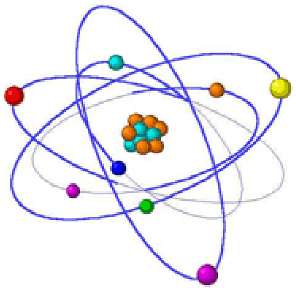
Radioisotope and Radiation Applications (FS2013)



Industrial Applications: Gauges (Week 5a, 1st part)

Pavel Frajtag

05.11. 2013

Industrial Applications and Applications in Research

- Week 1a (17.09.13): Introduction: Motivation and Physical Basics
- Week 1b (17.09.13): Radiation Sources, Detectors
- Week 1c (17.09.13): Seminar: Origin of the Nuclides & Exercises/Organisation

- Week 2a (24.09.13): Radiation Shielding
- Week 2b (24.09.13): Biological Effects of Radiation
- Week 2c (24.09.13): Exercises (W1a) & Exercises (W1b)

- Week 3a (01.10.13): Medical Diagnostics, Radiopharmaceuticals
- Week 3b (01.10.13): Radiotherapy: Fundamentals, Methods (part 1)
- Week 3c (01.10.13): Seminar: Effects of Low Dose Radiation & Exercises (W2a)

- Week 4a (15.10.13): Radiotherapy: Methods (part 2)
- Week 4b (15.10.13): Treatment Planning
- Week 4c (15.10.13): ~~Seminar: Proton Therapy at PSI~~ & Exercises (W3b)

- **Week 5a (05.11.13): Industrial applications: Gauges and Radiotracers**
- **Week 5b (05.11.13): Polymerisation, Food Irradiation, Radioisotope Batteries**
- **Week 5c (05.11.13): ~~Seminar~~ & Exercises (W4a)**

- **Week 6a (12.11.13): Gamma and Neutron Radiography**
- **Week 6b (12.11.13): Applications in Natural Sciences: Neutron Activation Analysis, Nuclear Dating**
- **Week 6c (12.11.13): ~~Seminar: NEUTRA/ICON Facilities at PSI~~ & Exercises (W5a)**

- **Week 7a (19.11.13): Radiochemistry Applications**
- **Week 7b (19.11.13): Radionuclides to Protect the Environment**
- **Week 7c (19.11.13): ~~Seminar: PROTRAC Facility at PSI~~ & Exercises (W6b)**

Motivation for Industrial/Research Applications: Unique Advantages

- Applications based on Absorption and Scattering:
  - **measure remotely** (non-destructive), online and in hostile environments
- Radiotracer Applications: **high sensitivity**
  - Tritium (T) can be determined in an atomic ratio down to T:H~ 10^{-19} !
- Gamma and Neutron Radiography:
  - provide complementary information in comparison to other techniques
- Polymerisation, Sterilisation, Radioisotope Batteries:
  - radiation leads to products of higher density and higher softening temperature
  - maintenance-free energy source with high output related to mass and volume
- Applications in Natural Sciences (Neutron Activation Analysis, Nuclear Dating), Radiochemistry Applications, Life Sciences:
  - high sensitivity, unique method

Generally many factors: uniqueness, sensitivity, time, costs, efficiency, quality...

Industrial Applications, Gauges: Outline

- Motivation
- Physical Basics: (Supplement!)
  - Fluorescence
  - Inner shell transitions
  - Electron-capture (as a pure γ-source)
- Application of γ-rays:
  - level gauges, density gauges
  - applications of γ–ray attenuation
  - X-ray fluorescence analysis
- Application of β-particles:
  - paper (film) manufacture
  - thickness of thin coatings
- Application of neutrons:
  - γ and neutron backscatter gauges
  - neutron moisture meters
  - borehole logging
- Application of protons and α:
  - PIXE, PIGME
  - Smoke Detectors

Gauges-applications are based on absorption and scattering!

Motivation: Suva-Statistics on the purchase of radioisotopes in Switzerland

Source: BAG Jahresbericht Umweltradioaktivität und Strahlendosen 2007.

