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Gamma-ray Attenuations in Natural Rubber

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The application of gamma-ray attenuation techniques to characterise natural rubber, rubber compounds and latex has yet to be fully exploited. The mass attenuation coefficients of natural rubber and its non-rubber constituents have been calculated for photon energies of 0.1 keV to 1 MeV. The dependence of γ-ray attenuation on photon energy and the type of photon interactions has led to the development of multi-component and multi-interaction systems for the γ-ray attenuation study in natural rubber. The multi-interaction system has been employed to determine the effective atomic numbers of natural rubber and some rubber compounds and the results are compared with the calculations. Dry rubber content (DRC) of latex at various concentrations has been measured using the multi-component system and the results show a good correlation between the attenuation method and the Standard Laboratory Method.

There are several methods for molecular characterisation of biological materials. However, the radiation method based on γ-ray absorption and scattering is still relatively new and needs further study and development. The conventional technique is based on the attenuation of transmitted X-ray or γ-ray beam arising from photon interactions with electrons in the material studied[1]. The attenuation technique has been applied over a limited range of photon energies to measure the effective atomic numbers of tissue materials using X-ray beam[2] and computerised tomography[3]. This transmission technique has also been used in the multi-phase system of oil-water-air to determine the component ratios of the pipeline flow system[4].

This paper aims to present the theoretical and experimental aspects of γ-ray attenuations which may be very significant in the characterisation of natural rubber, rubber compounds and rubber latex based on the multi-component and multi-interaction systems. Some experimental data related with the attenuation measurements of multi-component and multi-interaction systems are reviewed and discussed.

THEORETICAL

The basis of all measurements of the attenuation of γ-rays is the fact that the intensity of the radiation beam decreases as it passes through a material in such a way that if the radiation is homogeneous in a good geometry experiment, the attenuation of γ-rays is described by *Equation 1*.

$$I = I_o \exp \{ -\mu x \} \quad \ldots 1$$

where I is the intensity of radiation after a beam of initial intensity I_o has traversed the thickness x of the absorber and μ is the linear attenuation coefficient of γ-rays in the material. The attenuation of γ-rays depends on the energy of incident photons and the atomic number of the absorber. In the energy range below 1 MeV, the attenuation cross-section is mainly due to the following contributions, *i.e.* the photo-electric absorption in which photons

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interact with the innermost electrons of the atom. Compton scattering involving the interaction of photons with the outer electron, *i.e.* 'free electron', and Rayliegh coherent scattering in which photons are deflected by electrons at a small angle without losing energy[1]. In compound materials, however, the attenuation is related to the atomic composition. An attempt is made to use the dependence of γ-ray attenuation on the atomic composition and the type of interaction in the characterisation of natural rubber and rubber compounds.

Multi-interaction System

The dependence of the linear attenuation coefficient on γ-ray energy, E, the atomic number, Z, and the atomic interaction cross-section, σ, for the removal of photon by atoms in a unit volume may be written as

$$\mu_{ik}(Z,E_k) = \sum_i \sigma_{ij}(Z_i,E_k)N_i \qquad ...2$$

where N_i is the number of atom type i per unit volume and σ_{ij} is the interaction cross-section type j with atom type i. Below 1 MeV, three types of photon interactions are considered, thus *Equation 2* may be described as

$$\mu_{ik}(Z,E_k) = \sum_i \sigma_i^{P}(Z_i,E_k)N_i + \sigma_e(E_k)Z_iN_i + \delta_i^{R}(Z_i,E_k)N_i \qquad ...3$$

where the first, second and third terms of *Equation 2* are the cross-section for the photoelectric absorption, Compton incoherent scattering and Rayliegh coherent scattering respectively. The cross-section for each γ-ray interaction, *Equation 3*, may be expressed in the functional form[3], $\sigma = cE^{-m}Z^{n}$. Thus, *Equation 3* becomes

$$\mu(Z,E_k) = c_1E_k^{-m_1}\sum_i Z_i^{n_1}N_i + c_2E_k^{-m_2}\sum_i Z_i^{n_2}N_i + c_3E_k^{-m_3}\sum_i Z_i^{n_3}N_i \qquad ...4$$

