

Natural radioactivity and associated radiation hazards of some commonly used building materials in southwest Nigeria

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ABSTRACT The activity concentrations of primordial radionuclides (^{40}K , ^{226}Ra and ^{232}Th) in five different types of commonly used building materials in southwest Nigeria were measured using a low-background NaI(Tl) detector coupled to an IBM-compatible multichannel analyzer. The highest mean concentration of ^{40}K , which was $887.5 \pm 101.3 \text{ Bq.kg}^{-1}$, was obtained in gravel. Also, the highest mean concentrations of ^{226}Ra and ^{232}Th were obtained, respectively, in sand and brick samples. The lowest mean concentrations of ^{40}K , ^{226}Ra and ^{232}Th , respectively, were obtained in brick and cement sample. The annual gonadal effective dose (AGED) and other radiological parameters were estimated from the activity concentrations of the radionuclides. The highest mean values of the AGED and radium equivalent activity concentration obtained were $359.1 \mu\text{Sv.y}^{-1}$ and 110.9 Bq.kg^{-1} , respectively. All these radiological parameters were below the maximum permissible levels recommended for safe use of materials in building construction. It can therefore be concluded that these materials do not pose any radiological hazard to the dwellers of buildings constructed with them.

Keywords: building materials / primordial radionuclides / radiation hazards / NaI(Tl) detector

1. Introduction

All building materials derived from soil and rocks contain certain concentrations of natural radioactive substances. Naturally occurring radioactive materials are known to be the largest contributor to human radiation exposure (UNSCEAR, 2011). These include the radioactive substances in the uranium and thorium decay series (^{238}U and ^{232}Th) with their respective decay progenies, and the radioactive isotope of potassium (^{40}K). In the uranium decay series, the decay chain segment starting from radium (^{226}Ra) is radiologically most important and, therefore, reference is often made to ^{226}Ra instead of ^{238}U . In addition to naturally occurring radioactive materials, some industrial by-products also contain artificial radionuclides such as ^{137}Cs , which are spread into the environment due to the fallout from nuclear weapon tests and industrial nuclear accidents, *e.g.* the Chernobyl

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accident of 1986 (STUK, 2010). The contribution of artificial radionuclides to human exposure in the environment is known to be far smaller in comparison with the contribution from natural radionuclides (UNSCEAR, 2011).

Natural radionuclides in building materials can cause both external exposure due to direct gamma radiation and internal radiation exposure from radon gas (Mehdizadeh *et al.*, 2011). Radon is an alpha particle emitter with energy 5.48 MeV, a half-life of 3.82 days and a daughter product of ^{226}Ra . It is chemically inert and can move through the earth and structural materials to reach the outdoor atmosphere and indoor spaces (Nazaroff and Nero, 1988; Kobeissi *et al.*, 2008). Its accumulation indoors in rooms is believed to form more than 50% of the dose equivalent received through inhalation by the general public from all sources of radiation exposure (Kobeissi *et al.*, 2008). Exposure to high concentrations of radon gas and its progeny for a long period of time can cause pathological effects and the occurrence of lung cancer (BEIR, 1999). The radiation which humans are exposed to may increase appreciably if they live in houses constructed using materials whose radiation doses are above the normal background radiation level of the area (Cliff *et al.*, 1985; Otoo *et al.*, 2011).

