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Studies on energy absorption and exposure buildup factors in some solutions of alkali metal chlorides

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Buildup of photons has been investigated through the solutions LiCl, NaCl and KCl with different salt contents in the energy region 0.015-15 MeV up to a penetration depth of 40 mean free paths. Two types of buildup factors, the gamma ray energy absorption ($EABF$) and exposure buildup factors (EBF) have been calculated simultaneously using the five parameter geometric progression (G-P) fitting formula. The influence of photon energy, penetration depth and chemical composition on the buildup factors has been studied. Also, a comparison has been made between the values of $EABF$ and EBF if any significant variation occurs between them. Moreover, the Monte Carlo simulation study has been made for the purpose of the comparison.

Key words: Gamma ray buildup factor, energy absorption, exposure, solution, Monte Carlo method.

INTRODUCTION

The buildup of photons in various kinds of materials has long been considered as the subject of various investigations in radiation shielding and dosimetry. Brar et al. (1999) have focused on the buildup factor studies of HCO-materials as a function of weight fraction of constituent elements. Brar et al. (1998) have investigated the effect of weight fractions of Fe and Si on buildup factors of some soil samples. Singh et al. (2008) have investigated the variation of energy absorption buildup factors with incident photon energy and penetration depth for some commonly used solvents. Manohara et al. (2010) have studied the energy absorption buildup factors for thermoluminescent dosimetric materials and their tissue equivalence which are of importance in radiation dosimetry, diagnostics and therapy. Recently, chemical composition dependence of exposure buildup factors for some polymers has been studied (Singh et al., 2009). Singh et al. (2010) have studied the buildup of gamma ray photons in fly ash concretes. An experimental investigation based on the effect of finite sample dimensions and total scatter acceptance angle on the gamma ray buildup factor has been made before (Singh and Kumar, 2008). There are different available methods to calculate the buildup factor such as geometric progression (G-P) fitting method (Harima et al., 1986) and invariant embedding method (Sakamoto and Tanaka, 1988; Shimizu, 2002; Shimizu et al., 2004). A reliable document for these methods is American National Standards ANSI/ANS 6.4.3 (1991) which provided buildup factor data for 23 elements, one compound and two mixtures (that is, air and water) and concrete at energies in the range 0.015-15 MeV up to penetration depths of 40 mean free path by using the G-P method. The developed G-P fitting formula is known to be accurate within a few percent errors (Harima et al., 1986; Harima, 1983). Harima (1993) has made an extensive historical review and an assessment for the status of buildup factor calculations and applications.

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When it comes to material in dissolved form, it is noteworthy that the use of absorbers in the form of liquids has some definite advantages as compared with solid absorbers: the criterion of homogeneity of the absorber is satisfied instantly, and the strength of the absorber can easily be varied by changing the relative amount of solute and solvent (Kumar et al., 2006). However, there is almost no study based on the calculation of buildup factors for materials in their liquid forms such as solutions with different salt contents. Hence, we embarked on a study including the calculation of energy absorption and exposure buildup factors including their dependence on energy, penetration depth and chemical composition in the energy region of 0.015-15 MeV up to a penetration depth of 40 mean free paths. Also, the calculated buildup factors have been compared with those obtained using Monte Carlo method.

COMPUTATIONAL WORK

To calculate the buildup factors, the G-P fitting parameters were obtained by the method of interpolation from the equivalent atomic number (Z_{eq}). Computations are illustrated step by step as follows:

(i) Calculation of the equivalent atomic number Z_{eq}
(ii) Calculation of geometric progression (G-P) fitting parameters
(iii) Calculation of energy absorption and exposure buildup factors

However, the interaction of gamma rays with materials is based on domination of different partial photon interaction processes in different energy regions, thus Z_{eq} is an energy dependent parameter. Since the buildup effect arises from multiple scattering events, Z_{eq} is derived from the contribution of Compton scattering process. At the first step, the equivalent atomic number for a particular material has been calculated by matching the ratio, $(\mu/\rho)_{Compton}/(\mu/\rho)_{Total}$, of that material at a specific energy with the corresponding ratio of an element at the same energy. Thus, firstly the Compton partial mass attenuation coefficient, $(\mu/\rho)_{Compton}$, and the total mass attenuation coefficients, $(\mu/\rho)_{Total}$, were obtained for the elements of $Z = 4-40$ and for the solutions in the energy region 0.015-15 MeV, using the WinXCom computer program (Gerward et al., 2001a; Gerward et al., 2004b) (initially developed as XCOM (Berger and Hubbell, 1999)). It was reported by Hubbell (1999) that the envelope of the uncertainty of mass attenuation coefficient is in the order of 1-2% in the energy range from 5 keV to a few MeV. In case of the energies of 1 to 4 keV, the discrepancies are known to reach to a value of 25 to 50%. Recently, Chantler (2000) has extended the investigations below 5 keV concluding the presence of huge discrepancies below 4 keV and derived new theoretical results of substantially higher accuracy in near-edge soft X-ray regions in detail. De Jonge et al. (2005) have measured the mass attenuation coefficients and determined the imaginary component of the atomic form factor of molybdenum over the 13.5-41.5 keV energy range. Tran et al. (2005) have measured the X-ray mass attenuation coefficient of silver using the X-ray extended energy range with high accuracy.

At the second step, to calculate the G-P fitting parameters for elements were taken from the ANSI/ANS-6.4.3 (1991) standard reference database which provides the G-P fitting parameters for elements from beryllium to iron in the energy region 0.015-15 MeV up to 40 mfp. The G.P. fitting buildup factor coefficients of the used materials were interpolated according to the given formula (Gupta and Sidhu, 2012).