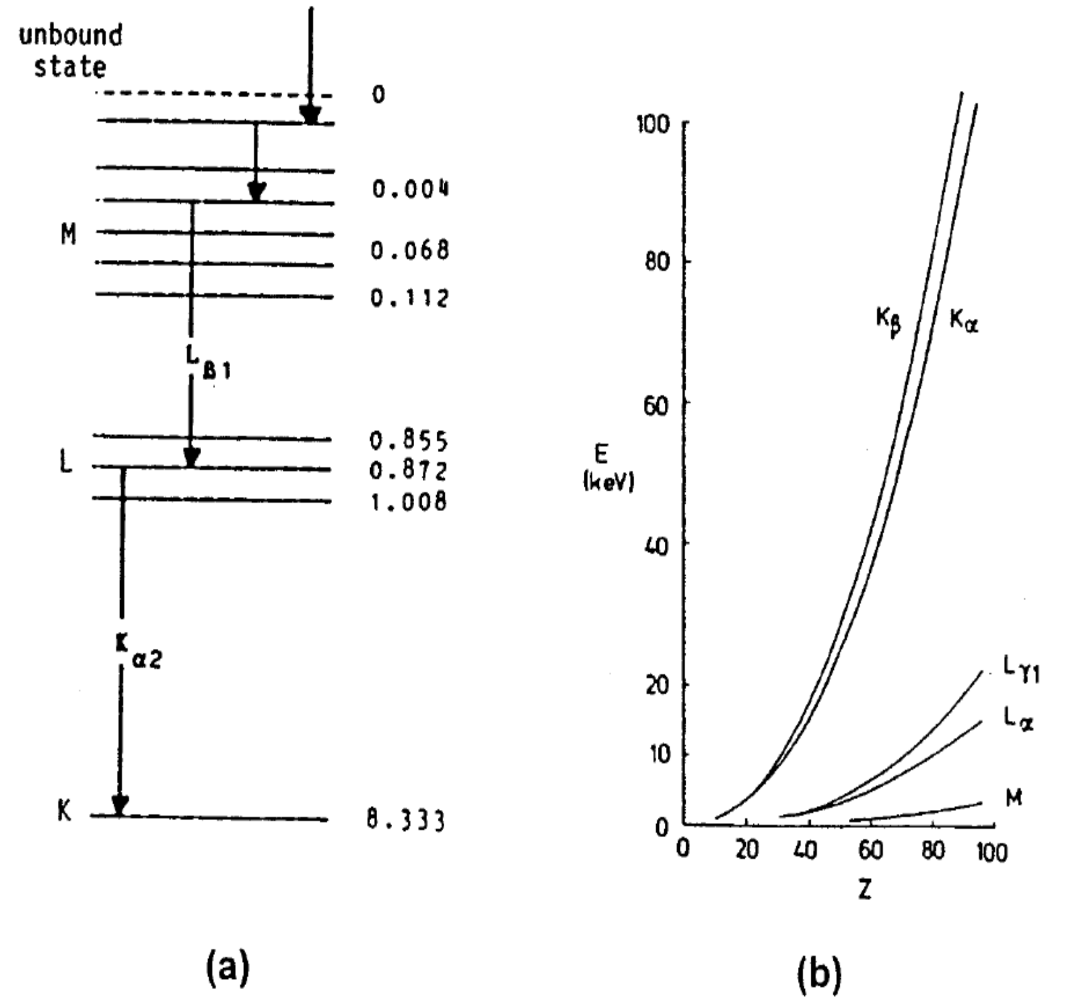
Einkauf radioaktiver Stoffe 1997 – 2007

	Isotope	2007	2006	2005	2004	2003	2002	2001	2000	1999	1998	1997	Einheit
Produktions-betriebe	^{3}H	11.51	3.85	5.237	5.493	5.506	6.216	11.4	7.3	8.9	8.1	6.5	PBq
	^{14}C	0.38	0.13	0.234	0.012	0.84	0.04	0.3	0.2	0.1	0.3	0.1	TBq
	^{147}Pm	13.16	13.19	40	0	32.618	28	19.0	26.1	21.3	25.1		TBq
	^{241}Am	0	0	0	0	0	3.7	13.0	3.3	15.2	12.0	31.3	GBq
Leuchtfarben betriebe	^{3}H	5.28	10.93	12.98	24.66	18.78	37.95	155.0	253.8	361.8	628.0	812.0	TBq
Forschungs-betriebe	^{3}H	28.4	23.6	15.2	19.4	4.3	15.4	18.1	9.7	4.9	11.7	6.8	TBq
	^{14}C	207.6	295.4	397.9	343.4	1552.8	1005	422.7	566.5	438.1	819.9	381.7	GBq
	^{32}P	7.3	9.1	11.3	20.8	6.4	28.5	30.2	45.3	36.3	56.2	76.9	GBq
	^{35}S	7.9	10.3	63.2	51.3	14.9	15.3	23.6	25.2	36.2	42.8	58.3	GBq
	^{45}Ca	0	0	0.04	0	0.06	0	0.3	1.2	1.5	1.5	1.9	GBq
	^{51}Cr	4.1	5.1	6.7	6.5	7.9	7.0	3.4	7.1	18.6	18.7	18.4	GBq
	^{125}I	1.1	1.7	3.2	23.9	27.2	18.7	3.4	22.2	32.9	41.7	53.6	GBq
Analytische Laboratorien	^{125}I	0.5	0.7	1.45	0.9	0.9	1.1	1.1	1.4	1.5	1.6	2.2	GBq
	^{3}H	0	0	0	0	0	1.1	19.7	0.0	3.0	5.3	3.9	MBq
	^{57}Co	0	0	3.08	3	5	3.4	3.7	16.1	17.2	20.4	30.4	MBq
	^{14}C	116.2	133.6	525.4	703.6	884.4	882.3	1498.0	2010.0	861.0	1246.0	443.8	MBq

Die Produktion der ^{241}Am Folien für die Ionisationsrauchmelder wurde 2003 eingestellt.

Physical Basics: Fluorescence

- Binding energies of electrons decrease from inner to outer shells: $E_{BK}>E_{BL}>E_{BM}>\dots$
- Vacancies in inner shells can be filled by e⁻ from outer shells releasing Xrays or Auger-electrons.
- Binding energy of electrons given by: $E_n \sim Z^2/n^2$;thus X–rays characteristic for element.
- QM selection rules: not all transitions are allowed.



(a) A typical series of de-excitations in atoms of nickel ($Z=28$), showing the energies of the K. L, M electronic shells. (b) The energies of K, L, M X rays as functions of the atomic number of the emitting atoms (Debertin and Helmer, 1988, Figures 1.4 and 1.7).

Inner Shell Transitions: $E_{KX}=E_{BK}-E_{BL}$ characteristic for element

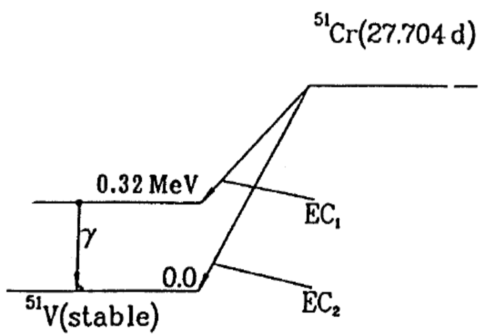
Atomic electron binding energies and fluorescent X ray energies for the atoms of selected elements.[a]

		Atomic electron binding energies in stated shells (keV)				X ray energies (keV) due to electron transfers to the K shell and their weighted average			
Z	Element[b]	K	L_2	L_3	M_3	K–L_2	K–L_3	K–M_3	$(E_{KX})_{av}$[c]
20	Ca	4.04	0.35	0.35	0.03	3.69	3.69	4.01	3.7
21	Sc	4.49	0.41	0.40	0.03	4.08	4.09	4.46	4.1
22	Ti	4.97	0.46	0.46	0.03	4.51	4.51	4.94	4.6
23	V	5.47	0.52	0.51	0.04	4.95	4.96	5.43	5.0
24	Cr	5.99	0.58	0.57	0.04	5.41	5.42	5.95	5.5
25	Mn	6.54	0.65	0.64	0.05	5.89	5.90	6.50	6.0
26	Fe	7.11	0.72	0.71	0.05	6.39	6.40	7.06	6.5
27	Co	7.71	0.79	0.78	0.06	6.92	6.93	7.65	7.0
28	Ni	8.33	0.87	0.85	0.07	7.46	7.48	8.26	7.6
29	Cu	8.98	0.95	0.93	0.07	8.03	8.05	8.91	8.2
30	Zn	9.66	1.04	1.02	0.09	8.62	8.64	9.57	8.7
35	Br	13.47	1.60	1.55	0.18	11.88	11.92	13.30	12.4
40	Zr	18.00	2.31	2.22	0.33	15.69	15.78	17.67	16.0
45	Rh	23.22	3.15	3.00	0.50	20.07	20.22	22.72	20.9
50	Sn	29.20	4.16	3.93	0.71	25.04	25.27	28.49	25.8
55	Cs	35.99	5.36	5.01	1.00	30.63	30.98	34.99	31.7
56	Ba	37.44	5.62	5.25	1.06	31.82	32.19	36.38	32.9
60	Nd	43.57	6.72	6.21	1.30	36.85	37.36	42.27	38.2
70	Yb	61.33	9.98	8.94	1.95	51.35	52.39	59.38	53.6
73	Ta	67.42	11.14	9.88	2.19	56.28	57.54	65.23	58.9
74	W	69.52	11.54	10.21	2.28	57.98	59.31	67.24	60.7
79	Au	80.72	13.73	11.92	2.74	66.99	68.80	77.99	70.5
80	Hg	83.10	14.21	12.28	2.85	68.89	70.82	80.25	72.5
83	Bi	90.53	15.71	13.42	3.18	74.82	77.11	87.35	79.4

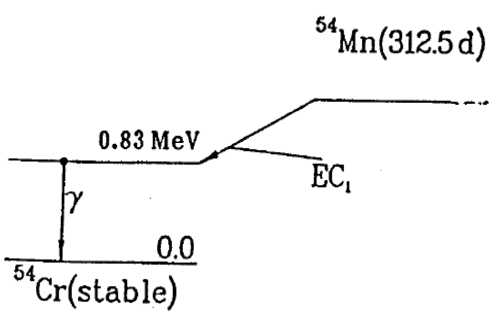
Electron Capture as a pure γ-source

- $^{A}Z + e^{-} \rightarrow {}^{A}(Z-1) + \nu_e$
- Only for a few radionuclides the decay goes directly to the ground state of the daughter. Otherwise subsequent γ-transitions.
- When only one excited state: radionuclide is a quasi-pure γ-ray emitter.