Knowledge of the distribution pattern of natural radionuclides in building materials is very important due to the fact that building materials constitute the largest source of population exposure because humans spend most of their time indoors. The results of radioactivity measurements in building materials help in setting up radiation protection guidelines and aid in decision-making processes. They also assist in the implementation of precautionary measures whenever the dose level is found to be above recommended limits. However, in Nigeria, there is virtually no regulation currently controlling the use of materials in relation to their radioactivity levels. During the last three decades, there has been increased interest in the study of the radioactivity levels of diverse building materials. Several national and regional surveys have been conducted to establish the radioactivity concentrations and radiological impact of various building materials such as sand, bricks, concrete, granite, cement, ceramic tiles, clay, etc. (Ingersoll, 1983; Berekta and Matthew, 1985; Malanca *et al.*, 1993; Hewamanna *et al.*, 2001; Khan and Khan, 2001; Stoulos *et al.*, 2003; Xinwei, 2004; Papastefanou *et al.*, 2005; Xinwei, 2005; Al-Jundi *et al.*, 2006; Tufail *et al.*, 2007; Bridigo Flores *et al.*, 2008; Turhan *et al.*, 2008; Righi *et al.*, 2009; Hassan *et al.*, 2010; Stojanovska *et al.*, 2010; Otoo *et al.*, 2011; etc.). In Nigeria, a few studies have focused on the radioactivity levels of building materials (Farai and Ademola, 2005; Farai and Ejeh, 2006; Farai and Isinkaye, 2009). This recent work is therefore aimed at evaluating the activity concentration of five commonly used building materials; cement, gravel, bricks, ceramic tiles and sand, in order to estimate the radiological problems associated with their use in constructing houses in southwest Nigeria.

2. Materials and methods

Samples of different commonly used building materials were collected from five cities (Abeokuta, Ado Ekiti, Akure, Lagos and Osogbo) in southwest Nigeria. The samples consist of sand, gravel, cement (which includes two popular brands, Dangote and Portland cement), ceramic tiles and bricks. The ceramic tiles used in southwest Nigeria are imported mainly from China, Spain and Italy, while the cements are produced locally with some imported additives. The Spanish and Italian tiles are seldom used due to high cost. The building materials were collected randomly from different brick-making industries and building construction sites, except ceramic tiles, which were obtained from the retailers. Each sample was taken to the laboratory, pulverized and homogenized in order to have the same matrix and characteristics as the standard sample. They were then packed into cylindrical thin-walled plastic containers whose dimensions are 7.5 cm × 6.5 cm in height and diameter, respectively.

The sealed samples were then left for about 28 days in order to achieve secular equilibrium between ^{226}Ra and its progeny, as well as between ^{232}Th and its own progeny. Since radium (^{226}Ra) and its progeny produce 98.5% of the radiological effects of the uranium series, the contributions of ^{238}U and the precursors of ^{226}Ra are normally ignored (Hassan *et al.*, 2010). Therefore, the reference of the uranium series is often ^{226}Ra rather than ^{238}U (Farai and Ademola, 2005).

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were measured using a low-background NaI(Tl) detector coupled to an IBM-compatible multichannel analyzer with Sampo 90 software for data analysis. The detector was calibrated using ^{109}Cd , ^{57}Co , ^{137}Cs , ^{54}Mn and ^{22}Na for their known energy and peak width of γ -ray emissions, while the counting efficiency was determined using a standard soil sample (IAEA soil-375) obtained from the IAEA, Vienna Austria. The radionuclides were identified according to their individual photo peaks, which are 1.76 MeV (^{214}Bi) for ^{226}Ra , 2.62 MeV (^{208}Tl) for ^{232}Th and 1.4603 MeV for ^{40}K (Yasir *et al.*, 2007). The activity concentration of individual radionuclides was obtained by direct comparison of the total counts per second (cps) obtained from individual samples of the building materials with the count per second (cps) obtained for the same radionuclide in the standard sample. The activity concentration was obtained using the following equation:

$$C_s = \frac{A_s M_{\text{std}}}{A_{\text{std}} M_s} C_{\text{std}} \quad (1)$$

where C_s is the concentration of the radionuclide in the analyzed sample (Bq.kg^{-1}), C_{std} is the concentration of the radionuclide in the standard sample (Bq.kg^{-1}), M_s is the mass of the analyzed sample (g), M_{std} is the mass of the standard sample (g),

TABLE I
Activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th measured in different types of building materials used in southwest Nigeria.

