Portable Instrumentation for Determination of Lead in Painted Surfaces

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Introduction

Ingestion by young children of paint chips is a major cause of lead poisoning. There has therefore been much interest in the development of portable instrumentation for measuring non-destructively and in situ the lead content of painted surfaces in dwellings. Isotope-excited X-ray fluorescence is essentially the only analysis technique that satisfies these requirements, and several instruments have been introduced for this specific task. In this paper we discuss the efficacy of currently used devices, with emphasis on accurate determination of levels close to legally permissible limits. We show that a more sophisticated device may be constructed, taking advantage of new developments in instrumentation. This improves lead detectability limits from $\sim$ 1 mg/cm^2 to $\sim$ 0.03 mg/cm^2.

Current techniques

Most current instruments employ a radionuclide source of a few millicuries activity to excite K or L X-rays of lead, and a NaI(Tl) scintillation counter for their detection. The X-ray energy spectrum recorded by the detector contains the lead characteristic X-rays superimposed on an intense background of scattered primary radiation. The latter is discriminated against by one or both of two means: first, absorbing filters on the detector face will pass preferentially the desired energies; then a 'window' can be set electronically to limit the voltage pulses accepted to a narrow band centred on the Pb X-rays. The pulses are finally counted by a scaler or ratemeter, and by pre-calibration the count can be converted to mg/cm^2 of lead.

Inevitably, background pulses in precisely the same energy band occupied by the X-rays cannot be rejected by these simple methods and are therefore counted. The statistical fluctuation in this background determines the detectability limits. KAPLAN et al (1975) have reported an intercomparison of four commercial XRF instruments (using ^{57}Co) and atomic absorption spectroscopy; they found that XRF readings greater than 3.2 mg/cm^2 indicated, with over 95% confidence, that

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AAS would yield over 1 mg/cm^2; zero XRF reading indicated an AAS value of < 1 mg/cm^2. Thus a single XRF instrument used in isolation has detectability limits in the 1 mg/cm^2 region, and readings in the 0-3.2 mg/cm^2 interval are too near the limit to be useful. KAPLAN et al. chose the 1 mg/cm^2 figure because the city of Chicago requires remedial action where this is exceeded. An even lower level of 0.03 mg/cm^2 is recommended by KING (1971).

Since the detectability limit is in the region of the recommended or enforced criteria, an improvement is desirable. But any attempt in this direction must compete with the low cost, portability and convenience of NaI(Tl) devices.

One other point is that Pb L X-rays have a half-distance of about 6 mg/cm^2 in lead. Devices using L X-rays (see eg. BARLTROP et al. 1971) cannot be sensitive to lead in older coats covered by a few more recent coats.

Use of Ge(Li) detectors

LAURER et al (1971) discussed use of a Ge(Li) detector with a ^{109}Cd source to excite Pb K X-rays. The very high energy resolution (a 70 keV peak is 50 times narrower than with NaI(Tl)) eliminates the need for filters. A single-channel analyser was set to encompass the Kβ peak, which is almost free of backscatter interference, and this fed a scaler. The results agreed with chemical analysis, and the detection limit for a 1 minute count was 0.25 mg/cm^2.

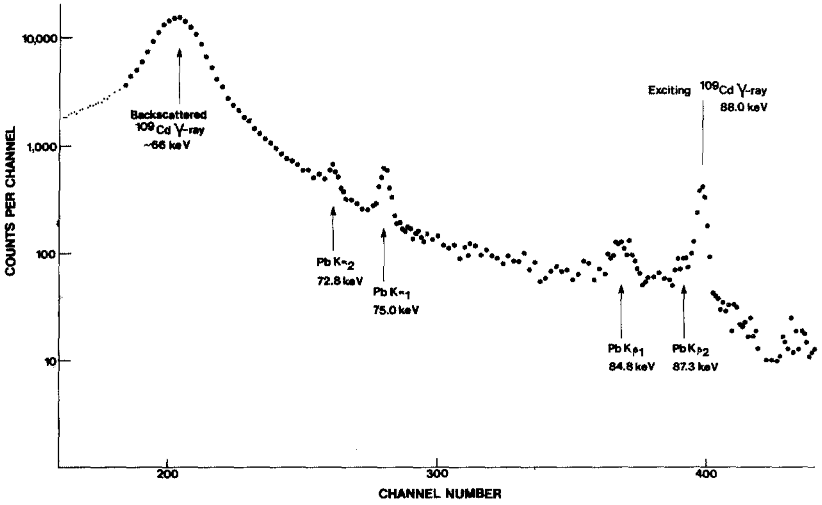

Figure 1. Ge(Li) spectrum from painted plaster panel.

Ge(Li) systems have not found widespread use since they are relatively bulky, require constant liquid nitrogen and are expensive.

Measurements

We recorded the entire pulse height spectrum excited by the ^{109}Cd 88 keV gamma rays, using an annular source mounted on a Ge(Li) detector. The spectrum is in Fig. 1.