(a)



(b)

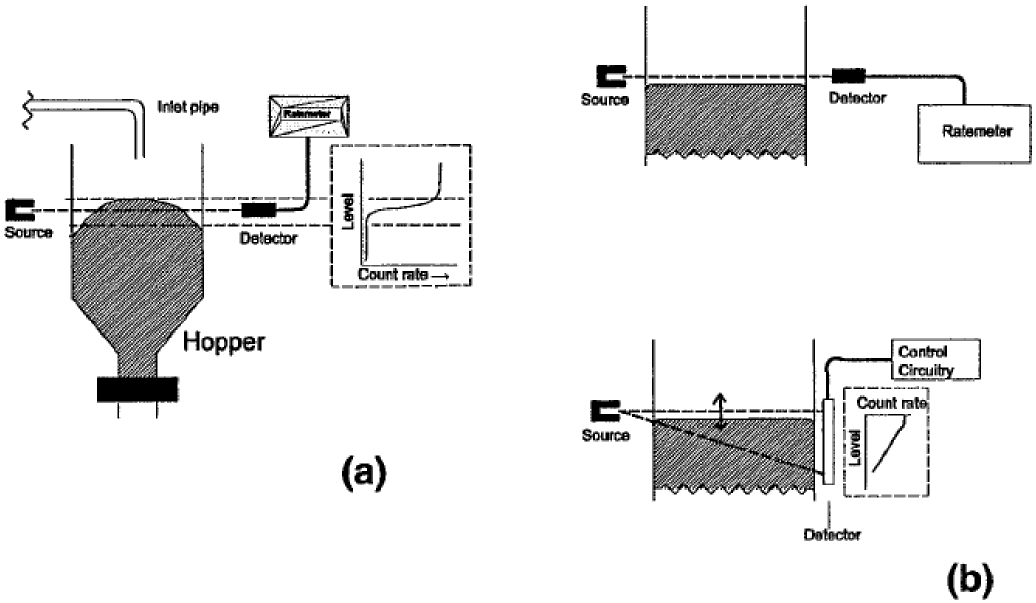


Gamma ray emitters extensively employed for applications. (Decay data from Charts of Nuclides *published since 1985.)* $E(_{KX})_{av}$ *and* E_γ *are in kiloelectronvolts;* f_{KX} *and* f_γ *are expressed as percentages.*

Nuclide	Decay mode	$T_{1/2}$	$E(_{KX})_{av}$ (f_{KX})	$E_\gamma(f_\gamma)$
^{7}Be	EC	53.28 d		478(10.6)*
^{11}C	β^+	20.38 m		511(200)*, from β^+
^{18}F	β^+	109.7 m		511(194)*, from β^+
^{22}Na	β^+	2.602 y		511(180), from β^+, 1275(100)*
^{24}Na	β^-	14.96 h		1369(100), 2754(100)*
^{46}Sc	β^-	83.3 d		889(100), 1121(100)*
^{47}Sc	β^-	3.351 d		159(68)*
^{51}Cr	EC	27.70 d		320(9.85)*
^{52}Mn	EC	5.59 d		744(90), 935(95), 1434(100)
^{54}Mn	EC	312.2 d		835(100)*
^{56}Co	EC	77.49 d		847(100), 1038(14), 1238(67), 1771(16), 2598(17) plus many others
^{57}Co	EC	271.7 d		122(86), 136(11)
^{58}Co	β^+	70.82 d		811(100), 511(30)*, from β^+
^{59}Fe	β^-	44.51 d		1099(56), 1292(44)
^{60}Co	β^-	5.27 y		1173(100), 1332(100)*
^{65}Zn	EC	243.9 d		1116(51)*
^{67}Ga	EC	3.26 d		93(38), 185(21), 300(17)
^{75}Se	EC	119.8 d		121(17), 136(59), 265(59), 280(25), 401(11)
^{82}Br	β^-	35.34 h		554(71), 619(43), 698(29), 777(84), 828(24), 1044(27)
^{85}Sr	EC	64.85 d		514(98)*
^{88}Y	EC	106.61 d		898(94), 1836(99)* (Figure 6.6(a))
^{95}Zr	EC	64.00 d		724(44), 757(55)*
^{99}Mo	β^-	65.92 h		141(91), 740(14), plus many others
^{99m}Tc	IT	6.007 h		141(89)* also in equilibrium with Mo-99, see also Figure 4.7
^{109}Cd	EC	462.6 d	23(102)	88(3.7)*
^{110m}Ag	IT	249.8 d		658(94), 678(11), 707(17), 764(22), 885(73), 937(34), 1384(24), 1505(13), plus many others
^{124}Sb	β^-	60.20 d		603(98), 723(11), 1691(48)
^{125}Sb	β^-	2.76 y	28(46)	428(29), 463(10), 6 00(18), 636(11)
^{131}I	β^-	8.021 d		364(81)
^{133}Ba	EC	59.6 d	33(118)	79/81(37), 356(62) (Figure 3.9)
^{134}Cs	β^-	2.065 y		569(15), 605(98), 796(85)
^{137m}Ba	IT	2.55 m		662(85)*, from Cs-137
^{139}Ce	EC	137.6 d	35(80)	166(79)*
^{140}Ba	β^-	12.76 d		537(24)*, (Section 1.6.3)
^{140}La	β^-	40.28 h		329(21), 487(46), 816(24), 1596(95) in equilibrium with Ba-140
^{141}Ce	β^-	32.50 d	37(16)	145(49)*