At the final step, these parameters were used to calculate the energy absorption and exposure buildup factors from the G-P fitting formula (Harima et al., 1986). While the exposure buildup factor, EBF is based on the energy absorption response of air; that is, exposure is assumed to be equivalent to absorbed dose in air as measured by a nonperturbing detector, the energy absorption buildup factor, $EABF$ refers to that absorbed or deposited energy in the attenuating material.

In radiologic sciences, practical problems usually are not legible for analytical solutions. Thus Monte Carlo techniques as strong computational tools are applied in radiation protection and dosimetry. Utilization of Monte Carlo codes such as MCNP (versions 4C, X and 5) has increased over past decade.

The MCNP code being acronym for Monte Carlo N-Particle, was developed at Los Alamos Laboratory (Los Alamos, NM) originally as a neutron and photon transport for reactor analysis in general. This code has been updated and improved repeatedly and their latest version, MCNPX and MCNP5, includes charged particle transport algorithms based on the best available models. This program provides several options for developing spatial and energetic distributions using complex geometric shapes (Briesmeister, 2000).

The code supports a wide variety of scoring options and radiation source modeling. Several variance reduction techniques are also available, allowing performance optimization for a more efficient determination of results. The user must also specify for each problem the *tallies*, or memory regions in which quantities such as energy, flux, etc. are recorded by MCNP. In our problem, we are interested in absorbed energy and/or photon flux either individual after passing through layers of material.

RESULTS AND DISCUSSION

The equivalent atomic numbers of the solutions are given in Tables 1, 2 and 3. The energy absorption and exposure buildup factors for various energies and mixtures are shown in graphical form at specific penetration depths (Figures 1 to 2a, b). In the energy range of interest (0.015-15 MeV), the photon interaction processes namely photoelectric absorption, Compton scattering and pair production partially dominate in different energy regions. Since the buildup of photons arises mainly from multiple scattering, the absorption processes (photoelectric absorption, pair production) reduces the values of $EABF$ and EBF in the low and high energy regions, respectively, and the scattering process (Compton scattering) increases the values of $EABF$ and EBF at the intermediate energy region. The maximum values of $EABF$ and EBF have been observed for LiCl, NaCl and KCl solutions at energy 0.15 MeV except for the KCl solution of highest Z_{eq} (salt content = 0.2) for which the maximum values occur at 0.2 MeV. At this energy the main photon interaction process is Compton scattering. It has been observed that the solutions of high Z_{eq} (that is, KCl (salt content = 0.2) mainly possess the lower values of $EABF$ and EBF whereas the solutions of

Table 1. Equivalent atomic numbers of LiCl solutions.

Energy (MeV)	Equivalent atomic number				
	LiCl solution				
	0.04*	0.08	0.12	0.16	0.2
1.50E-02	8.1	8.7	9.3	9.7	10.1
2.00E-02	8.2	8.8	9.4	9.8	10.3
3.00E-02	8.3	8.9	9.5	10.0	10.4
4.00E-02	8.3	9.0	9.6	10.1	10.5
5.00E-02	8.4	9.1	9.6	10.1	10.6
6.00E-02	8.4	9.1	9.7	10.2	10.6
8.00E-02	8.4	9.1	9.7	10.2	10.7
1.00E-01	8.4	9.2	9.8	10.3	10.8
1.50E-01	8.5	9.2	9.8	10.3	10.8
2.00E-01	8.5	9.2	9.9	10.4	10.9
3.00E-01	8.5	9.3	9.9	10.4	10.9
4.00E-01	8.5	9.3	9.9	10.4	10.9
5.00E-01	8.5	9.3	9.9	10.5	10.9
6.00E-01	8.5	9.3	10.0	10.5	11.0
8.00E-01	8.5	9.3	10.0	10.5	11.0
1.00E+00	8.5	9.3	10.0	10.5	11.0
1.50E+00	7.0	7.4	7.7	8.1	8.5
2.00E+00	6.9	7.2	7.5	7.8	8.2
3.00E+00	6.9	7.2	7.5	7.8	8.1
4.00E+00	6.9	7.2	7.5	7.8	8.1
5.00E+00	6.9	7.2	7.5	7.8	8.1
6.00E+00	6.9	7.2	7.5	7.8	8.0
8.00E+00	6.9	7.2	7.5	7.7	8.0
1.00E+01	6.9	7.2	7.4	7.7	8.0
1.50E+01	6.9	7.1	7.4	7.7	8.0

*refers to the salt content (g/cm^3).

low Z_{eq} (that is, NaCl (weight fraction = 0.04)) mainly dominate the higher values of $EABF$ and EBF. Actually, the reason for higher values of buildup factor in NaCl solutions is its lower Z_{eq} which leads to abundance of Compton scattering events.