For compound materials, the atomic number, Z_i, and the electron density, Z_iN_i may be replaced with the effective atomic number, ζ and the electron density, ζ.η, respectively[2]. The concept of effective atomic number for compound materials, such as natural rubber is not valid over a wide range of γ-ray energies. However, over a limited range of photon energies it is an important quantity which may be used to characterise the compound materials. The compound materials may be assumed homogeneous with the effective atomic number $\zeta = \Sigma \alpha_i Z_i^{n-1}$, where α_i = is the relative electron fraction of element i, given by $\alpha_i = N\omega_iZ_i/A_in_o$ in which n_o is the electron density given by $N\ \Sigma\ (\omega_iZ_i/A_i)$ where N is the Avogardo's number. *Equation 4* then becomes

$$\mu(\zeta,E_k) = c_1E_k^{-m_1}\sum_i \zeta_i^{n_1}\eta_i + c_2E_k^{-m_2}\sum_i \zeta_i^{n_2}\eta_i + c_3E_k^{-m_3}\sum_i \zeta_i^{n_3}\eta_i \qquad ...5$$

Equation 5 may be used to calculate the effective atomic number, ζ and electron density, ζ.η of compound materials by the attenuation measurements at three different γ-ray energies.

Multi-component System

In the multi-component system[2], γ-rays of various energies may be used to measure the attenuation coefficients of a compound material from which the amount of each component in the sample may be determined. Assuming the constituent i is uniformly distributed throughout the sample and each constituent has the thickness x_i along the beam path, *Equation 1* becomes

$$I(E_k) = I_o\ exp\ \{ -\Sigma\mu_{ik}(E_k)x_i \} \qquad ...6$$

where μ_{ik} is the linear attenuation coefficient of the constituent i at photon energy E_k. Knowing ω_i is the proportion by weight of the i-th constituent, *Equation 6* may be reduced further to

$$I(E_k) = I_o\ exp\ \{ -(\Sigma\ \omega_i\mu_{ik}(E_k))\ x \} \qquad ...7$$

where $\omega_i = x_i/x$, $\Sigma\omega_i = 1$ and $\Sigma x_i = x$. Expressing the linear attenuation coefficient of the compound materials by $\mu_{mix}(E_k) = \ln [I_o(E_k)/I(E_k)]$,

$$\mu_{mix}(E_k) = \sum_i \omega_i \mu_{ik}(E_k) \quad ...8$$

Knowing the linear attenuation coefficient $\mu_{ik}(E_k)$ at energy E_k, *Equation 8* may be solved for ω_i by making k number of linear attenuation measurements of $\mu_{mix}(E_k)$ at each γ-ray energy E_k. Note that in our case $k = i$.

Mass Attenuation Coefficient of Natural Rubber

The linear attenuation coefficient varies with the density of an absorber, even though the absorber is made up of the same materials. If the thickness x in *Equation 1* is expressed in $x.\rho$ (g/cm^2), μ (cm^{-1}) may be replaced by the mass attenuation coefficient, μ/ρ (cm^2/g), which is independent of the density or the physical state of the absorber. The mass attenuation coefficient of a mixture or compound material is made up of the sum of contributions from its constituent elements, according to the 'mixture rule'[5]:

$$(\mu/\rho)_{mix} = \sum_i \omega_i (\mu/\rho)_i \quad ...9$$

where ω_i is the proportion by weight of the *i-th* constituent with its mass attenuation coefficient of $(\mu/\rho)_i$. Knowing the elemental composition of compound materials, it is possible to derive the values of mass attenuation coefficients of some biologically important materials, however complex the molecular structure may be.