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

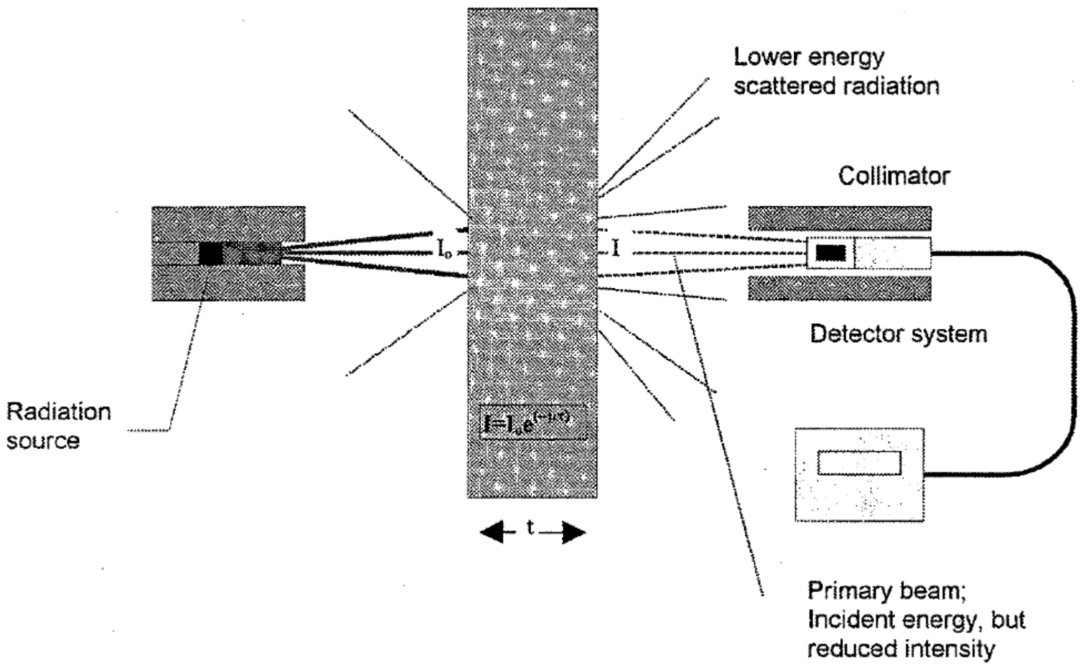
Nucleonic Level Gauges



Nucleonic level gauge comprising a collimated radioactive source, a detector and a control system. (a) Monitoring the level of material in a hopper. The response of a fixed detector to the raising and lowering of the surface of the material is shown. (b) Monitoring the level of liquid in a tank. The gamma ray beam in the lower figure has an angle of about 20° and a linear detector is used (after Charlton, 1986, Ch. 13). The response of the detector to changes in the liquid level is shown.

- ❑ Widely used for monitoring or controlling the level of material in tanks or hoppers in the refining and chemical processing industries.
- ❑ Information can be:
  - Fed online to central operation rooms.
  - Used to control pumping and valving systems.
- ❑ Advantages:
  - Measure remotely in hostile environments.
  - Device is rather robust.
- ❑ Radioactive source is chosen according to the properties:
  - energy of emitted γ-rays
  - activity of the source
  - half life

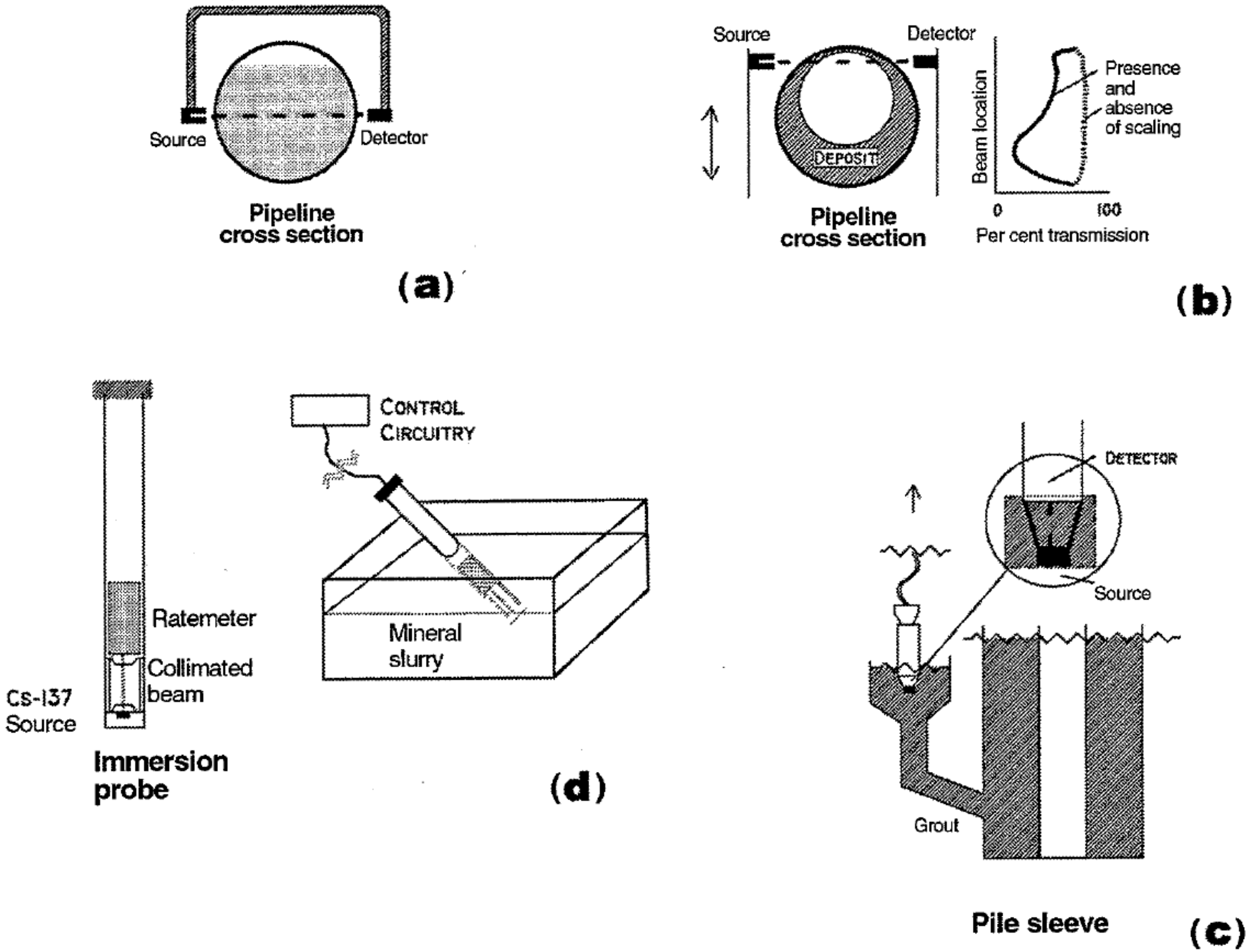
Density Gauges



The attenuation of a collimated gamma ray beam.