Figures 3 and 4a, b, c, d show the influence of penetration depth on buildup factors at the fixed energies. From the above mentioned figures and data in Table 5, it is understood that $EABF$ and EBF values lie between 1 to 1.7, 1 to 1.7 and 1 to1.6 at 0.015 MeV for LiCl, NaCl and KCl solutions, respectively. At this low energy the main interaction process is photoelectric absorption hence the fast removal of photons give rise to the lower values of buildup factors. For photon energies of 0.15 MeV or lower, $EABF$ and EBF increase with the decrease in Z_{eq}. $EABF$ and EBF seem to be independent of variation in chemical composition at 1.5 MeV, thus the buildup factor values remain the same for different solutions. At 15 MeV, an inverse variation occurs as such buildup factors increase with the increase in Z_{eq} after 20 mfp for the given materials. $EABF$ and EBF for the given solutions as a function of weight fractions of Na and H_2O are shown in Figures 5 and 6 a,b. Similar figures have been produced for LiCl and KCl in the course of our studies. It has been shown that for the energies below 1.5 MeV, $EABF$ and EBF increase with the increase in the weight fraction of H_2O whereas they decrease with the increase in weight fraction of Li, Na and K present in solutions. Also, there are significant variations between $EABF$ and EBF where the larger buildup factors occur. In general, $EABF$ have higher values than EBF due to the fact that the materials under investigation have higher Z_{eq} values than that of air. Thus, when the Z_{eq} increases the energy absorption in the medium will be more than that of absorption in air. Figure 7 a, b gives the relative differences (%) between $EABF$ and EBF. In these figures, the positive values

Table 2. Equivalent atomic numbers of NaCl solutions.

Energy (MeV)	Equivalent atomic number				
	NaCl solution				
	0.04	0.08	0.12	0.16	0.2
1.50E-02	8.0	8.6	9.0	8.4	9.8
2.00E-02	8.1	8.6	9.1	8.4	9.9
3.00E-02	8.2	8.7	9.2	8.5	10.1
4.00E-02	8.2	8.8	9.3	8.6	10.1
5.00E-02	8.2	8.8	9.3	8.6	10.2
6.00E-02	8.2	8.8	9.4	8.7	10.2
8.00E-02	8.3	8.9	9.4	8.7	10.3
1.00E-01	8.3	8.9	9.4	8.8	10.3
1.50E-01	8.3	9.0	9.5	8.8	10.4
2.00E-01	8.3	9.0	9.5	8.9	10.4
3.00E-01	8.3	9.0	9.5	8.9	10.4
4.00E-01	8.3	9.0	9.6	8.9	10.5
5.00E-01	8.4	9.0	9.6	9.0	10.5
6.00E-01	8.4	9.0	9.6	9.0	10.5
8.00E-01	8.4	9.1	9.6	9.0	10.5
1.00E+00	8.4	9.1	9.6	9.0	10.5
1.50E+00	7.0	7.3	7.7	7.5	8.3
2.00E+00	6.9	7.2	7.5	7.4	8.1
3.00E+00	6.9	7.2	7.5	7.3	8.1
4.00E+00	6.9	7.2	7.5	7.3	8.0
5.00E+00	6.9	7.2	7.4	7.3	8.0
6.00E+00	6.9	7.2	7.4	7.3	8.0
8.00E+00	6.9	7.2	7.4	7.3	8.0
1.00E+01	6.9	7.1	7.4	7.3	8.0
1.50E+01	6.9	7.1	7.4	7.3	8.0

Table 3. Equivalent atomic numbers of KCl solutions.

Energy (MeV)	Equivalent atomic number				
	KCl solution				
	0.04	0.08	0.12	0.16	0.2
1.50E-02	8.4	9.2	9.9	10.5	11.0
2.00E-02	8.5	9.4	10.0	10.6	11.2
3.00E-02	8.6	9.5	10.2	10.8	11.4
4.00E-02	8.7	9.6	10.3	10.9	11.5
5.00E-02	8.7	9.6	10.4	11.0	11.6
6.00E-02	8.8	9.7	10.4	11.1	11.6
8.00E-02	8.8	9.8	10.5	11.1	11.7
1.00E-01	8.9	9.8	10.5	11.2	11.8
1.50E-01	8.9	9.9	10.6	11.3	11.8
2.00E-01	8.9	9.9	10.7	11.3	11.9
3.00E-01	9.0	10.0	10.7	11.4	12.0
4.00E-01	9.0	10.0	10.7	11.4	12.0
5.00E-01	9.0	10.0	10.8	11.4	12.0
6.00E-01	9.0	10.0	10.8	11.4	12.0
8.00E-01	9.0	10.0	10.8	11.4	12.0

Table 3. Contd.

1.00E+00	9.0	10.0	10.8	11.4	12.0
1.50E+00	7.2	7.7	8.2	8.7	9.2
2.00E+00	7.0	7.5	7.9	8.4	8.8
3.00E+00	7.0	7.4	7.8	8.3	8.7
4.00E+00	7.0	7.4	7.8	8.2	8.7
5.00E+00	7.0	7.4	7.8	8.2	8.7
6.00E+00	7.0	7.4	7.8	8.2	8.6
8.00E+00	7.0	7.4	7.8	8.2	8.6
1.00E+01	7.0	7.4	7.8	8.2	8.6
1.50E+01	7.0	7.4	7.8	8.2	8.6