Rubber is a polymer of *cis*-1,4-polyisoprene which contributes as much as 94% of the natural rubber *(Hevea brasiliensis)*. The non-rubber constituents are protein, galactose and moisture. The composition of rubber used in this work was according to the skim rubber and ribbed smoked sheet (RSS) prepared by the Dunlop Rubber Company Ltd[6] and contained acetone extract. The elemental compositions of skim rubber and RSS were deduced from *Tables 1* and *2*. *Table 2* shows the elemental composition of galactose and glycoprotein which includes the N-acetyl-neuraminic acid, sialic acid and N-acetyl-galactosamine. Other minor molecules of non-rubber materials were treated as oxygen. The compiled data for photo-electric, Compton scattering and coherent scattering cross-sections for H, C, N and O by Veigele[7] were used to calculate the mass attenuation coefficients of skim rubber, rubber hydro-carbon, RSS, acetone extract and some non-rubber constituents as shown in *Figures 1* and *2*.

EXPERIMENTAL

Multi-interaction System in Determination of the Effective Atomic Number

Using the compilation data of Veigele[7] and optimised for carbon (Z = 6), we could analyse

TABLE 1. COMPOSITION OF SKIM RUBBER AND RIBBED SMOKED SHEET[6]

Composition	Skim rubber (%)	RSS (%)
Hydrocarbon	91.13	94.45
Protein	2.75	2.5
Acetone	5.48	2.5
Moisture	0.41	0.3
Ash	0.03	0.025
Copper	7 p.p.m.	4 p.p.m.

TABLE 2. ELEMENTAL COMPOSITION OF RUBBER AND NON-RUBBER CONSTITUENTS

Rubber/Non-rubber constituent	H	C	N	O
Hydrocarbon $(C_5H_8)_n$	11.8	88.2	—	—
N-acetylneuraminic, $C_{11}H_{19}O_9N$	7.0	50.8	5.4	36.8
N-acetylgalactosamine, $C_8H_{13}O_6N$	6.0	43.8	6.4	43.8
Sialic acid, $C_2H_4O_2$	6.7	40.0	—	53.3
Galactose, $C_6H_{12}O_6$	6.7	40.0	—	53.3
Acetone, C_3H_6O	10.4	62.0	—	27.6

the photo-electric and coherent scattering cross-sections for the energy region and the atomic number of interest and obtained the values[8] for c_1, c_3, m_1, m_3, n_1 and n_3 as 28.44, 3.18, 3.30, 1.78, 4.47 and 2.50 respectively. Thus, *Equation 5* becomes:

$$\mu(\zeta,E) = 28.44\, E^{-3.30}\zeta^{4.47}\,\eta + \sigma_e(E)\zeta\eta + 3.18\, E^{-1.78}\zeta^{2.50}\,\eta \qquad ...10$$

where $\sigma_e(E)$ is Klein-Nishina cross-section for the Compton scattering. To obtain the effective atomic number, ζ, and electron density, $\zeta.\eta$, three measurements of $\mu(\zeta,E)$ must be made at three different energies and the functional ratio of $\mu(\zeta,E_1)/\mu(\zeta,E_2)$ or $\mu(\zeta,E_2)/\mu(\zeta,E_3)$ solved.

Seven types of rubber samples supplied by the Rubber Research Institute of Malaysia (RRIM) were used in our experiment. Except for the natural rubber sample 25%, 50% and 75% carbon black were added to other samples of sulphur and peroxide vulcanised rubbers. Only one type of carbon black was used and the particle size of the carbon composite was not considered significant since the interaction of photons with matter takes place at the atomic level. All samples were prepared in sheet form of thickness 2.4 mm. The linear attenuation coefficients of the samples were determined by the transmission attenuation method at photon energies 17.7 keV, 26.4 keV and 59.5 keV from ^{241}Am source using a lithium drifted silicon Si(Li) solid state detector. A multi-channel analyser (MCA) was used to display the intensity of gamma spectra used in our experiment[8].