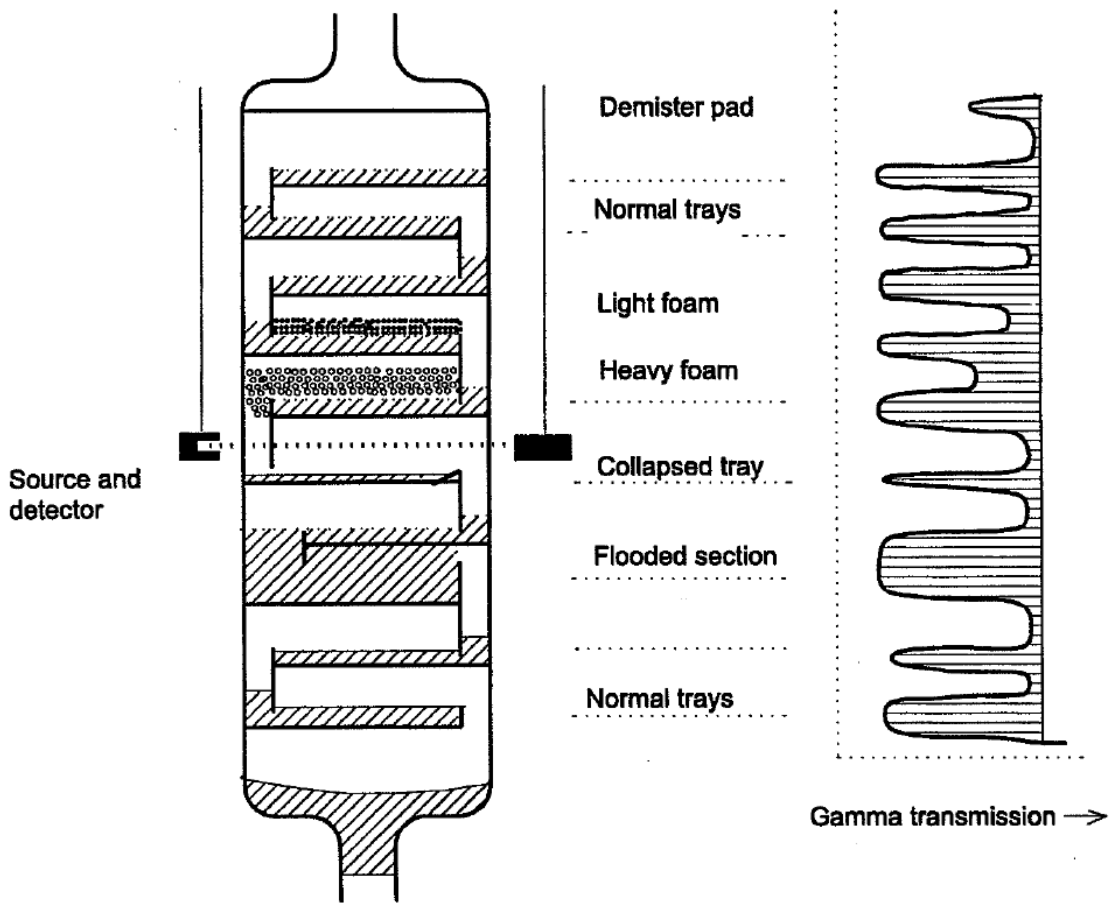
- Used to measure the density of material between source and detector.
- Basic relationship (atten. law):
  - $I(x) = I(0)\exp(-\mu_x \cdot x)$
  - $I(x) = I(0)\exp(-\mu_m \cdot \rho \cdot x)$
- Advantages:
  - measure remotely in hostile environments
  - detect differences in density as low as 0.1%
- Preferred radiation sources are:
  - ^{60}Co, ^{137}Cs, ^{241}Am
- Application areas:
  - Minerals processing industry (on stream analysis in combination with XRF).
  - Coastal engineering (sediments in rivers and estuaries).

Four Applications of γ-ray Attenuation



Four applications of gamma ray attenuation. (a) The monitoring of slurry in a pipeline. (b) Investigation of scale deposits in pipelines. (c) Monitoring of density of grout during the construction of, for example, offshore platforms (ICI Synetix Tracerco, product information). (d) The *in situ* measurement of the density of mineral slurries (Cutmore *et al.*, 1993).

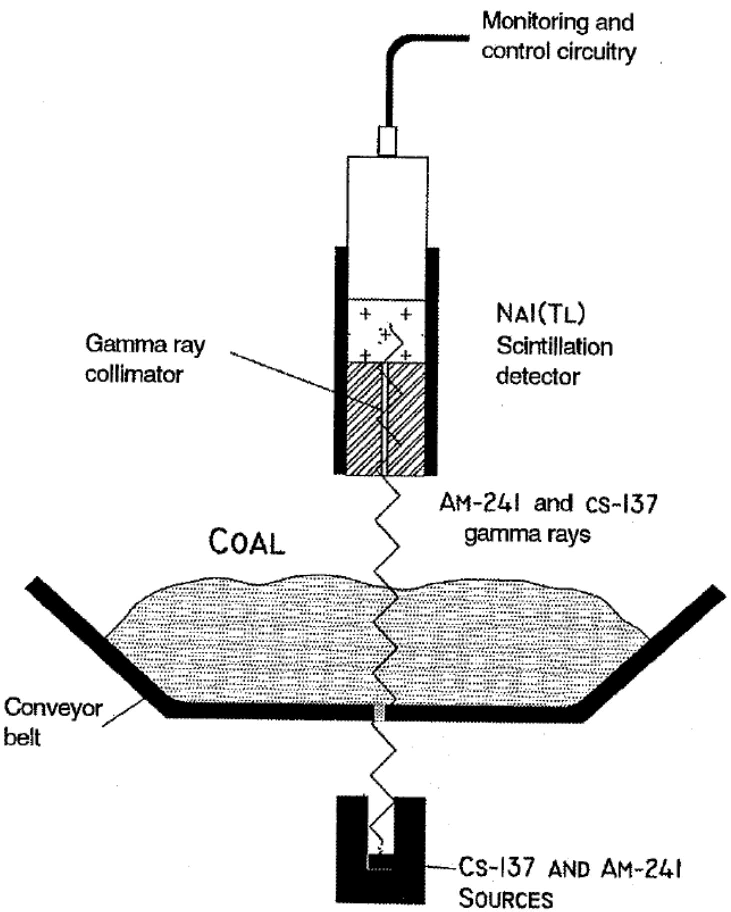
Scanning of distillation columns in the oil refining industry



Gamma transmission scanning of a distillation column illustrating identifiable features (after Charlton, 1986, Ch. 13).

- Large columns (10m high, 2-3m in diameter).
- Diagnose problems with **no interference to plant operation**.
- Source and detector are lowered in parallel on opposite sides.
- Full understanding of scan-results requires experience.