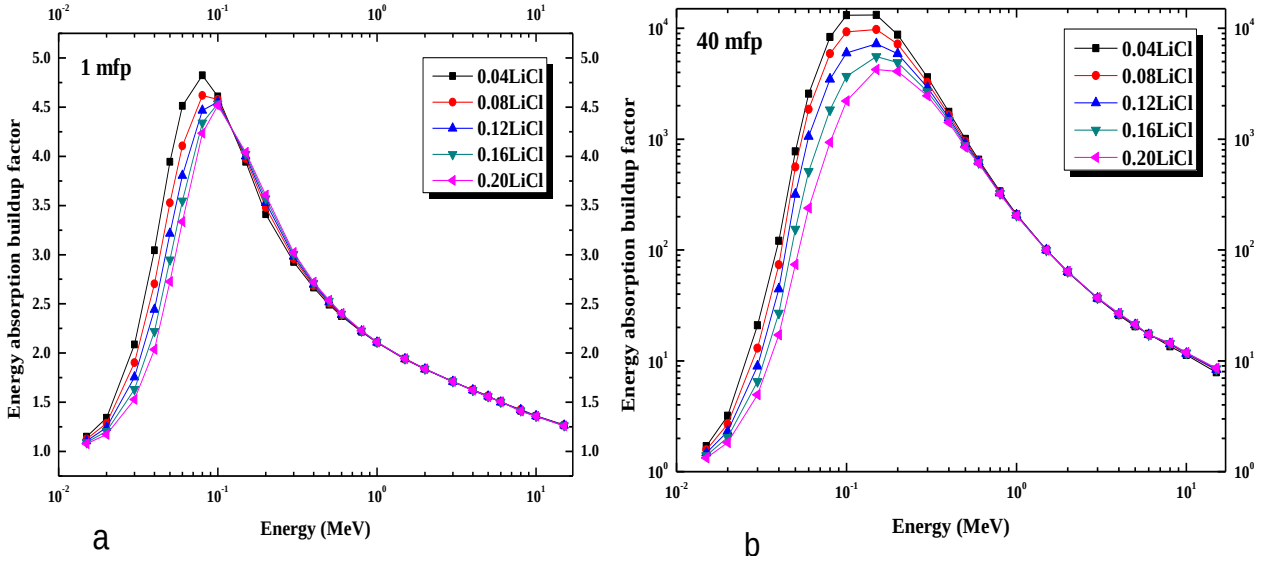

Figure 1(a, b). The energy absorption buildup factor for NaCl solution in the energy region 0.015-15 MeV at 1 and 40 mfp.

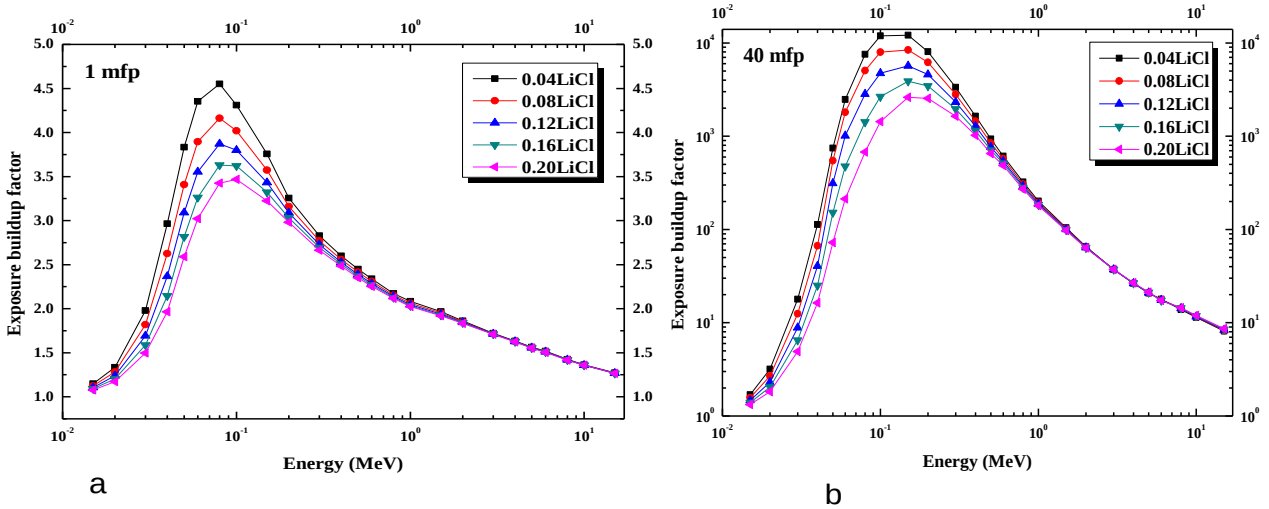

Figure 2(a, b). The exposure buildup factor for NaCl solution in the energy region 0.015-15 MeV at 1 and 40 mfp.