Type of building material	No of sample	Activity concentration ($\text{Bq}\cdot\text{kg}^{-1}$)		
		^{40}K Range (Mean)	^{226}Ra Range (Mean)	^{232}Th Range (Mean)
Cement	15	3.7 – 722.7 (218.7 ± 11.5)	8.7 – 217.0 (58.9 ± 12.2)	4.7 – 45.9 (16.3 ± 3.9)
Tiles	15	160.7 – 854.1 (397.9 ± 41.0)	7.8 – 115.2 (40.1 ± 12.0)	6.3 – 51.9 (19.2 ± 3.3)
Gravel	15	38.6 – 887.5 (400.9 ± 42.4)	6.4 – 110.2 (39.8 ± 12.5)	3.7 – 50.8 (17.3 ± 4.0)
Bricks	15	10.2 – 611.2 (193.5 ± 20.5)	4.4 – 113.9 (33.2 ± 11.7)	2.6 – 132.3 (27.4 ± 4.0)
Sand	10	22.0 – 641.9 (274.7 ± 4.3)	6.4 – 152.5 (61.3 ± 23.2)	10.5 – 33.5 (20.0 ± 3.1)

A_s is the count rate (cps) of the analyzed sample, and A_{std} is the count rate of the standard sample (cps).

3. Results and discussion

The specific activities of the measured radionuclides, *i.e.* ^{40}K , ^{226}Ra and ^{232}Th , in the five commonly used building materials in southwest Nigeria expressed in $\text{Bq}\cdot\text{kg}^{-1}$ dry mass of the samples are presented in Table I. The activity concentration of ^{40}K ranged from 3.7–887.5 $\text{Bq}\cdot\text{kg}^{-1}$, ^{226}Ra ranged from 4.4–217.0 $\text{Bq}\cdot\text{kg}^{-1}$ and that of ^{232}Th ranged from 2.6–132.3 $\text{Bq}\cdot\text{kg}^{-1}$ in all the analyzed samples. The highest mean concentrations of ^{40}K , ^{226}Ra and ^{232}Th are found in gravel, sand and bricks, respectively, while the lowest mean concentrations of the three radionuclides, respectively, are found in brick and cement samples, as shown in Table I. When compared with the results from other locations around the world, the mean activity concentrations of the radionuclides in the five different building materials analyzed in this study are comparable with the values reported in the literature. Table II presents the comparison of the results of the present work with values obtained by different researchers from other countries. It is observed that the activity concentration of ^{40}K in cement is lower in this study than most of the values from the other countries considered, except Australia and China. ^{226}Ra concentrations are higher in this study than the results from other countries, except Brazil, while the ^{232}Th concentrations obtained for cement in this study are lower than the values from other countries, except Greece. For tiles, the concentrations of ^{40}K , ^{226}Ra and ^{232}Th are all lower than the values from most countries. The trend in bricks is similar to that of tiles, but concentrations of ^{40}K , ^{226}Ra , and ^{232}Th in gravel are higher in value compared with the concentrations obtained in other countries.

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TABLE II
Comparison of activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in building materials analyzed in this study with values from other countries.