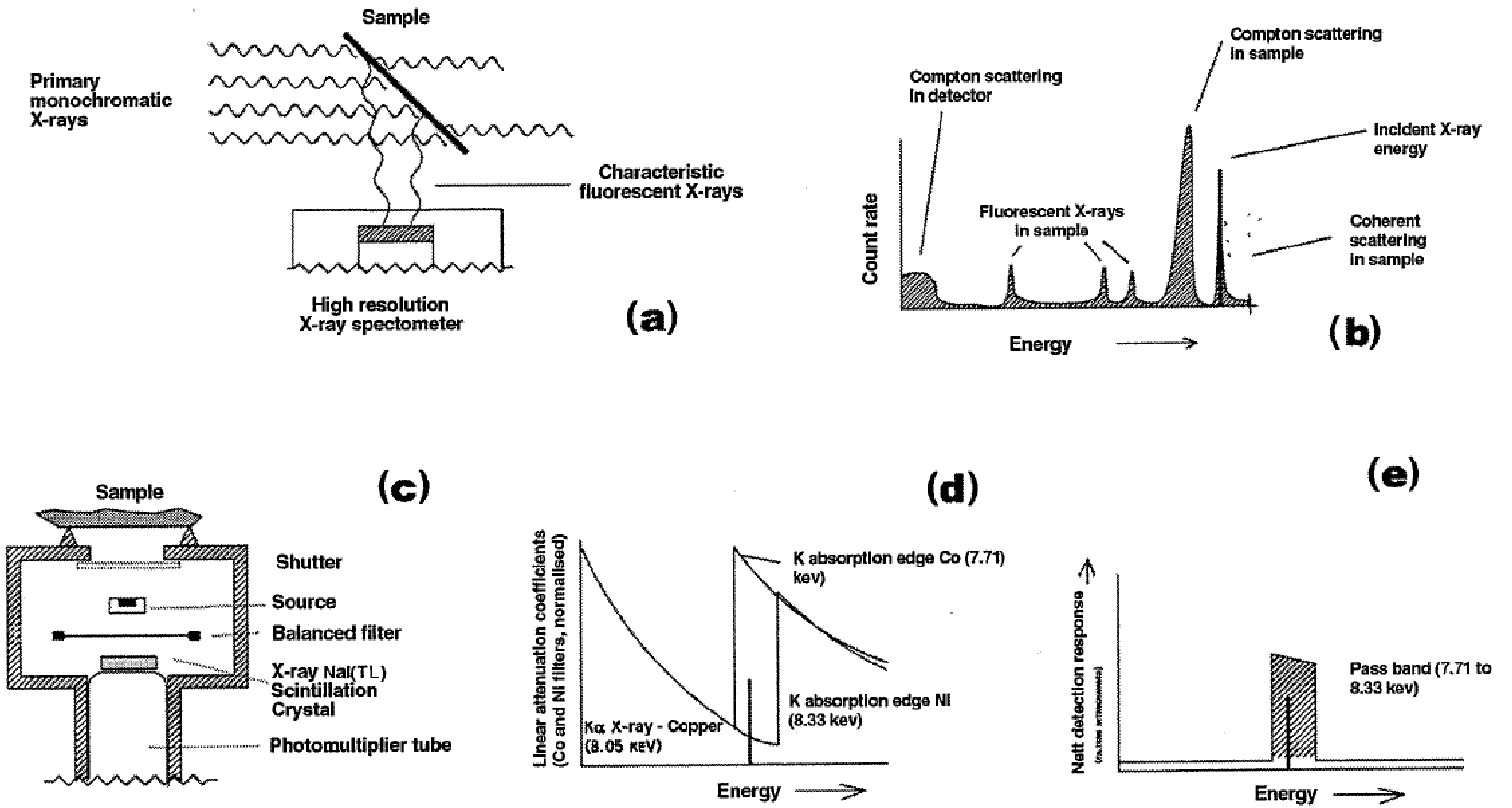
On-line Measurement of Ash in Coal



The 'on-belt' analysis of ash in coal using the dual-energy gamma transmission technique (after Cutmore *et al.*, 1993).

- Dual isotope applications:
  - ^{137}Cs (662keV): attenuation due to Compton scatter monitors the total mass.
  - ^{241}Am (59.5keV): attenuation due to photoelectric (~Z^n, n=4-5) interaction sensitive to elemental composition.
- Hybrid technology: Combination with microwave phase shift measurements to determine moisture in coal.
- Multi element analysis of coal (ash, moisture, density, key elements) by adding prompt γray neutron activation analysis.

X-ray Fluorescence Analysis (XRF, XFA)



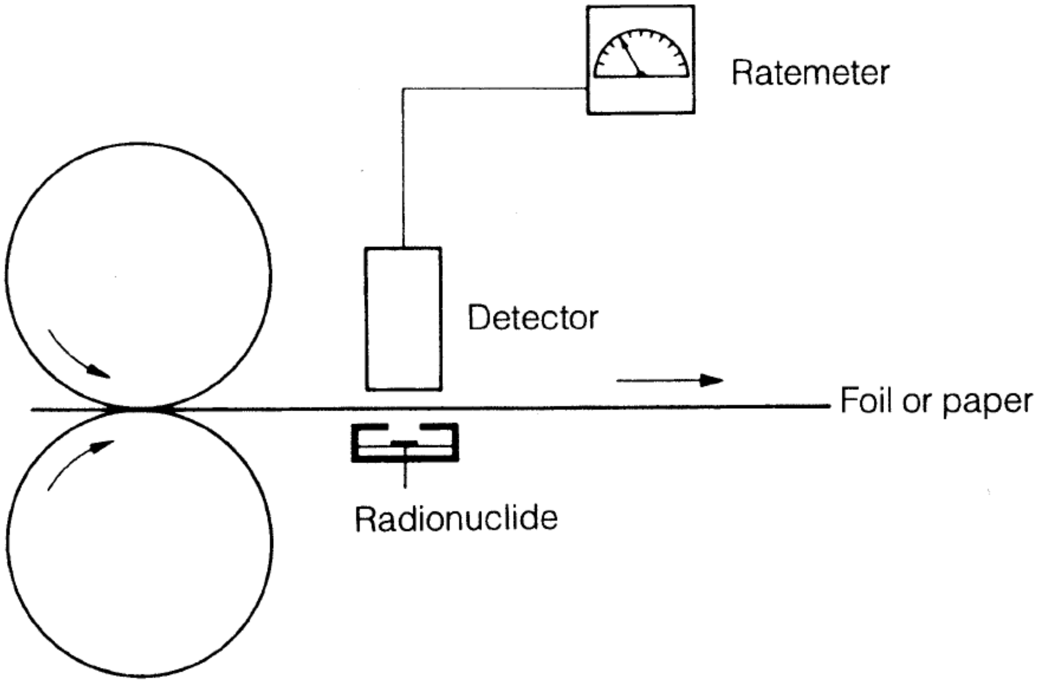
X ray fluorescence analysis. (a) Schematic representation of XRF analysis system. (b) Features of an XRF spectrum (after Jaklevic *et al.*, 1977). (c) Schematic diagram of the head of a portable XRF analyser showing the location of the source, the sample, the balanced filter and the scintillation crystal. (d) The linear attenuation coefficients of the balanced cobalt and nickel filters in the vicinity of the copper K_α X ray emission. (e) The pass band, i.e. the change in the detector response when the balanced cobalt and the nickel filters are interchanged.

Radionuclides suitable as excitation sources for XRF

Radionuclide	Half-life	Decay mode	Energy of the emission lines used [keV]
^{55}Fe	2.73 y	ε	5.9 (Mn K)
^{238}Pu	87.74 y	α	12–17 (U L)
^{109}Cd	462.6 d	ε	22.1 (Ag K)
^{125}I	59.41 d	ε	27.4 (Te K); 35.4 (γ)
^{210}Pb	22.3 y	β^-	46.5 (γ)
^{241}Am	432.2 y	α	59.6 (γ)
^{170}Tm	128.6 d	β^-	84.4 (γ)
^{153}Gd	239.47 d	ε	103.2 (γ); 97.4 (γ); 69.7 (γ)
^{57}Co	271.79 d	ε	136 (γ); 122 (γ)

- Advantages of radionuclide sources: monoenergetic radiation, possibility of measuring the K-rays of heavy elements by excitation with γ-ray emitters, no need for high-voltage installation.
- ^{109}Cd is applied most frequently.