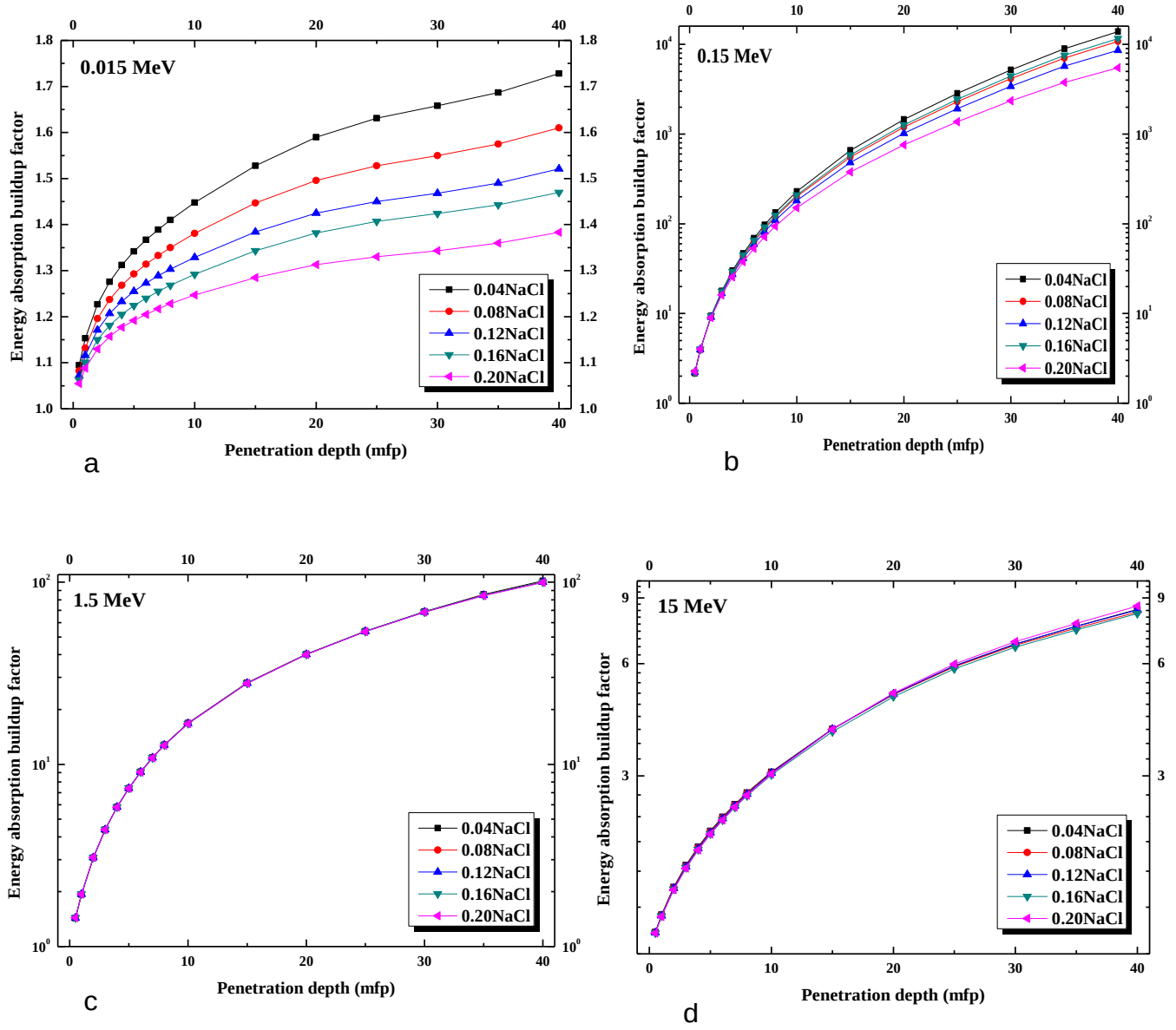

Figure 3(a, b, c, d). The energy absorption buildup factor for NaCl solution up to 40 mfp at 0.015, 0.15, 1.5, 15 MeV.