Type of building materials	Country	^{40}K	^{226}Ra	^{232}Th	Reference
Cement	Algeria	422.0	41.0	27.0	Amrani and Tahtat (2001)
	Australia	114.7	51.8	48.1	Berekta and Matthew (1985)
	Brazil	564.0	61.7	58.5	Malanca <i>et al.</i> (1993)
	China	173.2	56.5	36.5	Xinwei (2005)
	Greece	241.0	20.0	13.0	Stoulos <i>et al.</i> (2003)
	India	432.2	37.0	24.1	Kumar <i>et al.</i> (1999)
	Iran	290.8	39.6	28.9	Mehdizadeh <i>et al.</i> (2011)
	Italy	218	38	22	Rizzo <i>et al.</i> (2001)
	Netherlands	230.0	27.0	19.0	Ackers <i>et al.</i> (1985)
	Macedonia	264 ± 50	42 ± 10	28 ± 8	Stojanovska <i>et al.</i> (2010)
	Turkey	267 ± 102	41 ± 27	26 ± 19	Turhan (2007)
	Nigeria	218.7 ± 11.5	58.9 ± 12.2	16.2 ± 3.9	Present study
Tiles	Algeria	410.0	55.0	41.0	Amrani and Tahtat (2001)
	China	480.0	73.0	62.0	Xinwei (2004)
	Egypt	569.0	77.0	64.0	Higgy <i>et al.</i> (2000)
	Greece	411–786	25–174	29–47	Kristić <i>et al.</i> (2007)
	India	24	28	64	Kumar <i>et al.</i> (2003)
	Italy	708.0	53	53	Righi <i>et al.</i> (2009)
	Pakistan	476.9	70.3	62.1	Turhan <i>et al.</i> (2008)
	Nigeria	397.9 ± 41.0	40.1 ± 12.0	19.1 ± 3.4	Present study
Gravel	Australia	171.0	13.9	14.8	Soranin and Steger (1983)
	Brazil	933.0	10.3	ND	Malanca <i>et al.</i> (1993)
	Iran	450.0	20.4	6.3	Mehdizadeh <i>et al.</i> (2011)
	Lebanon	4.6	27.5	13.8	Kobeissi <i>et al.</i> (2008)
	USA	14.8	33.3	33.3	Ingersoll (1983)
	Nigeria	400.9 ± 42.4	39.8 ± 12.5	17.3 ± 4.0	Present study
Bricks	Algeria	675	65	51	Amrani and Tahtat (2001)
	China	714	59	50	Xinwei (2005)
	Greece	710	35	45	Papaefthymiou and Gouseti (2008)
	Iran	851.4 ± 15	37.0 ± 1.5	12.2 ± 0.7	Mehdizadeh <i>et al.</i> (2011)
	Kuwait	332	12	7	Bou-Rabee and Bem (1996)
	Pakistan	692.63	45.50	61.76	Tufail <i>et al.</i> (2007)
	Turkey	923.4 ± 161.0	69.9 ± 11.7	11.7	Turhan <i>et al.</i> (2008)
	Nigeria	193.5 ± 20.5	33.2 ± 11.7	27.4 ± 4.0	Present study

TABLE II
Continued.

Type of building materials	Country	⁴⁰ K	²²⁶ Ra	²³² Th	Reference
Sand	Algeria	74 ± 7	12 ± 1	7 ± 1	Amrani and Tahtat (2001)
	Bangladesh	303.1 ± 141.9	14.5±8.2	34.7±2.4	Mantazul <i>et al.</i> (1998)
	Brazil	51	10.2	12.6	Malanca <i>et al.</i> (1993)
	Cameroon	586 ± 13	14 ± 1	31 ± 1	Ngachin <i>et al.</i> (2007)
	Greece	367 ± 204	18 ± 7	17 ± 10	Stoulos <i>et al.</i> (2003)
	India	65.5	9.4	52.05	Kumar <i>et al.</i> (2003)
	Kuwait	360 ± 14	7.9 ± 0.7	7.2 ± 0.3	Bou-Rabee and Bem (1996)
	Turkey	527.2 ± 129.2	22.9 ± 12.9	26.4 ± 16.2	Turhan <i>et al.</i> (2008)
	Zambia	714 ± 17	24 ± 1	26 ± 2	Hayambu <i>et al.</i> (1995)
	Nigeria	274.7 ± 4.3	61.3 ± 23.2	20.0 ± 3.1	Present study

Estimation of radiological hazard indices

The measured activity concentrations of the natural radionuclides in the building materials were used to estimate the radiological hazards associated with the use of such materials in building construction. The radiation hazard parameters estimated in this study include: radium equivalent activity, the internal hazard index (H_{in}), the gamma index (I_γ), the alpha index (I_α) and the annual gonadal equivalent dose (AGED).

The radium equivalent activity (Ra_{eq}) is a common index used to represent the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K by a single quantity, which takes into account all the radiation hazards associated with the materials. It is calculated based on the estimation that 370 Bq.kg⁻¹ of ²²⁶Ra, 259 Bq.kg⁻¹ of ²³²Th and 4.810 Bq.kg⁻¹ of ⁴⁰K produce the same gamma-ray dose rate. The radium equivalent activity is expressed as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \quad (2)$$

where A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in Bq.kg⁻¹. In order to limit the external dose to below 1.5 mSv per year, indicating safe use of any material in building construction, the radium equivalent must be less than 370 Bq.kg⁻¹ (UNSCEAR, 2000). The calculated Ra_{eq} for the building material samples considered in this study is presented in Table III. It is observed that the Ra_{eq} for the locally sourced materials and the imported ceramic tiles are all below the maximum permissible limit of 370 Bq.kg⁻¹. It can therefore

TABLE III
Range and mean values of calculated radiation hazard parameters in building materials.

