1. Introduction

Nuclear power is confronted with the problem of safe storage of long lived radioactive wastes, especially very hazardous transactinides. Recently proposals have been made to transmute (or "burn") these wastes either within a reactor, or via very high flux secondary neutron sources powered by high current, intermediate energy accelerators [BO92a, NA92]. In addition, accelerator-based energy amplification has been proposed as an alternative to the present nuclear energy production using the uranium fuel cycle [CA93]. Based on a spallation neutron sources subcritical arrangements shall be used as energy amplifier, at the same time minimizing the problem of possible proliferation of fissile materials by using the thorium fuel cycle. Evaluation of the practicality of these proposed schemes requires, among other factors, the ability to calculate the spectrum and fluence of secondary neutrons from targets of various materials and geometric design. This requires either experimental measurements or nuclear reaction codes capable of reproducing both the microscopic nuclear physics, and the radiation transport through the target.

Experimental measurements are long, expensive, and for some aspects, such as total nucleon emission multiplicity, on the edge of available technology. Additionally, at the energies of 800 - 1600 MeV being suggested in some proposals, there are just few accelerators on which the measurements could be made. The more appealing option is to use nuclear reaction codes. Many exist; we need to gain some insights as to the limits of each, whether results are to be trusted to 20%, 50%, or less accuracy. The present code intercomparison exercise is intended to provide such insight. It is the third one in a series up to now three exercises. The first part was dealing with the calculation of (p,xpyn) double differential cross sections for incident proton energies of 25, 45, 80, 160, 256, 800 and 1600 MeV [BL94a]. The second involved modeling of particle production and transport for nuclear reactions in thick targets [FI95, SO96].

In this report we test the capabilities of models and codes to predict activation yields. Since nuclear reactions at all energies from thresholds up to the highest energies of the primary particles will contribute to residual nuclide production in accelerator-based waste transmutation and energy amplification systems, such an intercomparison has to cover the entire energy range. Reaction types such as compound nucleus reactions, preequilibrium and direct reactions, spallation, fragmentation and fission have to be considered.

In Section 2 we present a summary of contributors to this exercise, and a qualitative description of some of the nuclear models and code options employed. Since in this exercise emphasis is laid upon a comparison of the various calculated contributions with experimental data, we describe the sources and selection criteria of the experimental data in section 3. Section 4 gives details of the methodology of the intercomparison, i.e. the calculations necessary to make the contributed theoretical data comparable to experimental cross sections, the systematics of graphical presentation of the results and, last but not least, an attempt to derive a quantitative measure of the agreement or of the deviations between experimental and theoretical data. In Section 5 we discuss results in some detail, and in Section 6 we present conclusions of this exercise. In a number of appendices we then give the detailed results of this intercomparison in tabular (Appendix I) and graphical form (Appendix II). Finally, the specifications of this intercomparison are given (Appendix IV) along with questionnaires returned by many of the participants (Appendix III) summarizing the models used in the various codes and giving references to more detailed reports or to manuals.

The motivation for this exercise came from data needs of accelerator-based waste transmutation and energy amplification. The same capabilities of nuclear modeling and radiation transport calculation are valuable in many other areas of science and technology. They relate to dosage calculation in radiation oncology, personnel dosimetry for space stations and a possible new generation of high altitude supersonic aircraft, and stability of microelectronics for satellite communications and commercial avionics. In basic research they are valuable for simulation needed in detector design. There is a further wide range of applications of medium energy nuclear reactions connected with the interactions of cosmic ray particles with matter. Cosmogenic nuclides are widely used as natural radioactive tracers. To understand their production in terrestrial and extraterrestrial matter has to rely on models of nuclide production at medium energies. We think that this exercise will be valuable for many of these needs, in addition to fulfilling the primary motivation of accelerator-driven waste transmutation and energy amplification data needs.

2. Summary of Contributions and Types of Codes

Historically, reactions at intermediate energies have been treated with the intranuclear cascade model [SE47, GO48, BE69], at the early stages of the reaction, followed by an evaporation calculation [WE40, DO59] for nucleons falling below some arbitrary level of excitation (typically of the order of 7-10 MeV above the binding energy or coulomb plus binding energy). The physics is based on the impulse approximation of Serber [SE47], with consideration of Fermi motion mediating collisions according to the work of Goldberger [GO48]. The evaporation phase was often modeled after the early works of Dostrovsky et al. [DO59], using classical sharp-cutoff approximations for charged particle barrier penetrabilities, and simplified approximations for the nuclear level densities, all applied in the Weisskopf evaporation model [WE40].

Many of the approximations made in these codes rest with the fact that their development was contemporary with the evolution of digital computers. As computational abilities improved, some of the codes were revised to take advantage of improved speed and core size, although this was not always the case.

In the early 1970s, semi-classical precompound models [BL75] which treated the intranuclear cascade in a closed form evolved, which easily allowed the direct cascade contributions to be followed until the system was fully equilibrated. In the master-equation formulation [CL71a], this approach may even be used to treat the equilibrium decay channels. Precompound decay models were incorporated into some cascade plus evaporation codes to fill the gap between the arbitrary cutoff of the INC and the equilibrium evaporation decay [GU83]. This produced a marked improvement in the reaction models. Angular distributions in precompound decay are characteristically put in using nucleon-nucleon scattering for an incident nucleon colliding with nucleons having a Fermi momentum distribution [ZI82], or more simply by using expressions which have been fitted to experimental results for purposes of interpolation - so called systematics [KA88].