Application of β-particles in paper (film) manufacture



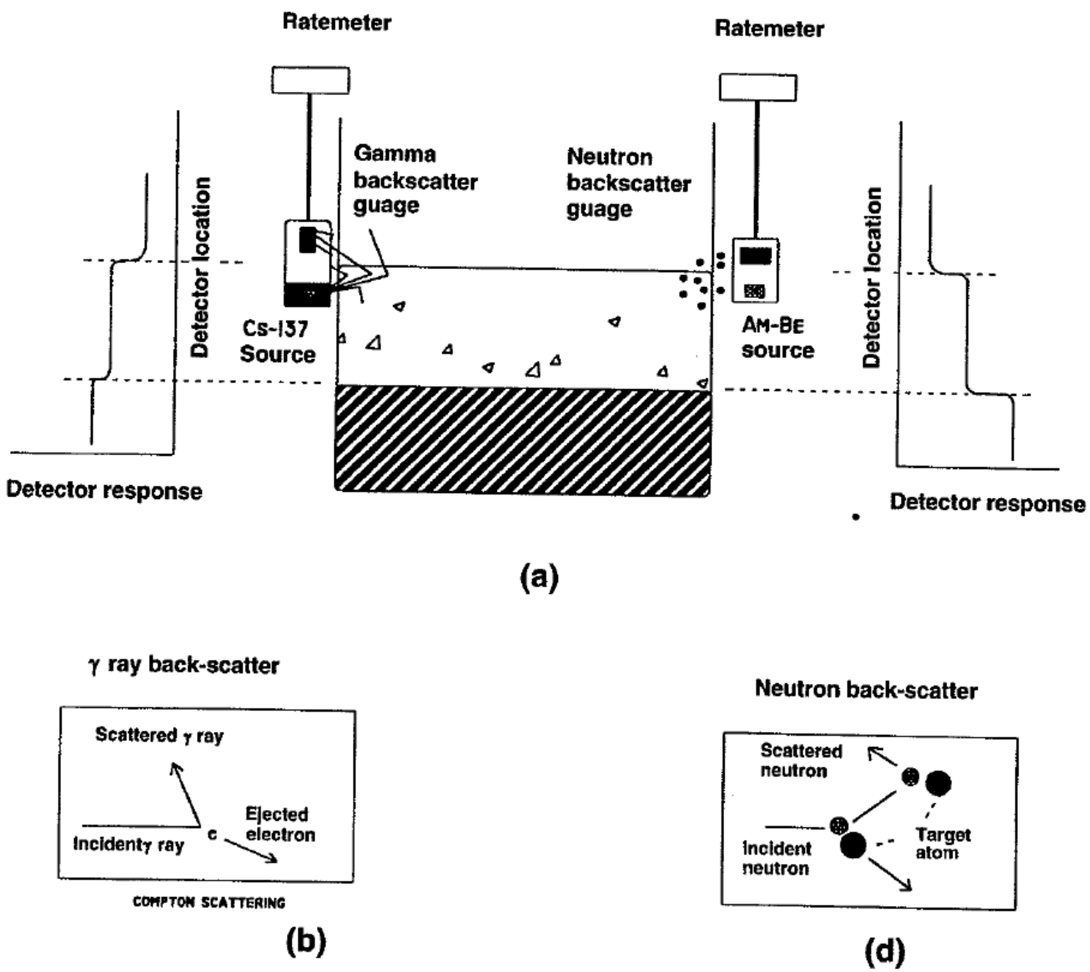
- Measurement of the thickness of films in paper, plastics and rubber industries.
- The detectors are saturation ionization chambers filled with argon.
- On-line processing of the output data from the detectors.
- Frequently used β-particle sources are:
  - ^{85}Kr ($T_{1/2}$=10.73y, E_{β}^{max}=672keV)
  - ^{147}Pm ($T_{1/2}$=2.62y, E_{β}^{max}=225keV)

Industrial Applications of β-particle Backscatter

- Beta-particle backscatter methods are well suited to measurements of the thickness of thin coatings on substrates, e.g., electroplated gold, plastic coatings on metals.
- Substrates must be thick enough for saturation backscatter.
- Radiation source is a pure or nearly pure β-emitting isotope (see table).

Nuclide	$T_{1/2}$[b]	E_β(keV) max	E_β(keV) ave[c]	Average range (mg/cm^{2}Al[d])	Per cent[e]
^{3}H[f]	12.4 y	18.6	6.7		100
^{14}C	5730 y	156	50	5.5	100
^{63}Ni	100.1 y	67			100
^{85}Kr[g]	10.73 y	672	251	80	100
^{90}Sr –	28.3 y	545	195	55	100
^{90}Y[h]	(64.1 h)	2270	940	500	100
^{106}Ru –	369 d	39	10	–	100
^{106}Rh	(30.4 s)	3550	1480	850	79
^{144}Ce –	284.3 d	316	91	–	77
^{144}Pr	(17 m)	3000	1230	710	98
^{147}Pm	2.62 y	225	62	8.0	100
^{204}Tl	3.78 y	763	245	80	98
^{210}Pb –	22.3 y	15	5	–	100
^{280}Bi	(5.01 d)	1160	390	160	100

γ and neutron backscatter gauges

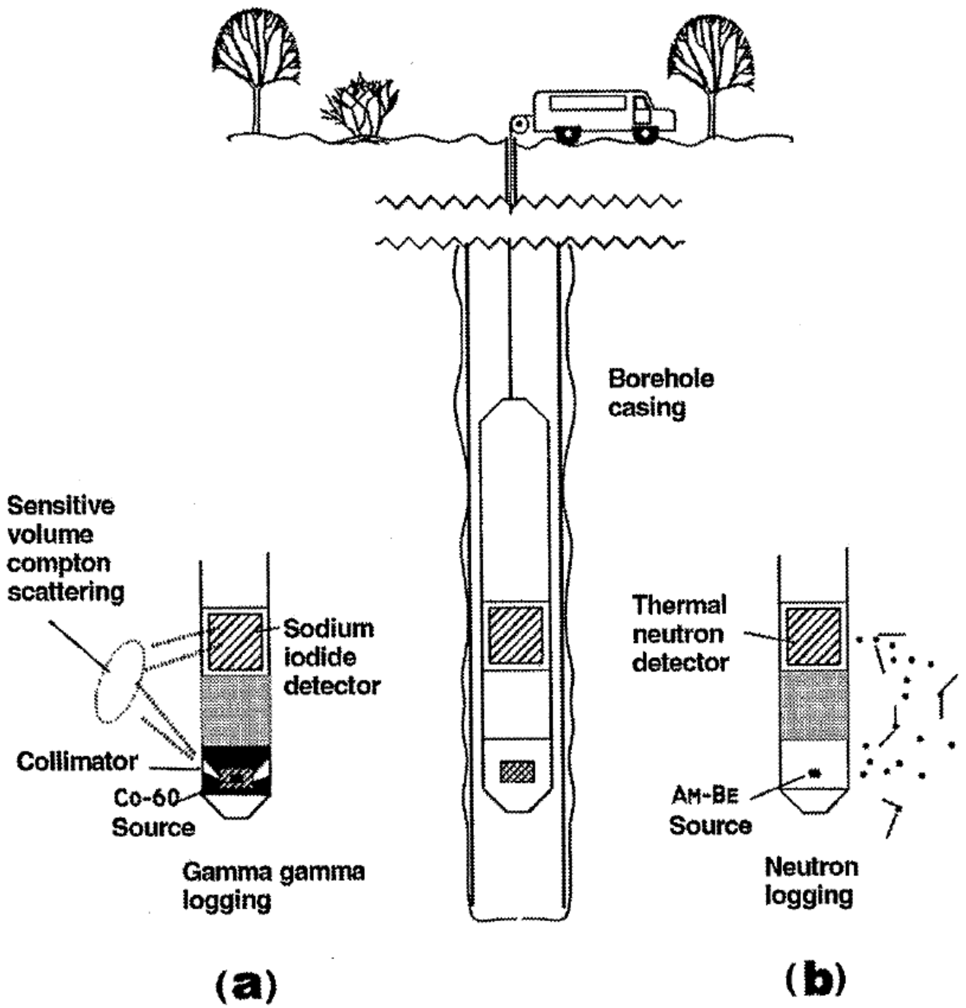


- Frequently used to monitor the levels of liquids in tanks.
- Intensity of backscattered γ-radiation depends on the bulk density of the material in the tank.
- Gauges are designed to optimize the backscatter angle.
- Fast neutron sources are used and $^{10}BF_3$ or ^{3}He proportional counters.
- With neutrons mainly the concentration of H in the liquids is measured.