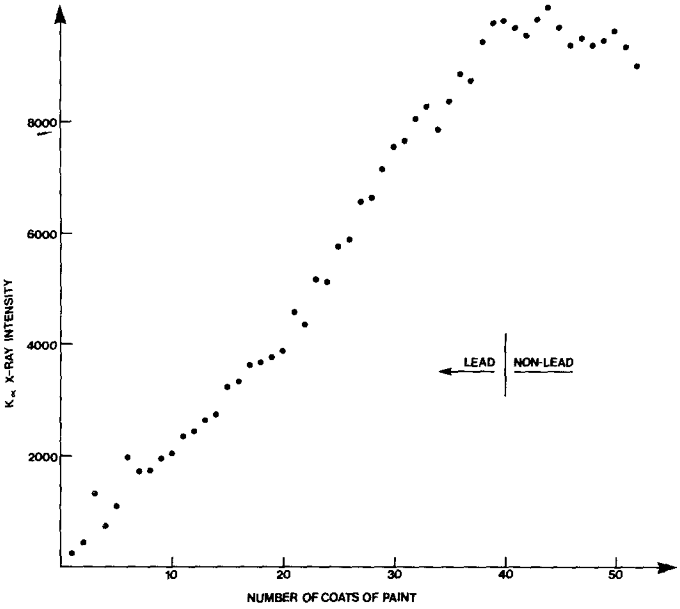

Figure 2. Dependence of fluorescent Kα X-ray intensity on number of paint layers.

The pulse-height-analysis system is described later. Background under the $K\alpha_1$ peak, taken as a measure of lead, was defined using two regions on either side of the peak. In Fig. 2 we plot the $K\alpha_1$ intensity measured during application of 40 coats of lead-bearing paint to a plaster panel. Each coat contained 0.034 mg/cm^2 of lead; thus Fig. 2 demonstrates the effectiveness of the system in the region below the 1 mg/cm^2 limit of conventional devices. Indeed a NaI(Tl) device (working on K X-rays) read zero lead after application of the fortieth coat. The linear response plus the failure of 10 non-leaded coats to influence the count, indicate absence of absorption problems. In contrast, Fig. 3 shows intensity of L X-rays of lead, excited strongly by the ^{109}Cd 22 keV X-rays. The response to lead-bearing layers is non-linear, and a few overlying clean layers hide the presence of lead effectively. It is clearly mandatory to use the lead K X-rays.

Portable pulse-height-analysis system

Our system is semi-portable. Its high sensitivity

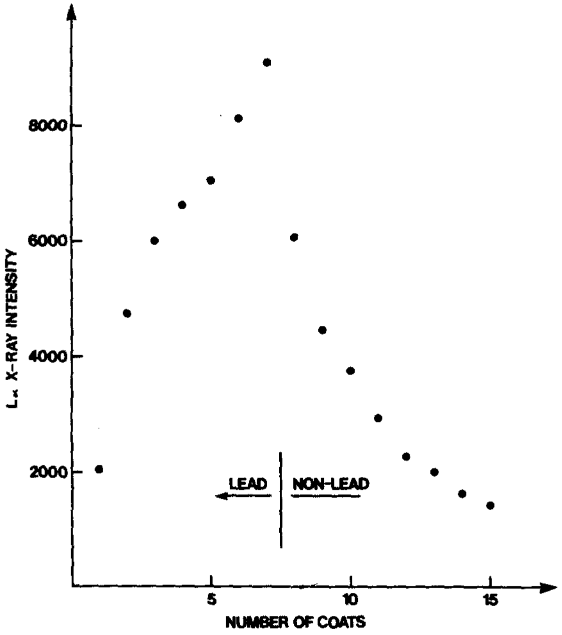

Figure 3. Dependence of fluorescent Lα X-ray intensity on number of paint layers.

makes it worthwhile to explore the feasibility of a truly portable system based on a solid-state detector (for improved detection limits) and pulse height analyser. Several recent advances in instrumentation can contribute.

First high-purity Ge detectors are available; these may be stored at room temperature and only need liquid nitrogen during measurements. Thus warmup is no longer catastrophic and a very small dewar (eg. 3 litre) may be used. Cryostats that rotate through 180° are available. Thus a light and versatile hand-held probe can be obtained to replace the NaI(Tl) probe.

Portable multi-channel analysers have appeared, incorporating detector voltage supply and pulse amplifier in the same unit, eliminating three of the four units (including bin) that accompany a lab-based detector. Output hardware is also dispensed with; a pair of markers on the CRO screen can be moved to enclose a peak or background region, whose integrated intensity then appears digitally on the screen. The discriminator/ratemeter/high voltage box for NaI(Tl) is thus replaced by an MCA in a one to one exchange.

This visual display is of great value in defining background and peak properly. In NaI(Tl) systems the operator is working 'blind'. Our background contributed from 20 to 85% of the intensity of the Pb window.

Our system comprised an Aptec GE detector with 3 litre dewar and a Canberra Omega MCA, overall weight being 30 kgm and dimensions 45x45x55 cm. These figures could be reduced by further streamlining the MCA. A GE detector of 80 mm^2 with a 20 mCi source provided a detection limit of 0.03 mg/cm^2 for a 10 minute measurement; this limit scales inversely as the square root of the time, and is defined conventionally as the lead mass affording counts equal to two background standard deviations. The cost is between $15000 and $20000, and could also be reduced by streamlining.

Conclusions

With recently available portable equipment one can effect considerable improvement in detection limits for lead in painted surfaces. Our unit is admittedly less convenient and more expensive than the currently used NaI(Tl) units, but its excellent sensitivity should provide motivation for further miniaturisation. The visual display of the X-ray energy spectrum eliminates errors due to drift and to substrate variations since the operator can make his own decisions as to definition of background. The most important point is that the device operates accurately and linearly well below the 1 mg/cm^2 level which appears to be the practical lower limit of present devices. Comparison of lead levels with legally set limits as low as 0.03 mg/cm^2 is feasible.

Acknowledgements

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