Precompound decay models effect great simplifications over INC calculations due to the use of statistical few quasi particle distribution functions [ER60, GR66, WI70, WI71] (or exciton state densities) with an equal a-priori population assumption in place of explicit calculation of energy partition following two body scattering. It has been shown that, for the most important leading term in the series, this result is consistent with the kinematics of nucleon-nucleon scattering [BL83] at energies below the pion threshold (approximately 270 MeV). Because pion production is not part of the exciton distribution functions, it is at best questionable to employ this approach at incident nucleon energies in

excess of the pion threshold. This limitation has indeed been observed for the submissions in this work. The INC codes, in contrast, have single and double pion production channels (with some approximations) as well as additional particle production channels as appropriate for the energies involved, and may be valid for incident energies of many GeV or tens of GeV.

More recently quantal approaches have been suggested for treating direct continuum reactions in the region below the pion threshold [FE80, TA82, NI88]. There are still some constraints on these approaches; they represent work in progress, and a hope for improved understanding and success in the future. In many regimes they are already starting to become a useful tool in our ability to model nuclear reaction data. Several examples of this type of approach are a part of this intercomparison. Additionally, results have been submitted using a "quantum molecular dynamics" code, which is like the INC for N-N collisions, but with nonlinear nucleon trajectories between collisions as each nucleon is permitted to interact with every other nucleon via a two body force [AI88]. This also represents work in progress; practical use of the QMD models may await massively parallel computing.

In Table 1 we summarize the codes which have been used in this intercomparison, give an alpha-numerical reference to each contribution distinguishing different contributions from the same participant, and list the models used in each. The contribution reference code used is of the form *ABmn* having the following meaning: *AB* is an abbreviation of the (first) contributor's name, *m* and *n* are running numbers, *m* counting the number of models or codes used by a particular participant and *n* counting different options used for the same code indicated by *Abm*. This reference code is used both in tables and figures making sure that always the same plot symbol is used for a particular contribution reference code. Note that several different laboratories have used some of the same models and codes, sometimes exercising different options or modifications. The names and laboratory affiliations of participants are given in Table 2.

We represent intranuclear cascade approaches as INC, precompound exciton model approaches as PE, compound nucleus evaporation models as EVAP, and use FKK for the only quantal approach used in this exercise, that due to Feshbach, Kerman and Koonin [FE80]. The evaporation routines are all forms of the Weisskopf-Ewing [WE40] formulation, except for the FKK-GNASH code, for which a Hauser-Feshbach approach (specific coupling of angular momenta) may be used, and the master equation model from the Bratislava group.

Many of the INC codes are taken from the HETC transport code of Armstrong and Chandler [AR72], which incorporated the INC code of Bertini [BE69, BE63]. The HETC/KFA2, HETC/Bruyere, HETC-FRG and HETC-3STEP codes are modifications of the HETC code. In addition to the Bertini INC code, there are also contributions using the ISABEL code of Yariv and Fraenkel [YA81], derived from the Vegas code of Chen et al. [CH68], one of them coupling the ISABEL code to the SMM code [BO92b] in order to account for nuclide production by fragmentation reactions. Many additional options and physics have been incorporated into the HETC/KFA2, HETC/Bruyere, HETC-FRG and HETC-3STEP versions of HETC [AR84, CL88, BE96, NA86, IS93]. For further details of the physics used in each set of results presented, we refer to the questionnaires contained in Appendix II, and to the references cited therein.

The codes in Table 1 can be categorized from the practical point of view according to the energy regions for which they are applicable. It is convenient to distinguishing coarsely three energy regions which are dominated by nuclear reactions involving a compound nucleus in statistical equilibrium (0 - 50 MeV), by preequilibrium reactions (50 - 200 MeV) and by an initial intranuclear cascade (above 200 MeV). It is quite natural that preequilibrium codes such as ALICE 92 [BL91] (*BL11, BL12,*

BL13), ALICE-IPPE [BL82] (*SH11*), FKK-GNASH [CH93] (*CM11*,*CM12, CM13*), MINGUS [KO94] (*KO11*) and PEQAG2 [BE89] (*BE11*) are confined to energies below 200 MeV, the actual upper limits in the contributions depending a little on the degree of confidence of the different contributors.

There are, however, two contributions in which the pre-equilibrium approach is extended to higher energies. The AREL code [BL94b] (*GL11, GL12*) is a modification of the hybrid model of preequilibrium reactions [BL71, BL72] which formally can be run up to energies of 900 MeV. It shows some improvements compared to earlier attempts of such an extension (code ALICE 900 [BL90]) by taking into account relativistic kinematics, but being still limited by the neglect of multiple preequilibrium emission of particles [SC96]. This problem was overcome by using Monte Carlo techniques to describe multiple preequilibrium decay. The contributions (*BL21, BL22*) gives results for this approach using the code HMS-ALICE [BL96a] for energies up to 290 MeV.

Among the contributions calculated by PE-EVAP-type codes there are some which tested different sets of parameters or options which can be chosen in the respective codes. This may be due to which set of level density parameters are considered best for particular target elements, which nuclear mass formulas or mass data are used or what level of complexity has been adopted for the calculations.

The first of these reasons applies to the contributions (*BL11, BL12, BL13* and *BL21, BL23*). For the ALICE 92 calculations Fermi gas level densities were used for all target elements (*BL11*). In addition, calculations were done for the target element oxygen using Chadwick level density parameters [CH96] (*BL12*) and for the target element cobalt with Kataria Ramamurthy level densities [KA90] (*BL13*). For the same reason the HMS-ALICE calculations were done for the target element gold with Fermi gas level densities (*BL21*) and with Kataria-Ramamurthy level densities for cobalt (*BL23*).

The second reason applies to the choice of calculational options for the AREL calculations (*GL11, GL12*). As investigated in detail elsewhere [BO93, SC96] the choice of mass options and the neglect or consideration of shell effects in the mass formulas has partially strong influences on the results of the calculations. The two options used here are the extremes, namely Myers and Swiatecki (MS) mass formula [MY66] neglecting shell corrections and pairing effects (*GL11*), on the one hand, and experimental nuclear masses according to Wapstra and Audi [WA85] as far as available, else MS masses with both shell and pairing corrections (*GL12*).