Multi-component System in Dry Rubber Content Determination

To test the model of multi-component system for natural rubber, we considered *Hevea* latex as consisting of a two-component system of water suspension, that is water, α_1 and solid material of rubber, α_2. The composition of the solid content could be calibrated for DRC in latex[9]. If the attenuation coefficients μ_{21} and μ_{22} of solid natural rubber in latex could be measured at two photon energies E_1 and E_2, α_1 and α_2 could be solved using *Equation 8*. Thus

$$\mu_{mix}(E_1) = \alpha_1\mu_{11}(E_1) + \alpha_2\mu_{21}(E_1) \qquad ...11$$

$$\mu_{mix}(E_2) = \alpha_1\mu_{12}(E_2) + \alpha_2\mu_{22}(E_2) \qquad ...12$$

where μ_{11} and μ_{12} are the linear attenuation coefficients of water at the two γ-ray energies E_1 and E_2 respectively.

To determine DRC using the attenuation method, bulk samples of latex were diluted with distilled water to obtain samples with low DRC ranging from 4% to 35%. The linear attenuation coefficients of latex at various concentrations were determined using the setup similar to that of the previous experiment but at photon energies of 17.7 keV and 26.4 keV.

The values of μ_{11}, μ_{12}, μ_{21} and μ_{22} were then deduced from two straight lines of the attenuation coefficient *versus* the DRC using the least square curve fitting method The values of μ_{11}, μ_{12}, μ_{21} and μ_{22} obtained in our experiment were 0 998 cm^{-1}, 0 418 cm^{-1}, 0 464 cm^{-1} and 0 163 cm^{-1} respectively Using *Equations 11* and *12*, the percentage by weight of solid material (α_2) or an estimated DRC in latex of various concentrations have been determined by the measurements of μ_{mix} at 17 7 keV and 26 4 keV and compared with that of the standard method For each latex concentration, DRC measurements by the Standard Laboratory Method were carried out based on Malaysian Standard[10] MS 3 35 1875

RESULTS AND DISCUSSION

Calculated values of the mass attenuation coefficients of skim rubber, RSS rubber, rubber hydrocarbon and acetone extract in the energy range of 0 1 keV to 1 MeV are shown in *Figure 1* *Figure 2* shows the mass attenuation coefficients of some non-rubber constituents of the natural rubber The results show that rubber hydrocarbon contributes the largest portion of the attenuation in natural rubber, while carbon and oxygen play a significant role in the γ-ray attenuation of non-rubber hydrocarbon Above the elemental absorption edge energies, two attenuation events were clearly shown Firstly, in the energy regime of about 1 keV to 100 keV, the interaction due to photo-electric energy decreases with the increase of γ-ray energy according to $\sigma \sim E^{3/2}$ Secondly, the Compton scattering dominates the attenuation of rubber materials at photon energies of 50 keV to 1 MeV Although experimental information on the mass attenuation coefficient of rubber materials has not been reported, the mixture rule is believed to hold true to a high level of accuracy except in the neighbourhood of an absorption edge of one constituent element The results indicated that above the elemental absorption edges, the energy regime of 1 keV to 100 keV would be the most appropriate photon energy region to be used in γ-ray attenuation study of biological materials Thus, the selection of energy of photons used in the attenuation measurements of natural rubber, rubber compounds and latex reported here is considered justified