Neutron Moisture Meters

- **Soil moisture meters** are specialized backscatter gauges:
  - Source of fast neutrons, detector responding only to thermalized neutrons.
  - The total hydrogen content determines principally the moderating power of the soil.
  - The sphere of influence around the borehole probe has typically a radius of 0.5m.
  - Calibration steps employing dried samples of soil may be necessary.
  - Quantitative estimates evaluate the macroscopic thermal neutron absorption cross section Σ_a: $\Sigma_a = n_x \cdot \sigma_{a,x} + n_y \cdot \sigma_{a,y} + n_z \cdot \sigma_{a,z} + \ldots$ (n_x from chemical analysis of the soil, $\sigma_{a,x}$ tabulated).
  - Problems due to traces of strong neutron absorbers in the soil.
- Moisture gauges are **widely used in agricultural research** for monitoring of soil moisture variations in the root zone, furthermore applied:
  - in civil engineering to measure the moisture levels in bulk material employed for the construction of roads and earth-filled dams,
  - in the concrete and glass industries to monitor the moisture levels in sands,
  - in the iron and steel industry to determine the moisture levels in coke and sinter mixtures.

γ-γ and neutron backscatter borehole logging



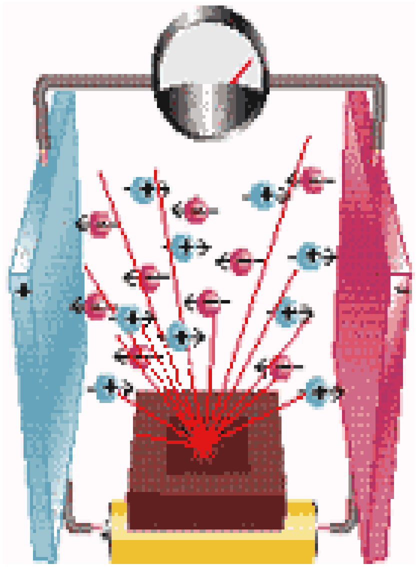
Borehole logging: (a) γ–γ and (b) neutron backscatter logging techniques.

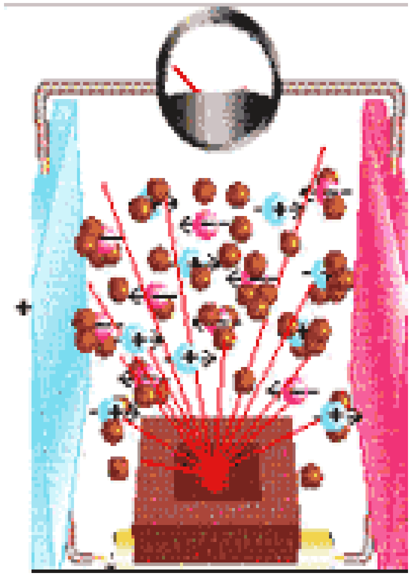
- The response of γ-γ gauges depends primarily on the energy of the incident radiation:
  - above 100-300 keV Compton dominates => measurement of bulk density
  - at lower energies photoelectric effect => sensitivity to Z_{eff} (composition of matter)
- In practice the ratio of the two corresponding intensities ($P_Z$ratio) is determined.
- Neutrons are used (mainly in the oil industry) to measure:
  - hydrocarbon volumes and viscosity
  - porosity and permeability
  - grain size and mineralogy
- Pulsed neutron sources have been applied.

Proton induced X-ray emission (PIXE) and γ-ray emission (PIGME)

- Principle:
  - Irradiation of a target material by protons (also α, γ, neutrons) leads to the excitation of the atoms and their nuclei with the resulting emission of X-rays and γ-rays.
  - The energies of the emitted radiations (spectra) are indicative of the elements present and the intensities reflect their concentrations.
- Mostly exploited for multi element analysis of natural materials.
- Frequently the proton beam comes from a Van de Graaff accelerator.
- It is possible to map the distribution of elements across a sample by focusing the beam to a spot (down to 1μm in diameter, microPIXE).
- The information from the X-rays and γ-rays can be combined with the collection of:
  - Rutherford backscattering spectrum
  - Proton transmission spectrum
- Widespread applications in geology, archeology, biology (protein analysis).

Ionization Smoke Detectors





Principle:

- α-particles from a small ^{241}Am source ionize the air between two electrodes and cause a small ionization current.
- Any smoke that enters the chamber absorbs the α-particles, which reduces the ionization and current, setting off the alarm.

Advantage/Disadvantage:

- Ionization smoke detectors are sensitive to particles over a much wider size range than those based on light scattering.
- Rejected for environmental reasons (produce radioactive waste).

Combination of ionization and photoelectric sensors possible:

- photoelectric detector senses the large, visible smoke particles (smoldering fire).
- The ion chamber detector senses the small, invisible particles (flaming fire).

Summary

INDUSTRIAL APPLICATIONS OF γ-RAYS		
Property of Radiation	**Application**	**Example**
Attenuation	Level gauges	Monitor and control the levels of liquids
	Column scanning	Diagnosis of malfunction in industrial columns
	Density gauges	Density of material in mineral processing streams
	Dual isotope	Ash in coal conveyor belts
Back-scatter	Level gauges	Levels of liquids in tanks
	γ-γ logging of boreholes	Bulk density of strata Monitoring of the water table Monitoring of oil/water interface
	On-line thickness monitoring	Monitor and control of surface coating thickness
Fluorescence	X-ray Fluorescence (XRF)	Multi element assay
INDUSTRIAL APPLICATIONS OF β-PARTICLES AND ELECTRONS		
Transmission	Thickness measurements	Thickness control in paper (film) industry
Back-scatter	Thickness of coatings	electroplated gold, plastic coatings on metals
INDUSTRIAL APPLICATIONS OF NEUTRONS		
Back-scatter	Measurement of H-content	Monitoring liquid levels in tanks Measuring moisture in soils
	Borehole logging	Porosity measurements Monitoring water/oil interface in oil wells

Literature

- G.C. Lowenthal, P.L. Airey, *"Practical Applications of Radioactivity and Nuclear Reactions"*, Cambridge University Press (2001)

  Chapter 8

- K.H. Lieser, *"Nuclear and Radiochemistry"*, WILEYVCH (2nd edition, 2001)