The third reason applies for the FKK-GNASH calculations (*CM11, CM12, CM13*). For the target element oxygen the contributors took into account emission of neutrons, protons, H-3, He-3 and Be-7 (*CM13*), for the target element aluminum no evaporation of Be-7 was accounted for (*CM11*), and for the target element iron emission of complex particles such as H-3, He-3 and Be-7 was neglected (*CM12*).

The restriction of PE-EVAP-type codes to energies below 200 MeV causes a problem in the calculations of activation yields for gold, because medium-energy fission is not included in the respective models though it could be treated within the frameworks of the respective models. In case of the ALICE-type codes a considerable recording would be necessary in addition since there is presently a restriction in the array sizes resulting in maximum numbers of emitted protons and neutrons.

All other codes which give results for energies significantly above 200 MeV include INC models or a QMD model and make use of Monte Carlo techniques. Among the 15 contributions, only one the code (*CS11*) makes use of a quantal approach for the intranuclear cascade in form of the

QMDRELP+SDMRELP code [NI95]. All others originate from HETC- or VEGAS-like approaches. These latter codes can be categorized depending on whether they include preequilibrium phases between the intranuclear cascade and the evaporation stages or not. Six contributions make use of INC-PE-EVAP codes, namely CEM95 [MA96, MA93] (*MA11*), HETC-FRG (*IS11*), HETC-3STEP [NA86, IS93] (*TA11*), INUCL [ST90, SH93] (*KA11*), MSDM [AD93, AM89, AM90, BO87, BO90, GU75, GU83, TO83] (*SO11*), and PACE/MSM [ER94] (*FO11*), while nine contributions use codes which follow the classical INC-EVAP scheme. These latter are CASCADE [BA85] (*SH21*), DISCA [KO96] (*SH31*), HETC/BRUYERE [BE96] (*FL11*), HETC/KFA2 [AR84, CL88] (*MI11*), ISABEL-EVA [YA81] (*FR11, FR12*), ISABEL/SMM [YA81, BO92b] (*LA11*) and MECC7+EVAP_F [BE69, DR62, AT80] (*YO11*).

A further distinction of the versatility of codes applicable to energies above 200 MeV is whether or not they take into account Fermi break-up and allow for calculation of fragmentation products. In this intercomparison there are three contributions which do so. The MSDM code (see refs. in App. 3) (*SO11*) considers Fermi break-up. In the contribution *LA11* the MSM code by Botvina and Mishustin [BO92b] was coupled to the ISABEL code [YA81] to improve the calculations of fragmentation products. HETC-FRG (*IS11*) considers fragmentation reactions by using a liquid-gas phase transition model.

While modeling of Fermi break-up is a necessary addition for many codes, all but the CEM95 [MA96,MA93] (*MA11*) and DISCA codes [KO96] (*SH31*) take into account medium-energy fission.

Among the contributions using Monte Carlo techniques to describe the intranuclear cascade different calculational options for the same code were only used in case of the ISABEL-EVA code [YA81] where to contributions for the target elements aluminum to gold were given, one using a local (*FR11*) and one a uniform (*FR12*) Thomas Fermi density approximation for momenta.

The INC codes and, in particular, the QMD code need large amounts of computation time per event. This causes partially problems with when calculating production cross sections which are small compared to the reaction cross section. Therefore, rare reaction channels show great statistical uncertainties in the cross sections due to the statistical limitations of running small numbers of events. Some INC contributions even show a general lack of statistical accuracy. Here, this intercomparison clearly demonstrates that excessive use of computing time is indispensable when running INC codes to calculate activation yields. This is particularly true and has another quality than for INC codes for the new QMD approach which, however, may considerably improve as massively parallel computer technology becomes increasingly available.

There is one contribution (*MI21*) which is basically different from all others since it makes us of a semi-empirical cross section formula. Besides models employing the wide range of nuclear reaction theories, semi-empirical models of medium energy nuclear reactions and semi-empirical cross section formulas [RU66, SI73,SU91,WE91] have a long tradition in attempts to satisfy the cross section data needs of various fields of applications. In this intercomparison, one contribution (*MI21*) applies the semi-empirical model by Silberberg and Tsao [SI73] in a modified and updated form. The particular appeal of the semi-empirical models is that a minimum of computing time allows for estimates of production cross section which, however, often are applied without precautions or with wrong presumptions about the achievable accuracy. Though semi-empirical model do not necessarily reflect the status of our basic understanding of nuclear reaction modes, it was felt to be important to include such a model into this intercomparison in order to evaluate the capabilities of such frequently applied models.

In this "part 3" code intercomparison we test the nuclear physics of the codes by comparing calculated activation yields with experimental results. Only in case of the reaction cross sections and of some important residual nuclides for which no experimental data exist we restrict ourselves to the mere comparison of the calculational results. Given the importance of experimental activation yields for this intercomparison we give a detailed survey on the sources of experimental data used in the next chapter. Since, moreover, we try to avoid a subjective view when comparing experimental and calculated data we devote chapter 4 to the detailed description how the contributed calculated „zero time“ cross sections were consistently transformed to be comparable to measured cross sections and how quantitative methods are used to make this comparison a mathematically well-founded objective one.

3. Sources of experimental data

Though a large number of experimental investigations has been performed during the last four decades, see e.g. [TO71, MC76, BU80, BU81, HO82, HO85, KE73, IL91] and the EXFOR compilations at the international nuclear data banks] for references, the experimental data base of integral cross sections for the production of residual nuclides by proton-induced reactions is neither comprehensive nor reliable. It is widely contradictory and except for a few reactions [TO71] there do not exist evaluated data. For many of the reactions which were somewhat more intensely investigated a scatter of data of up to an order of magnitude is observed; see e.g. [MI95, SC96] for a detailed discussion. For a comparison with theoretical calculations as in this exercise the consistency of the experimental data set to which the model and code predictions should be compared is essential.