Type of building material	Radiation hazard parameters									
	Ra _{eq}		I _γ		H _{in}		I _a		AGED	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Cement	17.9 – 310.4	99.0 ± 18.7	0.06 – 1.10	0.35	0.07 – 1.35	0.43	0.04 – 1.08	0.29	54.3 – 1003.2	318.5
Tiles	34.7 – 208.8	98.1 ± 19.9	0.13 – 0.73	0.36	0.12 – 0.87	0.37	0.04 – 0.58	0.20	121.1 – 651.7	328.9
Gravel	11.7 – 227.7	93.2 ± 21.4	0.04 – 0.82	0.34	0.05 – 0.91	0.36	0.03 – 0.55	0.19	35.3 – 735.8	312.1
Bricks	13.4 – 266.9	84.7 ± 18.7	0.05 – 0.93	0.30	0.05 – 0.93	0.32	0.02 – 0.56	0.17	41.0 – 799.1	270.4
Sand	73.6 – 232.2	110.9 ± 27.9	0.27 – 0.81	0.40	0.22 – 1.04	0.47	0.03 – 0.76	0.31	243.9 – 741.1	359.1

TABLE IV
Comparison of radium equivalent activity with values obtained by other researchers.

Country	Radium equivalent activity (Bq.kg ⁻¹)					Reference
	Cement	Tiles	Gravel	Bricks	Sand	
Algeria	112	145	58	190	28	Amrani and Tahta (2001)
Bangladesh	172.8	–	121.29	127.14	87.52	Mantazul <i>et al.</i> (1998)
Cameroon	70.1	36.03	80.12	193.34	94.7	Ngachin <i>et al.</i> (2007)
China	128	–	79	95	202	Xinwei (2004)
Cuba	78	–	49	140	55	Brigido Flores <i>et al.</i> (2008)
India	–	121	72.1	69.15	135	Kumar <i>et al.</i> (2003)
Iran	103.32	–	64.11	58.58	–	Mehdizadeh <i>et al.</i> (2011)
Lebanon	92.2	–	28.4	–	23	Kobeissi <i>et al.</i> (2008)
Malaysia	69.6	93.7	–	130.7	102.6	Yasir <i>et al.</i> (2007)
Pakistan	69	–	–	106	91	Faheem <i>et al.</i> (2008)
Turkey	101.9	195.8	95.6	240.4	101.2	Turhan <i>et al.</i> (2008)
Zambia	79	–	33	180	84.15	Hayambu <i>et al.</i> (1995)
Nigeria	99.0 ± 18.7	98.1 ± 19.9	93.2 ± 21.4	84.7 ± 18.7	110.9 ± 27.9	Present study

be recommended that all the materials are safe for use in building construction. Table IV shows the comparison of the radium equivalent activity in this work with values obtained by other researchers from different countries of the world.

To assess the excess gamma radiation from the building materials and to further ascertain the safety of the usage of the materials in building, the gamma index (I_γ) was evaluated using the formula given by the European Commission as:

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \leq 1 \text{ (EC, 1999)}. \quad (3)$$

The European Commission introduces two dose criteria for the gamma dose of building materials, which includes an exemption level (lower limit) of 0.3 mSv.y⁻¹

and an upper limit of 1.0 mSv.y^{-1} . If an exemption level of 0.3 mSv.y^{-1} is considered, the value of I_γ should be below 0.5 for materials used in bulk quantities such as cement and bricks, but in the case whereby the upper limit of 1 mSv.y^{-1} is considered, the value of I_γ should be less than unity for bulk material usage (Mehdizadeh *et al.*, 2011). For covering and decorative materials with restricted use such as tiles, I_γ should be less than 2 and 6, supposing the control values of 0.3 and 1 mSv.y^{-1} , respectively. The range and mean values of I_γ for the five common building materials considered in this study are shown in Table III. It is observed that the highest values of I_γ obtained for all the building materials are higher than 0.5 but lower than the upper limit of 1.0. The mean values of the gamma index for the building materials are all lower than the 0.5 limit, indicating an excess gamma dose of less than 0.3 mSv.y^{-1} for the dwellers of buildings constructed with such materials.