Except for the peroxide rubbers, the effective atomic numbers of natural rubber and sulphur vulcanised rubbers have been reported earlier[8] *Figure 3* shows the correlation between the calculated and measured ζ values of natural rubber and the sulphur and peroxide vulcanised rubbers used in this study The calculated values are based on the *cis*-1,4-polyisoprene structure which contributes up to 95% of the total composition of natural rubber Since the major composition of natural rubber is carbon and hydrogen, its effective atomic number is less than the atomic number of carbon, $Z = 6$, or the effective atomic number of fats, $\zeta = 5\ 95$, which consists of hydrogen, carbon and oxygen[11] It can be seen that for the sulphur vulcanised rubber, the value of ζ is higher at the lower percentage of carbon black added and decreases as more carbon black is added to the vulcanised rubber This suggests that sulphur contributes to an increase in the ζ value and as more carbon is added into the rubber compound, the value of ζ decreases towards the atomic number of carbon In the case of the peroxide vulcanised rubber, the value of ζ increase towards the atomic number of carbon as the amount of carbon added increase It can be concluded that the effective atomic number of compound materials depends on the amount and atomic number of elemental compositions present as long as the experiment is carried out over a limited range of photon energies Knowing the effective atomic number of a compound material its molecular composition cannot be predicted directly but it can be