During the last two decades such a consistent data set was established for proton-induced reactions on target elements C, N, O, Mg, Al, Si, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Sr, Y, Zr, Nb, Ba and Au for energies up to 2.6 GeV [MI96a]. It was mainly aimed to satisfy the data needs of model calculations of cosmic ray interactions with extraterrestrial matter [MI96b]. Some target elements without cosmophysical relevance such as V, Co, Nb, and Au were included into these studies for systematic reasons to allow for some comparison with theories of nuclear reactions. This data base covers today 547 different target/product combinations and a total of more than 15,000 cross sections. An extension of this work contributing to waste transmutation and energy amplification studies is presently underway. In experiments at Laboratoire National Saturne/Saclay and the Svedberg Laboratory/University of Uppsala residual nuclide production by proton-induced reaction from target elements Na, Cr, Rb, Mo, Rh, Ag, Te, I, Cs, La, Ta, W, Re, Os, Ir, Hg, Pb, Bi, U and Th is investigated; see [GL96a, GL96b, BL96b] for first results. However, it will take some time until all the results from these ongoing experiments are available and therefore they could not be used for this exercise.

Therefore, it was decided to base this intercomparison on the data available in 1995 and to concentrate on the target elements oxygen, aluminum, iron, cobalt, zirconium and gold. Of all the experimental data used for the intercomparison only a minor part was published before the specifications of this intercomparison, [MI95] and references therein. In particular, data for target elements ($Z < 29$) for energies between 200 MeV and 400 MeV [SC96] were not yet published at that time and those for Zr and Au were not published at all.

In parallel to this intercomparison a report on the complete data base is prepared [MI96a]. All the new data of [MI96a] will be transformed to EXFOR format (*EXFOR number O0276*) and will be made available by the NEA Data Bank.

For this intercomparison it was decided that at first hand only data from this data base are used because of the consistency of this data set. Table 3 gives the sources of experimental data of all reactions used in this intercomparison. Cross sections from the data set described above are referenced to as „MI96". Under this label all data published by our collaboration [BO93] (*EXFOR number O0282)*, [DI90a] (*EXFOR number O0098)*, [DI90b] (*EXFOR number O0281)*, [MI78a] (*EXFOR number B0100)*, [MI78b] (*EXFOR number B0083)*, [MI79a] (*EXFOR number A0146)*, [MI79b] (*EXFOR number A0151)*, [MI80] (*EXFOR number A0145)*, [MI84a] (*EXFOR number A0100)*, [MI84b] (*EXFOR number A0100)*, [MI85] (*EXFOR number A0100)*, [Mi86](*EXFOR number A0344)*, [MI89a] (*EXFOR number O0078)*, [MI89b] (*EXFOR number O0280)*, [MI95] (*EXFOR number O0277)*, [SC96] (*EXFOR number O0284)*, [WE75] (*EXFOR number O0088)* as well as the unpublished ones [MI96a] (*EXFOR number O0276)* are summarized. From the published work cited above only two references were omitted in this intercomparison. The data published in a paper by Dittrich et al. [DI90a] (*EXFOR number O0281)* were not corrected for interference by secondaries and superseded by a later work [MI95] (*EXFOR number O0277)*. Further, the cross sections reported by Weigel et al. [WE75] (*EXFOR number O0088)* for the target element iron were omitted for reasons discussed elsewhere [MI95] (*EXFOR number O0277)*.

There are, however, some reactions which, on the one hand, are not or just incompletely covered by this data base. But, on the other hand, they were considered to be important for this intercomparison. For these reactions some selected work of other authors was added to the experimental data set. Table 3 gives detailed references to them. The different sources of experimental cross sections are not distinguished in the figures for better readability. But it must be kept in mind that for the respective reactions less internal consistency has to be anticipated.

For the target elements from oxygen to zirconium this was done for particular reactions only: ^{11}C and ^{14}C from oxygen, ^{3}H, ^{3}He, ^{4}He, ^{20}Ne, ^{21}Ne and ^{22}Ne from aluminum and iron, ^{24}Na, ^{28}Mg, ^{36}Cl, ^{36}Ar, ^{38}Ar and ^{55}Fe from iron, ^{56}Ni from cobalt, and ^{22}Na and stable Kr-isotopes from zirconium. For the target element gold it was more frequently necessary to add data from other authors. Some rejections were made in a quite subjective way if the data from a particular publication showed extreme deviations from the work of most other authors. Table 3 lists exclusively the references used in this work and is not meant as a compilation of all existing references.

4. Methodology of the intercomparison

This intercomparison is based on comparing the calculated cross sections with high quality experimental data. Only in some exceptional cases the results of different models and codes were compared with each other if no experimental data were available. This was done for products such as hydrogen and helium isotopes because of the importance of such data with respect to material damage, on the one hand, and because of the strongly differing results of some codes on the other hand. It is the purpose of this intercomparison to provide a basis for the model and code developers to become aware of still existing shortcomings and to be able to recognize particular products where the calculations fail because of not or wrongly considering particular reaction modes. To this end a comprehensive graphical presentation of all the results is necessary which, on the one hand, shows the general reliability or the failure of models and codes and which, on the other hand, enables the reader to look for all the individual reactions in detail.

Further, by asking for calculation of as many as possible target/product combinations and an as complete as possible coverage of the energy region from zero to 5000 MeV the ranges of applicability and the inherent restrictions of models and codes become evident.

In order to make the results more easily understandable for non-specialists we also tried to find a mathematical formulation of the degree of agreement or disagreement between theories and experiment. The procedures developed for this allow to reduce the large amount of results to a set of three or four tables or even to one figure. The so compressed results are for a quick glance and allow some judgment about different models and codes, but it has to be stressed that an improvement and qualified judgment is only possible by looking at the individual results for each reaction. Only by that the real shortcomings and some reasons for failure can be recognized which is a necessary prerequisite for improvements of models and codes.