There is also a radiation hazard threat to respiratory organs due to ^{222}Ra , and its short-lived decay products. To assess this threat, two radiation hazard indices were used in this study, which are: the internal hazard index and the alpha index. These indices are respectively given as:

$$H_{\text{in}} = \frac{A_{\text{Ra}}}{300} + \frac{A_{\text{Th}}}{200} + \frac{A_{\text{K}}}{3000} \quad (\text{Otoo } et al., 2011) \quad (4)$$

$$I_\alpha = \frac{A_{\text{Ra}}}{200} \quad (\text{Tufail } et al., 2007). \quad (5)$$

For the safe use of any material in the construction of dwellings, the internal hazard index should be less than or equal to unity. Also, the recommended exemption level and recommended upper limit of ^{226}Ra activity concentrations as applied in many countries of the world are 100 Bq.kg^{-1} and 200 Bq.kg^{-1} , respectively (Tufail *et al.*, 2007), which implies that the exemption level and the recommended upper limit of I are 0.5 and 1.0, respectively. If the concentration of ^{226}Ra in any building material exceeds the upper limit of 200 Bq.kg^{-1} , it is not impossible that radon exhalation from the material could cause indoor radon build-up exceeding 200 Bq.m^{-3} . As shown in Table III, the calculated mean values of H_{in} and I_α obtained for the different building materials examined in this study are all lower than the exemption levels and far lower than the upper limit, which implies that all the building materials can be used without any restrictions.

Organs with rapidly dividing cells, *e.g.*, gonads, the active bone marrow and bone surface cells, are known to be the most radiosensitive and are considered as organs of interest by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1988). Therefore, it is important to evaluate the annual gonadal equivalent dose for dwellers of houses built with known activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K . The annual gonadal equivalent dose

(AGED) is estimated using the formula presented by Xinwei *et al.* (2006) and Hassan *et al.* (2010):

$$\text{AGED} = (3.09A_{\text{Ra}} + 4.18A_{\text{Th}} + 0.314A_{\text{K}}) \mu\text{Sv.y}^{-1}. \quad (6)$$

The model used for arriving at this formula was implemented for a typical cavity-type house with infinitely thick walls, thereby making it appropriate to compare the AGED of a house containing concentrations of ^{226}Ra , ^{232}Th and ^{40}K equal to the world average values in soil (25, 25 and 370 Bq.kg^{-1} , respectively) with those obtained using a given material (Hassan *et al.*, 2010). The range and the mean values of the AGED evaluated for all the samples are presented in Table III, which range between 35.3 $\mu\text{Sv.y}^{-1}$ in gravel and 1003.2 $\mu\text{Sv.y}^{-1}$ in cement. The mean values of the AGED for the five different building materials lie within a narrow range of 270.4 $\mu\text{Sv.y}^{-1}$ in bricks and 359.1 $\mu\text{Sv.y}^{-1}$ in sand.

4. Conclusion

The gamma spectrometric method was used for the assessment of ^{40}K , ^{226}Ra and ^{232}Th concentrations in five commonly used building materials in southwest Nigeria. The lowest mean concentrations of ^{40}K and ^{226}Ra were found in bricks, while the lowest mean concentration of ^{232}Th was obtained in cement. The highest mean concentrations of ^{40}K , ^{226}Ra and ^{232}Th were found in gravel, sand and brick samples, respectively. The concentrations of the three radionuclides obtained in this study are in good agreement with the concentrations of building materials from other locations around the world. From the radiological point of view, the results of the various radiation hazard indices considered for these building materials are all lower than the maximum permissible levels. It can therefore be concluded that all the building materials are safe for use in building construction.

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