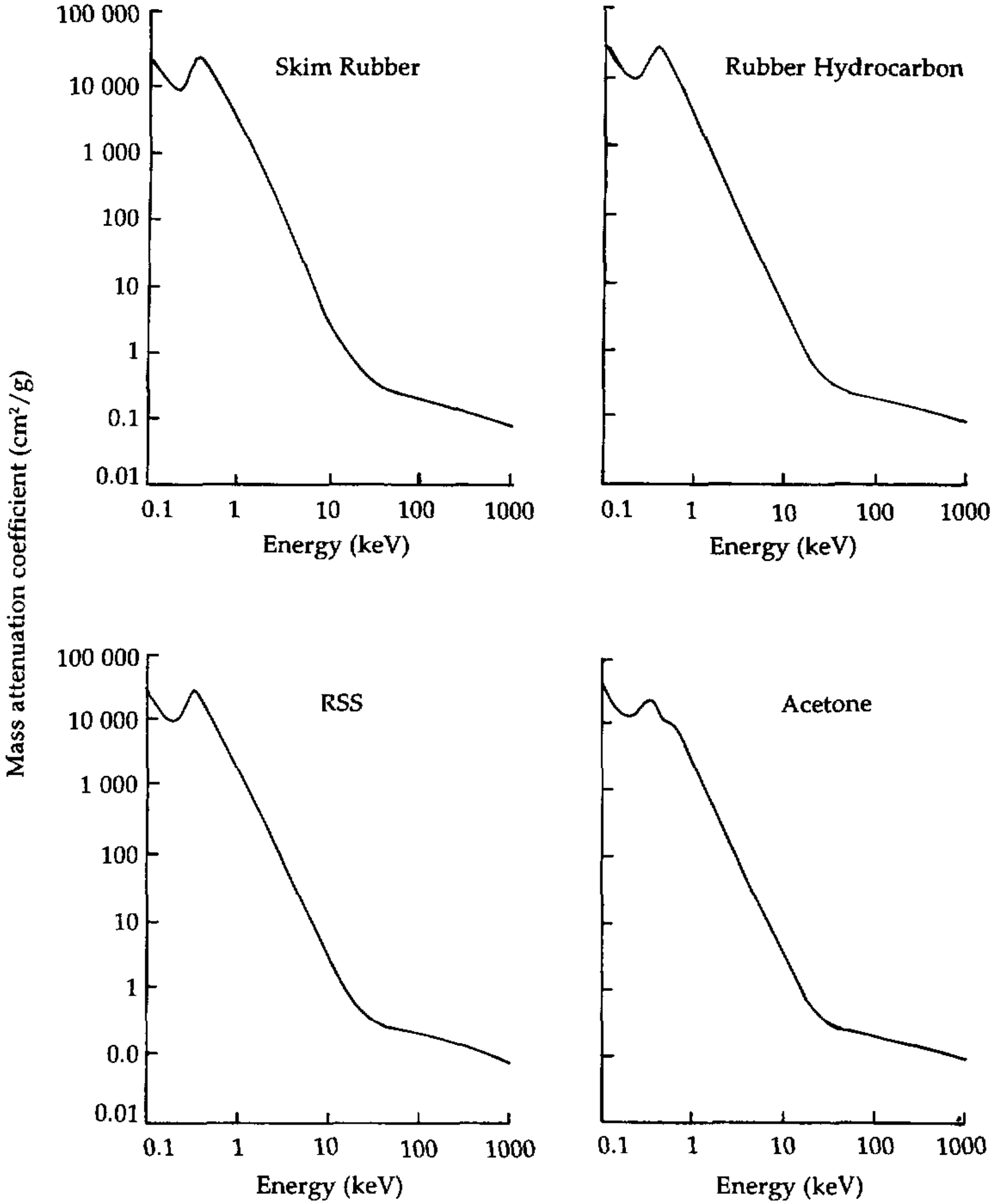

Figure 1. Mass attenuation coefficients of skim rubber, RSS, rubber hydrocarbon and acetone extract.