4.1 Calculation of cumulative cross sections

Calculated activation yields were reported by the participants as „zero time“ or independent cross sections not taking into account decay of short-lived progenitors. Among the nuclides used in this intercomparison, however, the shortest half-life is 10 min (^{11}C). Therefore, cumulative cross sections had to be calculated from the „zero time“ results delivered by the participants. Table 4 gives a survey on the radioactive progenitors considered and the nuclear data used to calculate the cumulative cross sections.

It is to emphasize that in particular for a heavy target element such as gold the cumulative cross sections can differ from the independent ones by an order of magnitude for special products. For other product nuclides and for light target elements this effect may be smaller and sometimes it is even negligible. In order to add no ambiguity to the interpretation of the results of this intercomparison due to possibly differing calculations of cumulative cross sections by particular contributors and to avoid one source of misinterpretation of the specifications the cumulative cross sections all were calculated from independent ones as described below.

Let there be a decay chain

$$nucl_1 \rightarrow nucl_2 \rightarrow nucl_3 \rightarrow ... \rightarrow nucl_n$$

of n radionuclides ($nucl_i$, i = 1,...,n) with decay constants λ_i and branching probability r_i when decaying from $nucl_i$ to $nucl_{i+1}$. σ_i are the independent or „zero time“ cross sections for the production of nuclide i from a target nucleus or element. Then the cumulative cross section $\sigma_{n,cum}$ for the production of the nth nuclide is calculated as

equ. 1: $$\sigma_{n,cum} = \sigma_n + (\lambda_{n-1} / (\lambda_{n-1} - \lambda_n)) * \Sigma_i \sigma_i * r_i$$

This approximation requires

equ. 2: $$\lambda_n / (\lambda_i - \lambda_n) << 1 \text{ for all } i > 1.$$

The condition given in equ. 2 is checked with a limit of 0.05 which is equivalent to a contribution less than 5 %.

If the half-lives of *all* radioactive progenitors are very short compared to that of the nuclide in question then all radioactive progenitors are completely decayed before measurement of an irradiated target. In

this case $\lambda_{n-1} / (\lambda_{n-1} - \lambda_n)$ is unity and the cumulative cross sections is the sum of all independent cross sections of nuclides decaying to the product times their respective branching probabilities plus the direct cross section of this nuclide.

The assumption of very short-lived progenitors holds in many cases, but not in all. There are a few cases (e.g. ^{86}Zr and ^{86}Y, ^{88}Zr and ^{88}Y, ^{95}Zr and ^{95g}Nb) where a progenitor (n-1) decays to the product nuclide (n) with a decay constant λ_{n-1} comparable to λ_n. In this case, equ. 1 gives a better approximation of the experimental reality.

The cumulative cross sections were calculated consistently from the delivered contributions and then compared with the experimental data. Since this procedure was announced in the specifications of the intercomparison, it is not discussed here in detail whether a particular contribution does consider all precursors necessary for such a calculation.

There may be some codes which according to dimensional limitations in the tables of residual nuclides are not capable to calculate each necessary precursor. But this problem will not show up frequently at lower energies (< 200 MeV). For Monte Carlo codes, it does not pose a problem if the histories are carefully analyzed.

Remark: Because of the isobaric yields being nearly Gaussian shaped with small half-value widths at intermediate energies, such a calculation is possible in most cases without particular problems. Only in the case of heavy target elements such as gold problems can occur. Due to the prevalence of neutron emission in the evaporation phase highly neutron deficient radionuclides far off from the valley of stability have to be considered as progenitors. The cumulative production for heavy target elements can be larger by an order of magnitude than the direct one. This may cause problems in codes which have fixed size tables of possible product nuclides being even limited in the numbers of emitted protons or neutrons. This particular problem can already show up at relatively low energies. However, for fission products from gold or for intermediate mass fragments this is not important. Nuclides very close to the target should also not be too much affected.

4.2 Graphical presentation of results

Generally, the intercomparison is based on the comparison of calculated and experimental cross sections. Therefore, figures in which such comparisons are performed make up the majority of plots. Only for a few reactions for which no experimental data exist, plots are included into this report and which only the calculated results of the different contributions are compared. This is commonly the case for the production of light nuclei and nucleons for which integral data are rare; e.g. ^{1}H, ^{2}H, ^{3}H, ^{3}He and ^{4}He. Because of the particular importance of the production of theses nuclei in view of material damage and because partially severe differences between the different contributions they shall also be exemplified here.

Since the various codes employed have different energy ranges of application a uniform type of graphical presentation of the intercomparison is not justified. Therefore, the results are presented in two different types of plots for

- 0 MeV to 200 MeV on a linear energy scale
- 1 MeV to 10 GeV on a logarithmic energy scale.

The numbers of figures covering energies from 0 to 200 MeV, only, was restricted by excluding all plots with less than 20 experimental plus theoretical data. Always logarithmic scales are used for the

cross sections, but not more than four decades were allowed for. Uncertainties of the cross sections which were given by the participants were plotted only if they exceeded the symbol sizes.

All calculated data are shown in the figures except for a very small number which was omitted for different reasons. These omitted cross sections were

- data which did not fit into the maximum of four decades which was allowed for when plotting the results. These were very few extreme outlyers as results of Monte Carlo calculations with bad statistics
- data with relative uncertainties exceeding 70 % were arbitrarily removed from the data set. Such data relied on one or two events found in the Monte Carlo histories.