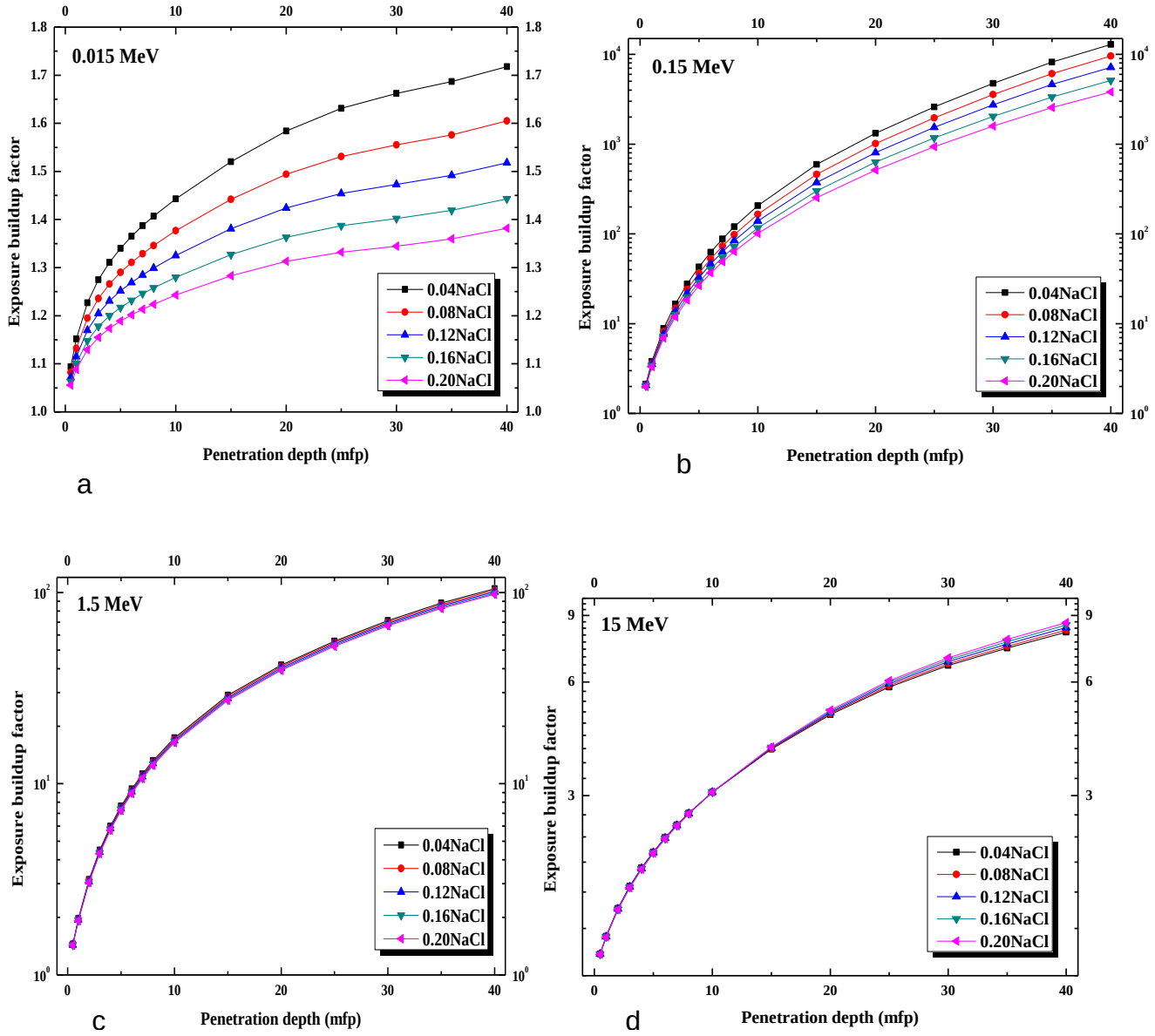

Figure 4(a, b, c, d). The exposure buildup factor for NaCl solution up to 40 mfp at 0.015, 0.15, 1.5, 15 MeV.

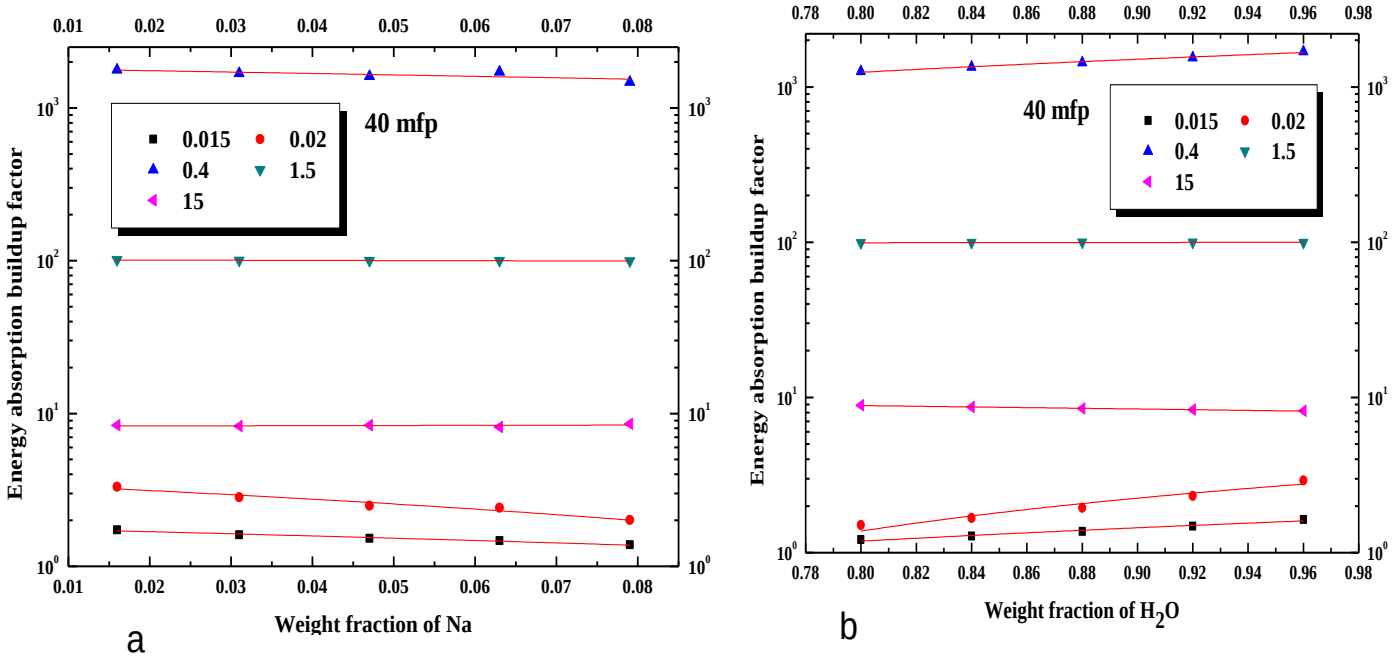

Figure 5(a, b). The energy absorption buildup factors for the given solutions as a function of weight fractions of Na and H_2O.