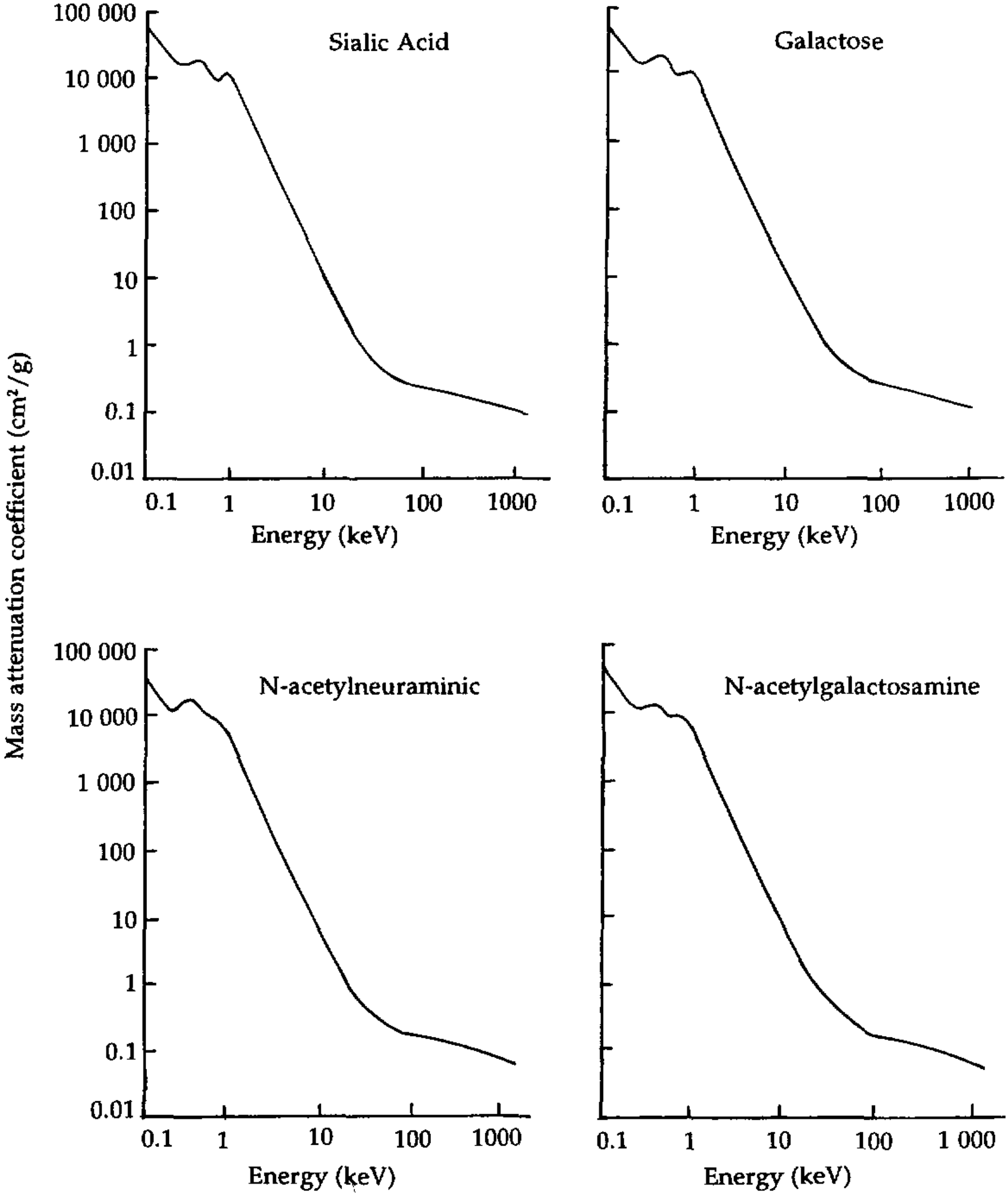

Figure 2. Mass attentuation coefficients of some non-rubber constituents of natural rubber.

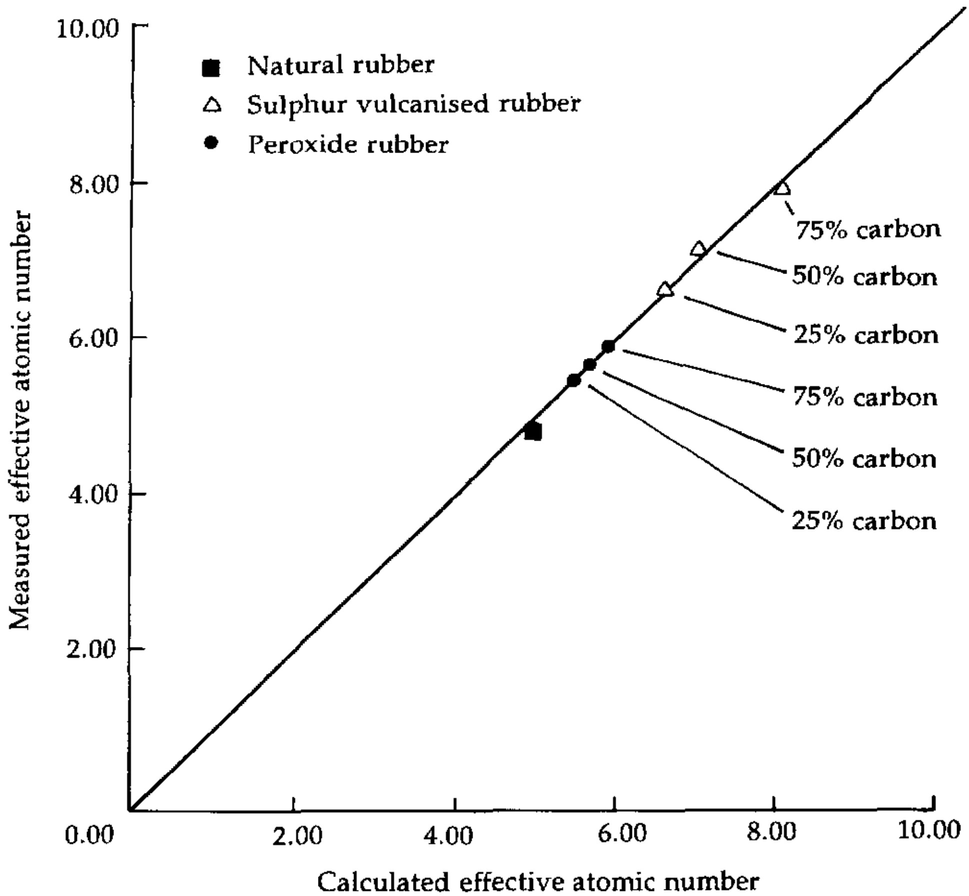

Figure 3. Correlation between calculated and measured effective atomic number of natural rubber, and sulphur and peroxide vulcanised rubbers.

used to determine the right mixture of compounds in some rubber products.