4.3 Quantification of agreement between experiment and theory

In order to quantify somehow the quality of a contribution in comparison with the experimental cross sections, mathematical measures were searched. It was found that the agreement between experiment and theory can be described by deviation factors which are calculated for each reaction point-wise at each energy for which an experimental cross section exists. These point-wise deviation factors can then be averaged over certain energy ranges and also over all or a part of the different reactions.

For a given reaction (target/product combination) we have ($\sigma_{exp,i}$, i = 1,...,n_{exp}) experimental cross sections at energies (E_i, i = 1, ..., n_{exp}). Then we define a *mean square logarithmic deviation* by

equ. 3: $$<(\log\sigma_{exp} - \log\sigma_{theo})^2> = \sum_i (\log\sigma_{exp,i} - \log\sigma_{theo,i})^2 / NS$$

The theoretical cross sections $\sigma_{theo,i}$ at the energies E_i were obtained from the calculated cross sections by double-logarithmic interpolation. No extrapolations were made. NS is the number of energy points with experimental cross sections for which this procedure is possible in a given energy interval.

Then the average deviation factor <F> is defined by

equ: 4: $$<F> = 10.^{**} SQRT(<(\log\sigma_{exp} - \log\sigma_{theo})^2>)$$

Logarithmic deviation factors have the advantage of being illustrative. They were chosen instead of linear ones since the variations observed for a given experimental cross section are often large compared to the cross section value. Consequently, a linear normal distribution of deviations between theories and experiment is unlikely. A log-normal distribution would then be the simplest assumption. For such a distribution <F> represents the standard deviation transformed back to a linear scale.

The uncertainties of the experimental cross sections have been neglected in the calculations for two reasons. Firstly, they are usually much smaller than the deviation between theories and experiments. Secondly, They are affecting all contributions is the same way.

Since the average deviation factor does not allow to distinguish underestimates from overestimates, we define in addition the maximum and minimum deviation factors, F_{max} and F_{min}, respectively by:

equ. 5: $$F_{max} = \max (\sigma_{exp,i} / \sigma_{theo,i} , i = 1, ..., NS)$$

equ. 6: $F_{min} = \min (\sigma_{exp,i} / \sigma_{theo,i}$, i = 1, ..., NS)

It is not meaningful to average such deviation factors over the entire energy range since the energy coverage of the different models and codes differs too much. Therefore, it was necessary to distinguish three energy regions, namely 0 MeV - 50 MeV, 51 MeV - 200 MeV and 201 MeV - 5000 MeV. This was done in order to coarsely distinguish energy ranges dominated by reactions involving a compound nucleus in statistical equilibrium, a precompound dominated preequilibrium region and a range in which intranuclear cascades dominate the initial phase of a nuclear reaction. For all reactions we calculated <F>, F_{max} and F_{min} for each of the three energy regions independently. In tables 6 - 8 for each reaction the values of the <F>, F_{max} and F_{min} are given for the energy regions up to 50 MeV, from 51 MeV to 200 MeV and above 200 MeV, respectively. This allows for a detailed judgment on the basis of individual reactions.

It is to note that for a given contribution and a given reaction the number NS of pointwise deviation factors may deviate from n_{exp} because of the different energy coverage of the contributions. Therefore, it is meaningful to define the number of cross sections of a given reaction i for which such a comparison was made NS_i. Since, moreover, the coverage of reactions is also differing from contribution to contribution one also has to know the number NR of reactions for which a comparison was possible.

In order to obtain also some global judgment about the quality of a given contribution we can now define a *global mean deviation factor* <<F>> by averaging for each contribution in addition over all reactions j:

equ. 7: $<<(\log\sigma_{exp} - \log\sigma_{theo})^2>> = \sum_{i,j} (\log\sigma_{exp,i} - \log\sigma_{theo,i})^2 / \sum_j NS_j$

equ: 8: $<<F>> = 10.^{**} SQRT(<<(\log\sigma_{exp} - \log\sigma_{theo})^2>>)$

For all three energy ranges these calculations were also performed. The results are shown in table 9 which in addition to the <<F>> values give the total $NS = \Sigma_j NS_j$ and NR values for each of the three energy regions. Thus a numerical result is derived which measures the global capabilities of a contribution in one number, <<F>>, for each energy region.

5. Results and Discussion

This model and code exercise provides an in-depth survey on the capabilities of *a priori* calculations of activation yields for target elements O, Al, Fe, Co, Zr and Au from thresholds up to 5 GeV. The comparison of calculational results with high-quality experimental data for more than 200 target/product combinations allows in an unprecedented way to analyze the advantages and shortcomings of nuclear models and codes. It provides a tool to recognize the weak points in models and codes and thereby it can serve as a basis for future improvements.

Surveying the results of this intercomparison and the different types of problems encountered in it one has to mention:

- Most contributions tried to cover as many requested target element/product combinations as possible. Some codes could only cover all requested reactions due to limitations in array sizes, some models have restrictions with respect to the range of applicable energies, and some contributors

hesitated to give results for one or some target elements. In spite of that the coverage of reactions and energies for a given target element allows for a good survey on the applicability of the models and codes in general.

- There was no contribution giving results for the production of isomeric states.

- There are just few contributions giving results for light complex particles.

- For many product nuclides the deviations between the different contributions fill a range of about two orders of magnitude and many calculated cross sections are widely contradictory to the experimental data.

- There are reactions for which the range of calculations is just a factor of two covering nicely the experimental data.

- Though there are just few experimental data for the production of light complex particles (H-3, He-3, He-4) to compare with, the discrepancies between the different contributions are striking. The calculated excitation functions partially differ by up to two orders of magnitude, e.g. for Fe, and the agreement with the rare experimental data often is poor.

- There are deviations among the different calculated reaction cross sections which are particularly important in the low-energy region. However, the differences in the reaction cross sections cannot account for the differences seen in the individual excitation functions for the production of residual nuclides.

