other
Exposure during nickel alloy production

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aerosol exposures in North America and in many other countries, there is strong
impetus towards new standards based on the inhalability criterion. Such studies are
therefore useful in providing the information needed to make a scientific assessment
of the impact of the implementation of such new standards. For nickel, discussion is
complicated by the fact that, in addition to a potential change in the criteria by
which exposure will be measured in the future, the ACGIH (1995) TLV booklet also
notes the intention to reduce the TLV for all forms of nickel to the all-time low level
of 0.05 mg Ni m\(^{-3}\). If either of these changes are implemented, the net result will be a
significantly higher incidence of exposures falling above the recommended threshold,
the impact of which could be considerable if such TLVs become influential in the
regulation process.

Nominally at least, and for aerosols in general, the British approach has
embraced the inhalability criterion since the late 1980s for all types of particulate
material which had previously been measured in terms of total aerosol. Currently,
the HSE Control of Substances Hazardous to Health (COSHH) Regulations require
compliance with the occupational exposure limits listed in the HSE Environmental
Hygiene Guidance Note EH 40 (latest version, HSE, 1993a) which in turn—for
aerosols—refers to Methods for the Determination of Hazardous Substances,
General methods for the gravimetric determination of respirable and total inhalable
dust, MDHS 14 (latest version, HSE, 1993b). In the latter, the seven-hole personal
aerosol sampler is described as 'The personal sampler for total inhalable dust...'.
This assertion is not supported by most of the evidence from wind tunnel
experiments, however. So, with the continued use of the seven-hole sampler, it is
possible that the impact of implementing the inhalability criterion for the coarse
aerosol fraction has not yet been felt. It is, therefore, strongly recommended that
side-by-side sampling involving the seven-hole sampler and an appropriate inhalable
aerosol sampler should be conducted in the near future in British workplaces.

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