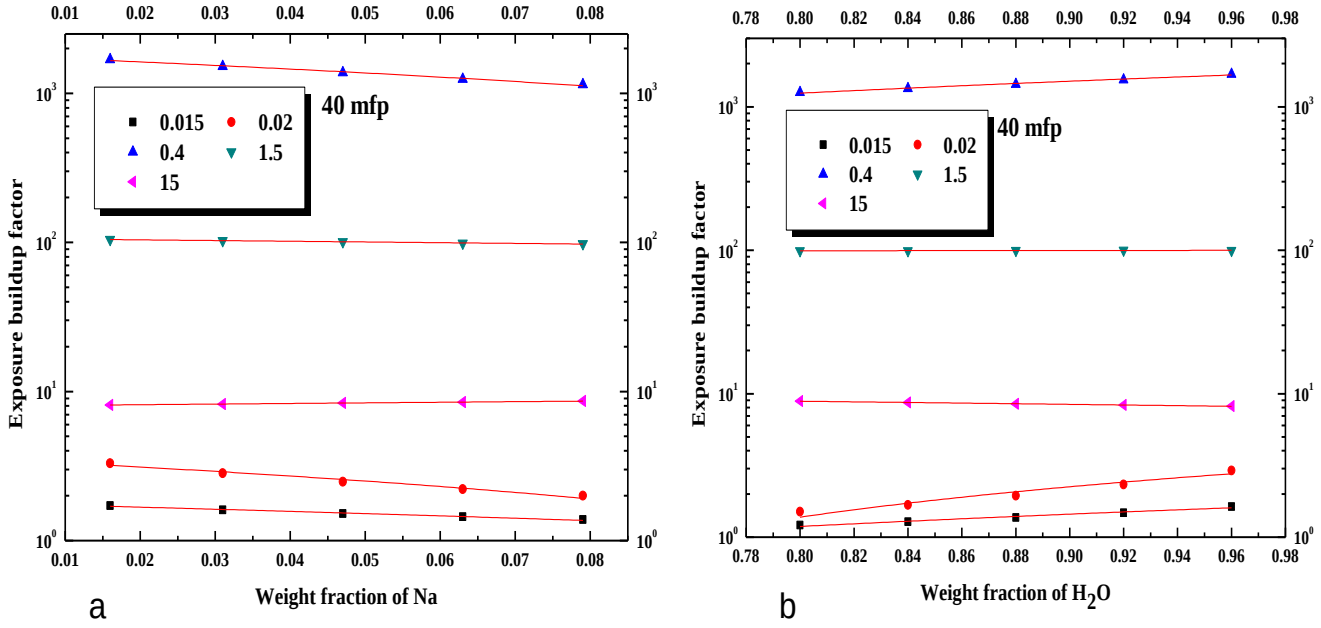

Figure 6(a, b). The exposure buildup factors for the given solutions as a function of weight fractions of Na and H_2O.

of differences (%) refer to the higher values of $EABF$ when compared to EBF.

Over past few years, Asano and Sakamoto (2007) have evaluated the buildup factors of two typical heavy concretes to improve the capability of the various materials for the shielding wall by using the Monte Carlo simulation code, EGS4. They also compared their calculated values by that of concrete in ANSI/ANS-6.4.3 (1991) standard reference database. Both of the calculations are in good agreement except for the slight differences which may be due to (a) the ANSI/ANS data are based on the calculation result data by using the

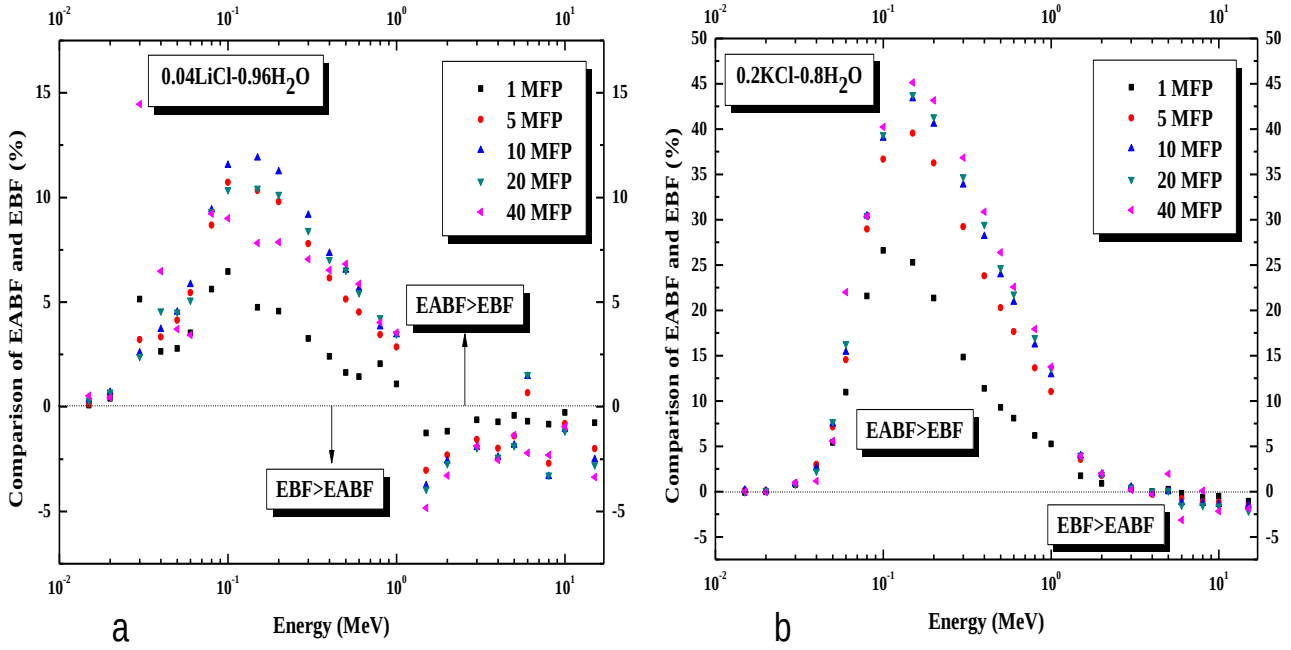

Figure 7(a, b). Difference (%) between $EABF$ and EBF for 0.04LiCl and 0.2KCl solutions in the energy region 0.015-15 MeV up to 40 mfp.

Table 4 (a,b,c,d). Energy absorption (*EABF*) and exposure buildup factors (*EBF*) of some solutions obtained by Monte Carlo simulation.