- There are some contributions where the calculations suffer from poor statistics. But, the wide range of calculational results in general is not dominated by statistical problems. It seems to be more likely that there are real differences in the understanding of the individual reactions by the different models and codes. The deviations show no systematics when globally comparing the reactions. There is no model or code which from the underlying physics is evidently wrong.

- There are problems with nuclear masses and the calculations of binding energies and consequently of reaction thresholds. In particular for reactions for which just apparent threshold can be given because no clear cut reaction paths are defined due to large numbers of emitted particles. Also the fact that some INC codes do not conserve energy, having always the same neutron and proton binding energies, can be a source of problems when calculating thresholds. Finally, the neglect of cluster channels can add to such failures, since the cluster binding energy is ‘lost’ if one makes a product by nucleon or nucleon plus alpha channels, only.

- Calculations for the target element oxygen were not given by many contributors, e.g. table 1. As a matter of fact one has to accept that quite a number of the statistical assumptions underlying preequilibrium and equilibrium reactions are not valid for systems with such small numbers of nucleons. Given, however, the importance of elements such as carbon, nitrogen and oxygen for calculations in radiation protection and dosimetry and for the activation of shields and ambient air of medium energy accelerators, also for the light target elements the calculational methods have to be available.

- There are extreme problems when calculating products near to or at double magic configurations such as ^{56}Ni and ^{57}Ni from cobalt. This points to real problems with accounting for shell effects and choosing level density formulas.

- There is clear evidence that preequilibrium emission of light complex particles, in particular He-4, has to be taken into account. Otherwise the structures in the excitation functions of reactions in the course one or two He-4 particles might be emitted cannot be adequately described.

- Particular problems are encountered when looking for the nuclide production by fission from gold, some nuclides being systematically over- and some underestimated.

- There are fission products which show strongly different shapes of the excitation functions, with respect to both the apparent thresholds and the energy dependence above 1 GeV. These differences point to these nuclides being produced either by fission of an excited nucleus with a mass close to the target nucleus, on the one hand, or by that of an nucleus resulting from a long intranuclear cascade with a large difference in mass between fissioning and target nuclide on the other. The first way of production results in relatively low apparent thresholds below 100 MeV, the second one exhibits thresholds significantly larger than 100 MeV. The fission models used in the different codes mostly do not adequately describe this phenomenon.

- There are strong discrepancies in the calculated excitation functions near the thresholds, in particular for the heavy target elements. This may be partially due to the use of simple mass formulas, but also can be caused by a neglect of the competition between gamma-emission and particle-emission in the deexcitation of the nuclides in the final stages of the reactions.

- There are some plots for the energy region below 200 MeV in which there are no experimental data. These have been included to demonstrate the partially extremely large differences among the calculations in the low-energy part.

- From this intercomparison only limited information about the influences of different level density formula can be derived. This problem has to be investigated by systematic variations of different level density formulas and parameters for a given code. This is one of the many task remaining for the code developers and evaluators.

- There is just one contribution describing the low-energy production (E < 100 MeV) of Be-7 from e.g. iron (*SH11*). The contribution of evaporation of Be-7 from highly excited equilibrated systems is not accounted for by all other models and codes.

- Calculations of activation yields from heavy target elements such as gold pose a particular problem since the measurable cumulative activation yields partially differ by an order of magnitude from zero time cross sections. Thus, for reliable modeling of medium and long-lived products from heavy target elements the suite of possible progenitors has to be carefully evaluated and covered by the model calculations to allow for reliable calculation of cumulative yields.

- An adequate description of Fermi break up and of residual nuclide production by fragmentation is urgently needed. The present intercomparison demonstrates that the up-to-now efforts are still not satisfying. Taking into account Fermi break-up removes the „orders of magnitude“ discrepancies when calculating the production of light fragmentation products. However, the differences between

experiment and theory and between the different calculational approaches are still in the range of those seen for other reaction modes.

This general survey makes evident that there is presently no model or code available which reliably predicts activation yields for all possible target-product combinations. Therefore, for the time being, calculation of the production of residual nuclides has to rely on experimental cross sections. In thick or extended targets a reliable modeling only is possible by combining experimental cross sections of the underlying nuclear reactions with energy dependent flux densities of primary and secondary particles. As inclusive data, the latter can be more reliably calculated than activation yields. In addition, computing power still is not sufficient to handle particle transport and activation in thick or extended targets.

There is a *caveat* with respect to use this intercomparison as a basis of comparative judgment about the different models and codes. The different models used in this intercomparison are not necessarily comparable. There are differences in applicability from the physics used with respect to energy, target and product ranges. Since there does not exist a comprehensive model of medium-energy nucleon-induced reactions, all models and codes are in one way or the other incomplete with respect to the coverage of nuclear reaction phenomena. Here this intercomparison may help to distinguish which phenomena have to be included if a global code system covering all aspects of the relevant nuclear reactions.

Moreover, such a comparative judgment would be erroneous since the contributions are biased by the personal presumptions of the contributors about applicability of the models and codes used. The different contributions cover different energy, target and product ranges reflecting the different capabilities of models and codes but also the estimate of this capabilities by the contributors some being more cautious than others. A quick survey on the coverage of an individual contribution is therefore helpful. For the energy ranges this is given in table 1, for the individual reactions in table 5.

Even with respect to semi-empirical systematics one has to consider that such systematics can only be as good as the experimental data which are used to derive the parameters of the semi-empirical formulas. Due to the fact that a large number of old experimental data is widely contradictory also in case of the systematics an estimate of the general applicability of this approach cannot be obtained from this exercise but rather a means to improve such systematics.

Each of the contributions deserves an in-depth discussion which is impossible within the limited space of this report. Such analyses are left to the contributors themselves hoping that they provide a basis for improvements. However, in order to obtain some comparability of the different contributions in this report and to distinguish some advantages and disadvantages, some quantification of agreement and deviation between experimental and calculated data was searched for.