Figure 4 shows the typical relationship of DRC measured by the γ-ray attenuation method with that measured by the Standard Laboratory Method. The correlation co- efficient between both methods was 0.998 which is very good. Therefore, the method proposed in this work would give the possibility of a direct and non-destructive method for determination of DRC.

Nevertheless, deliberately added impurities in latex samples could cause serious problem, especially if the effective atomic number of the adulterants is about equal to that of the natural rubber. To overcome this problem, the attenuation measurement should be carried out at various photon energies depending on the number of impurities added. Using the multi-component equation, *Equation 8,* the attenuation coefficient data may be computed to give the amount of each component of materials present in latex.

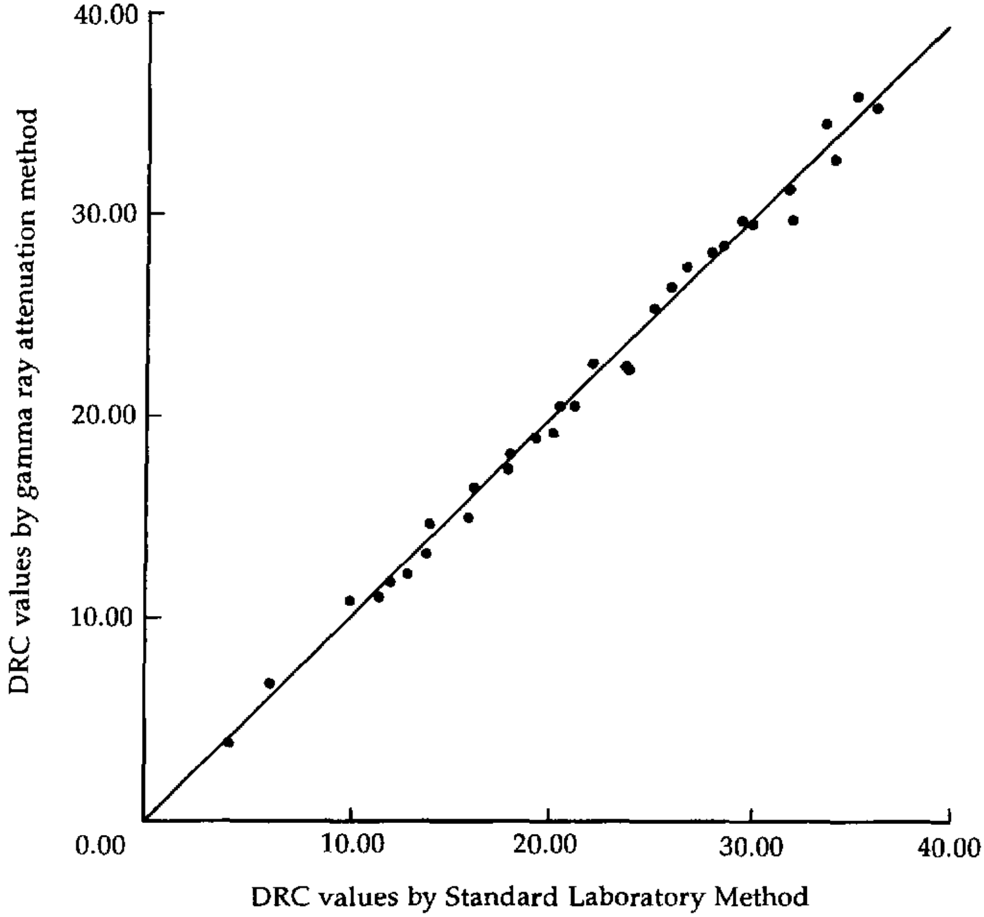

Figure 4. Typical relationship between dry rubber content measured by the γ-ray attentuation method and standard laboratory method.

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