Energy (MeV) \ Solutions	(a) Energy absorption buildup factor 1MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.10	1.10	1.07	1.07	1.07	1.05
0.15	3.81	3.78	3.39	3.38	3.38	2.96
1.5	2.10	2.09	2.05	2.06	2.04	1.99
15	1.36	1.36	1.36	1.35	1.35	1.35

Energy (MeV) \ Solutions	(b) Exposure buildup factor 1MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.10	1.10	1.06	1.07	1.07	1.04
0.15	3.27	3.26	3.02	3.01	3.00	2.72
1.5	1.86	1.85	1.83	1.83	1.82	1.79
15	1.23	1.23	1.23	1.23	1.22	1.22

Solutions / Energy (MeV)	(c) Energy absorption buildup factor 10MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.73	1.47	1.27	1.32	1.35	1.21
0.15	138.10	167.71	88.46	86.08	82.23	46.54
1.5	18.12	16.57	22.16	17.28	18.34	22.11
15	2.78	2.50	2.92	2.99	3.50	3.11

Solutions / Energy (MeV)	(d) Exposure buildup factor 10MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.70	1.47	1.27	1.31	1.35	1.21
0.15	97.45	114.07	68.69	67.97	64.68	40.50
1.5	14.07	12.66	18.08	13.51	13.61	18.03
15	2.32	2.17	2.48	2.45	2.86	2.46

Table 5 (a, b, c, d). Energy absorption (*EABF*) and exposure buildup factors (*EBF*) of some solutions obtained by G-P fitting approximation.

Solutions / Energy (MeV)	(a) Energy absorption buildup factor 1MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.13	1.13	1.11	1.08	1.09	1.05
0.15	3.98	3.97	4.00	4.04	4.02	4.09
1.5	1.94	1.94	1.94	1.94	1.94	1.94
15	1.26	1.26	1.26	1.27	1.27	1.25

Solutions / Energy (MeV)	(b) Exposure buildup factor 1MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.12	1.13	1.11	1.08	1.09	1.05
0.15	3.57	3.63	3.42	3.22	3.32	3.06
1.5	1.95	1.95	1.94	1.92	1.92	1.91
15	1.27	1.27	1.27	1.26	1.26	1.26

Solutions / Energy (MeV)	(c) Energy absorption buildup factor 10MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.36	1.38	1.30	1.21	1.25	1.14
0.15	191.30	200.92	167.24	137.72	151.23	111.54
1.5	16.80	16.81	16.77	16.69	16.70	16.62
15	3.05	3.02	3.03	3.08	3.07	3.03

Solutions / Energy (MeV)	(d) Exposure buildup factor 10MFP					
	0.08 LiCl	0.08 NaCl	0.08 KCl	0.2 LiCl	0.2 NaCl	0.2 KCl
0.015	1.35	1.38	1.30	1.21	1.24	1.14
0.15	152.01	165.25	120.54	85.56	101.03	63.20
1.5	17.07	17.10	16.82	16.34	16.40	15.96
15	3.06	3.06	3.06	3.06	3.06	3.08

moments method (Eisenhauer and Simmons, 1975) with parallel beam source and the Monte Carlo code, EGS4 with isotropic emission source, (b) the development of the low energy photon computations in EGS4 such as K-X ray, L-X ray and Bremsstrahlung. It was shown by Shimizu et al. (2004) that the methods based on invariant embedding, G.P fitting and Monte Carlo simulation agree for 18 low-Z materials within small discrepancies. In the present study, the solutions have Z_{eq} values ranging from 7 to 12. Hence, the used materials can be considered as low Z_{eq} materials. When compared with other available approximations such as Berger, Taylor and three exponential, the geometric-progression (G.P) fitting seem to reproduce the buildup factors with better accuracy. Harima et al. (1986) have shown that the absolute values of maximum deviations of exposure build factors for water in G. P. fitting is within 0.5-3%, in three-exponential approach is within 0.4 to 9.3%, in Berger approach is within 0.9 to 42.7% and in Taylor approximation is within 0.4 to 53.2%.

Conclusion

It is shown that G-P fitting is a proper method for estimating photon buildup factor in materials with Z < 20. To make a more robust comparison, in the present work the buildup factor of Alkali metals' salt solution in water was computed using MCNP code. This was carried out for penetration depths of 1 and 10 mean free path. The results appear in Tables 4 and 5. For EBF, in short distances from the source, G-P fitting results are in good agreement with MCNP. Namely, the average deviation between two methods is about 5%, the largest one being 10%. In all energies and solution concentration the buildup factor obtained by MCNP at one m.f.p is lower than G-P fitting method. The largest deviation appears at energy 0.15 MeV.

At longer penetration depth, that is, 10 m.f.p the percentage deviation between the results of two methods on average amounts to 20%. Except 0.015 MeV, the outcome of MCNP is lower than G-P fitting. Besides, the

agreement between two methods is much better in 0.015 MeV than higher energies.

In case of 1 m.f.p, the answers from MCNP and G-P fitting are consistent. In case of 10 m.f.p, remarkable disagreement is observed in some results but due to increasing number of interactions of photon in the matter, the MCNP results are more reliable. For EABF, again the deviation of results in longer penetration depth is much more pronounced. At 10 m.f.p on average being 20% while for 1 m.f.p. it is 8%. At short distance and energies above 1 MeV the buildup factor data resulting from MCNP is higher than G-P fitting. Since in EABF the absorbed dose is dealt with, this fact might be attributed to better precision of MCNP code in considering detailed interaction after occurring of photoelectric effect.

Regarding the data in Tables 4 and 5, one might say G-P fitting method is consistent with MCNP in following cases:

(i) For EBF at short distances and all energies and all type of solutions,
(ii) For EBF at long distances for energy below 50 keV and all types of solutions,
(iii) For EABF at short distances and energies away from 0.15 MeV, where buildup factor is maximum,
(iv) For EABF at long distances the consistency is poor but in some cases the data are consistent away from 0.15 MeV.

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