For such a quantification mean deviation factors were chosen. They can be used globally as well as reaction- or target-element-wise. The global mean deviation factors (Fig. 1, table 9) demonstrate that there is no contribution which is significantly better than predicting activation yields within a factor of two on the gross average. If there are now entries for a contribution in Fig. 1 or table 9, this means that the respective energy range was not covered by it. Apparently small deviation factors can also be the result of a small energy coverage of a contribution, therefore we have indicated also the numbers of reactions and individual cross sections which were used as basis to calculate the global deviation factors into table 9.

The global deviation factors range from a little less than two up to fourteen. It is to emphasize, however, that there is a considerable number of contributions for which the global mean deviation factors are significantly larger than four.

One has, however, to keep in mind that individual, reaction-wise deviation factors can reach even orders of magnitude. Therefore, a detailed judgment can only be made reactionwise. Such data are given in tables 6 - 8 for the three energy regions from 0 MeV to 50 MeV, from 51 MeV to 200 MeV and from 201 MeV to 5 GeV, respectively. Even these reactionwise deviation factors do not exhibit whether the shapes of the excitation functions have been correctly calculated which might indicate that all relevant reaction modes are accounted for properly and that there are just problems of book-keeping. Therefore, one has to look for the maximum and minimum deviation factors which are also given in tables 6 - 8. Small deviations between maximum and minimum deviation factors point to the shape of the excitation functions being correctly reproduced, while large differences indicate that there are problems with the calculated energy dependence of cross sections.

Looking for the deviation factors as function of energies two effects were observed. Extremely large deviation factors may be observed when comparing experimental and calculated cross sections near the thresholds since many codes occasionally wrongly calculate the thresholds or the excitation functions near their thresholds and, at the same time, give cross sections far below the μb-region. This can be due to different reasons, e.g. problems with nuclear masses, optical model parameters, and γ-competition in deexcitation. Furthermore, very large deviations are observed if cross sections are in the nano-barn region.

Since such extreme deviations strongly bias a realistic judgment on the basis of mean deviation factors, the calculation of deviation factors was restricted to theoretical cross sections larger than 1 μb. Since experimental data are available typically down to 10 μb this limit allows to see underestimates up to a factor of ten. The dependence of the mean deviation factors on the value of this limit was carefully tested. A limit of 1 μb does not remove any reaction from the intercomparison and the mean factors do not change significantly between 0.1 μb and 10 μb. An increase of the lower limit up to 1 mb does decrease the global mean deviation factors by up to a factor of two for some contributions. However, the best global mean deviation factors remain to have values of about two.

A global mean deviation factors of two can already be considered as the best what can be presently achieved. These deviation factors have, however, to be distinguished for different energy regions. The causes of deviations between theories and experiment differ for the three energy ranges considered in this intercomparison.

Finally, it has to be emphasized that the quantification of agreement used here for comparison favors our desire for simplicity. There must, however, be a *caveat* that the mere deviation numbers cannot provide the basis for a physically adequate judgment about any model. or code. The causes of the individual deviations are multi-factorial and can - for a given model, code or contribution - only be evaluated by model and parameter exercises for a wide range of reactions. The present intercomparison gives a first survey over the related problems. It should be understood by the modelers and code developers as a starting point for the improvement of models and codes.

6. Conclusion

This exercise has, as a main goal, the display of results of model calculations versus high quality experimental data and offer a tool to the model and code developer to work with in order to improve

their theoretical approaches or code formulations. The comparison given in this report can be regarded only as a first step. Detailed reactionwise discussion and interpretation of the results as well as systematic parameter studies aimed on the evaluation of the reasons for particular discrepancies between calculations and experiments are beyond the scope of this report and will rest with the model and code developers. In spite of that, it was a task of this intercomparison to derive a general survey and to draw *conclusions* about the capabilities of present days nuclear models and codes when calculating activation yields.

Conclusions of such comparisons are subjective in nature. We have tried, however, to give a quantitative judgment on the basis of individual and global average deviation factors between experiment and theory. Such numbers are biased due to the availability of experimental data which do not represent necessarily a meaningful grid of energy points for such a judgment. To achieve some grade of justification we distinguished three energy regions, namely 0 MeV - 50 MeV, 51 MeV - 200 MeV and 201 MeV - 5000 MeV. This was done in order to coarsely distinguish energy ranges dominated by reactions involving a compound nucleus in statistical equilibrium, a precompound dominated preequilibrium region and a range in which intranuclear cascades dominate the initial phase of a nuclear reaction.

From this exercise we may conclude that modeling calculations of intermediate energy activation yields on a predictive basis may at best have uncertainties of the order of a factor of two. Frequently, average deviations are much lager and individual reaction-wise deviations may go up to two or three orders of magnitude. There are no general over- or underestimates by individual models or codes, but rather a broad scatter of calculated data which occasionally among the different contributions are contradictory up to 3 orders of magnitude for a given reaction. It is not possible within the limited size of this report to trace the reasons for these discrepancies in detail. It can just be stated that the causes of the discrepancies are multi-factorial and not merely due to wrong book-keeping. Problems are encountered which are connected with the calculation of nuclear masses, binding energies and consequently Q-values, with the consideration of shell effects and the various level density formulas used, with the neglect of competition between γ- and particle deexcitation of excited intermediate nuclei, and , last but not least, with the basic modeling of medium energy fission and Fermi break-up.

Considering all these observations there is a need for major improvement of models and codes. Such efforts would be well spent given the importance of intermediate energy nuclear data for future technological development. In an ultimate conclusion one can state that calculation of activation yields turns out to be an extremely difficult task which cannot be adequately solved by present days nuclear models and codes. This emphasizes the importance of experimental work for future technological applications, on the one hand, and opens up a broad field of work for theoreticians and model and code developers, on the other.

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